Doctoral Thesis

Magnetic relaxation effects in organic, high-Tc, and classical superconductors flux-creep by thermal activation and quantum tunneling

Author(s):
Giovanni, Juri

Publication Date:
1992

Permanent Link:
https://doi.org/10.3929/ethz-a-000667006

Rights / License:
In Copyright - Non-Commercial Use Permitted
Magnetic Relaxation Effects in Organic, High-\(T_c\), and Classical Superconductors: Flux-Creep by Thermal Activation and Quantum Tunneling

A dissertation submitted to the
SWISS FEDERAL INSTITUTE OF TECHNOLOGY
ZURICH

for the degree of
Doctor of Natural Sciences

presented by
JURI GIOVANNI
Dipl. Phys. ETH
born on the 22\textsuperscript{nd} March 1961
citizen of Quinto (Ticino)

accepted on the recommendation of
Prof. Dr. A.C. Mota, examiner
Prof. Dr. J. L. Olsen, co-examiner
PD Dr. H. Keller, co-examiner

1992
A Pà, Mam e Cuchi
CONTENTS

ABSTRACT ................................................................................................................ iv

KURZFASSUNG ........................................................................................................ vi

I. INTRODUCTION ................................................................................................. 1

II. MODELS OF THE CRITICAL STATE AND FLUX MOTION ........................................ 5
   II.1 Bean's Critical State Model ........................................................................... 6
   II.2 Anderson's Flux-Creep Theory .................................................................... 15
   II.3 Theory of Collective Flux-Creep .................................................................. 18
   II.4 Theory of Quantum Collective Flux-Creep .................................................. 24
   II.5 Vortex-Glass Model ...................................................................................... 30

III. EXPERIMENTAL ARRANGEMENT AND METHODS ..................................................... 35
   III.1 Dilution Refrigerator ................................................................................. 36
   III.2 4K SQUID Magnetometer ........................................................................ 36
         III.2.1 Cryogenic Environment .................................................................... 36
         III.2.2 Coil Arrangement .......................................................................... 37
         III.2.3 Measuring System .......................................................................... 40
   III.3 Experimental Procedures .......................................................................... 41
         III.3.1 Isothermal dc Magnetization Curves ............................................. 42
         III.3.2 Decays of the Remanent Magnetization ........................................ 43
   III.4 Background ............................................................................................... 44
   III.5 Remarks on Chosen Units ......................................................................... 48
   III.6 Experimental Methods .............................................................................. 49
III.6.1 Analysis of Time Relaxation Data ......................... 50
III.6.2 Analysis of Remanent Magnetization Data as
Function of Field or Temperature ......................... 52

IV. MAGNETIC RELAXATION EFFECTS IN
(BEDT-TTF)$_2$Cu(SCN)$_2$ SINGLE CRYSTALS ......... 53

IV.1 Specimens .................................................. 53

IV.2 Magnetic Relaxation Effects of the Remanent
Magnetization at 4.2 K ..................................... 56

IV.2.1 Remanent Magnetization as Function of the
Field $H_i$ ..................................................... 57

IV.2.2 Time Relaxation of the Remanent Magnetization
as Function of the Field $H_i$ ................................... 63

IV.3 Magnetic Relaxation Effects of the Remanent
Magnetization as Function of Temperature
Between 5 mK and 8 K ....................................... 75

IV.3.1 Remanent Magnetization, Lower Critical Fields
and Critical Currents ......................................... 75

IV.3.2 Time Relaxation of the Remanent Magnetization:
Flux-Creep by Thermal Activation and Quantum
Tunneling ....................................................... 88

IV.4 ac Susceptibility Measurements ......................... 102

V. MAGNETIC RELAXATION EFFECTS IN A
Y$_1$Ba$_2$Cu$_4$O$_{8+x}$ SINGLE CRYSTAL AT 4.2 K ....... 106

IV.1 Specimens .................................................. 106

IV.2 Magnetic Relaxation Effects of the Remanent
Magnetization at 4.2 K ..................................... 108

IV.2.1 Remanent Magnetization at 4.2 K as Function of the
Field $H_i$ ..................................................... 108

IV.2.2 Time Relaxation of the Remanent Magnetization at
4.2 K as Function of the Field $H_i$ ....................... 110
VI. RELAXATION RATE OF THE REMANENT MAGNETIZATION OF CLASSICAL, CHEVREL PHASE, ORGANIC, AND HIGH-Tc SUPERCONDUCTORS AT 4.2 K

REFERENCES ............................................. 124

ACKNOWLEDGEMENTS ...................................... 130

CURRICULUM VITAE ...................................... 131

LIST OF PUBLICATIONS .................................... 132
ABSTRACT

A systematic study of magnetic relaxation phenomena in classical (NbTi, PbIn), Chevrel Phase, high-Tc (Y1Ba2Cu3O7-δ, Y1Ba2Cu4O8+x and Sr0.2La1.8CuO4) and organic ((BEDT-TTF)2Cu(SCN)2) superconductors was carried out at 4.2 K. Measurements of the time relaxation of the remanent magnetization show that at this temperature relaxation effects are much stronger in high-Tc and organic superconductors than in classical or Chevrel Phase superconductors. The logarithmic time dependence predicted by the Kim-Anderson theory of flux-creep is verified only for classical superconductors, where relaxation effects are very small. The relaxation curves measured for all the other superconductors exhibit marked deviations from the logarithmic law. All these curves can be well fitted by a power law of the type \( M_{\text{rem}}(t) = M_{\text{rem}}(\infty) + b(t/t_0)^{-\beta} \) within the time window of our experiments (1 s \( \leq t \leq 10^5 \) s, \( t_0 = 1 \) s). The values of \( \beta \) obtained from the fits range between 0.02 and 0.20.

From the initial logarithmic rates \( R = -\partial \ln M_{\text{rem}}(t)/\partial \log t \bigg|_{t=1 \text{s}} \) values of the average pinning energies \( U_c \) at 4.2 K were calculated. For the organic and the high-Tc superconductors the values of \( U_c \) range between 3 and 35 meV, whereas for Chevrel Phase and classical superconductors the values are between 0.15 eV and 0.7 eV.

Isothermal dc magnetization cycles and relaxation curves of \( M_{\text{rem}}(t) \) were measured on (BEDT-TTF)2Cu(SCN)2 single crystals between 5 mK and 8 K for applied fields \( H \) perpendicular and parallel to the superconducting planes (bc-plane) of the specimens. The values of the critical currents \( J_{\text{c}}^p(0) \) and \( J_{\text{c}}^p(0) \) obtained from the remanent magnetization data are about 2000 and 30 A/cm², respectively. At temperatures \( T < 300 \) mK the values of the initial logarithmic slopes \( S = -\partial M_{\text{rem}}(t)/\partial \log t \bigg|_{t=1 \text{s}} \) of the relaxation curves are constant for both field orientations. The corresponding constant values of the normalized decay rates \( R \) are of the order 0.015 and 0.005 for \( H \perp bc \) and \( H \parallel bc \), respectively. These results provide clear evidence of non-thermally activated flux motion. They can be, both qualitatively and quantitatively, explained by the theory of quantum collective creep (QCC) for layered superconductors in the limit of strong dissipation and for \( T \rightarrow 0 \). At higher temperatures the values of \( R \) follow
a $T^2$-dependence for both field orientations, as predicted by the QCC theory for the case of thermally-assisted quantum tunneling. For $H \perp bc$ the temperature dependence of $R$ above about 1 K deviates from the $T^2$-dependence and shows two peaks. This feature can be explained by the Kim-Anderson theory of flux-creep, if one assumes that pinning centres characterized by two different pinning strengths are effective in this material. For these two types of pinning centres we obtain values of the activation energy $U_c$ equal to $3 \pm 1 \text{ meV}$ and $14 \pm 4 \text{ meV}$.

The analysis of the magnetization curves indicates that the organic superconductor $(\text{BEDT-TTF})_2\text{Cu(SCN)}_2$ behaves essentially as a stack of superconducting layers (bc-planes) with extremely weak interlayer coupling in most of the temperature range. In fact, for $H \parallel bc$ the values of $H_{c1}^b$ are smaller than 0.5 Oe at all temperatures between 5 mK and 8 K. For $H \perp bc$, the $H_{c1}^b(T)$-curve exhibits an upward curvature and the values of $H_{c1}^b$ increase strongly below 4 K [$H_{c1}^b(0) \approx 380 \text{ Oe}$]. This temperature dependence can be explained in terms of a gain of condensation energy due to proximity-induced superconductivity between the layers as the temperature is lowered. It turns out that the $H_{c1}^b(T)$-data follow an exponential temperature dependence, that is typical of the breakdown fields in proximity systems. It is noteworthy, that the exponential temperature dependence of the “irreversibility line” observed at low temperatures for the highly anisotropic high-$T_c$ compound Bi-Pb-Sr-Ca-Cu-O has also been explained with this approach.

The relaxation of the remanent magnetization was measured at 4.2 K on a single crystal of Y$_1$Ba$_2$Cu$_4$O$_{8+x}$ as function of the (previously) applied field $H_i$ ($0 \leq H_i \leq 800 \text{ Oe}$) and of the field orientation ($H_i \parallel a$-, b- and c-axis). The values of the relaxation rates $R$ ($\sim 0.05-0.08$) are about the same as in Y$_1$Ba$_2$Cu$_3$O$_7$-8. Unfortunately, due to the limited field range of our experiments the fully critical state was not reached in the field orientation with $H_i \parallel c$, so that a conclusive study of the anisotropy of the relaxation rates was not possible.
Eine systematische Untersuchung magnetischer Relaxationseffekte ist an verschiedenen Supraleitern durchgeführt worden. Die Proben bestehen aus klassischen Supraleitern (NbTi, PbIn), Chevrel-Phase- (PbMo₆S₈), Hochtemperatur- (YBa₂Cu₃O₇₋₈, YBa₂Cu₄O₈₊ₓ und Sr₂La₁₈CuO₄) und organischen [(BEDT-TTF)₂Cu(SCN)₂] Supraleitern. Messungen der Zeitrelaxation der remanenten Magnetisierung M_{rem} bei 4.2 K zeigen, dass magnetische Relaxationseffekte in Hochtemperatur- und organischen Supraleitern viel stärker sind als in Chevrel-Phase- oder klassischen Supraleitern. Die Gültigkeit des logarithmischen Zeitgesetzes (Anderson-Flusskriechen) wurde im Fall der klassischen Supraleitern, die sehr schwache Relaxationseffekte aufweisen, festgestellt. Die an allen anderen Supraleitern gemessenen Relaxationskurven zeigen klare Abweichungen vom logarithmischen Zeitgesetz innerhalb der typischen Zeitspanne unserer Relaxationsexperimente (1 s < t < 10⁵ s). Solche Relaxationskurven konnten mit einem Potenzgesetz der Art M_{rem}(t) = M_{rem}(∞) + b(t/t₀)^{-β} gefittet werden (mit t₀ = 1 s und β zwischen 0.02 und 0.2). Die durchschnittliche Aktivierungsenergie U_c bei 4.2 K wurde aus der anfänglichen logarithmischen Relaxationsrate \( R = -\frac{\partial \ln M_{rem}}{\partial \log t} \big|_{t=1 s} \) der Relaxationskurven berechnet. Die Werte von U_c liegen im Bereich von 3 bis 35 meV für organische und Hochtemperatursupraleiter und zwischen 0.15 und 0.7 eV für Chevrel-Phase- und klassische Supraleiter.

Magnetisierungskurven und Relaxationskurven von M_{rem} bei konstanter Temperatur sind an zwei Einkristallen aus (BEDT-TTF)₂Cu(SCN)₂ im Temperaturbereich von 5 mK bis 8 K durchgeführt worden. Dabei wurde das magnetische Feld parallel bzw. senkrecht zu den stark supraleitenden Ebenen (bc-Ebenen) der Kristalle angelegt. Aus den Werten von M_{rem} im vollständig kritischen Bereich (Bean's critical state model) sind die kritischen Stromdichten \( J_{c}^{bc}(0) \) und \( J_{c}^{s}(0) \) abgeschätzt worden. Für die zwei anisotropen Komponenten von J_c erhalten wir 2000 bzw. 30 A/cm². Die normierte Relaxationsrate R ist bei tiefer Temperatur für beide Feldrichtungen konstant (R ~ 0.01). Dieses Resultat zeigt, dass ein nicht thermisch aktiverter Relaxationseffekt bei tiefer Temperatur wirksam ist. Die in diesem Gebiet erhaltenen Relaxationsraten sowie ihre Abhängigkeit von der Feldrichtung können im Rahmen der Theorie des Quantum Collective Creep (QCC) für geschichtete Supraleiter im Grenzfall \( T \rightarrow 0 \)
erklärt werden. Unsere experimentellen Daten zeigen bei höheren Temperaturen eine $T^2$-Abhängigkeit, die ebenfalls im Rahmen der QCC-Theorie verstanden werden kann. Die Relaxationsrate $R$ für $H \perp bc$ weist bei noch höheren Temperaturen ($T > 1.5$ K) ein aussergewöhnliches Verhalten mit zwei Maxima auf. Eine solches Verhalten kann mit Hilfe der Flux-Creep-Theorie von Anderson erklärt werden, wenn man annimmt, dass zwei verschiedene Arten von Haftzentren im Material vorhanden sind. Für diese zwei Typen von Haftzentren erhalten wir für $U_c$ Werte von $3 \pm 1$ meV und $14 \pm 4$ meV.

Die Analyse der Magnetisierungskurven als Funktion der Temperatur zeigt, dass (BEDT-TTF)$_2$Cu(SCN)$_2$ sich wie ein geschichteter Supraleiter mit sehr schwacher Kopplung zwischen den supraleitenden Schichten (bc-Ebenen) verhält. Die Werte von $H_{c1}$ für $H \parallel bc$ sind nämlich im ganzen Temperaturbereich (5 mK-8 K) kleiner als 0.5 Oe. Für $H \perp bc$ zeigt $H_{c1}$ eine unkonventionelle Temperaturabhängigkeit, die durch eine starke Zunahme unterhalb 4 K charakterisiert ist. Die Werte von $H_{c1}^c$ sättigen bei tiefer Temperatur ($T < 1$ K) bei etwa 380 Oe. Eine solche Temperaturabhängigkeit kann durch eine Zunahme der Kondensationsenergie bei tiefer Temperatur auf Grund von Proximity-Effekten zwischen den supraleitenden Schichten verstanden werden. Es stellt sich heraus, dass die $H_{c1}^c(T)$-Kurve einem Exponentialgesetz gehorcht. Dies entspricht dem typischen Verhalten von “breakdown fields” von Proximity-Systemen. Es ist bemerkenswert, dass das Auftreten eines Exponentialgesetzes bei tiefer Temperatur für die “irreversibility line” der stark anisotropen Hoch-$T_c$-Verbindung Bi-Pb-Sr-Ca-Cu-O auch durch Proximity-Effekte erklärt werden kann.

Die Relaxation von $M_{rem}$ wurde bei 4.2 K an einem $Y_1Ba_2Cu_4O_{8+x}$ Einkristall als Funktion des angelegten Feldes ($H \leq 800$ Oe) und der Feldrichtung ($H$ parallel zur $a$-, $b$- und $c$-Achse) untersucht. Für die Relaxationsrate erhalten wir etwa die gleichen Werte ($R \sim 0.05-0.08$) wie für $Y_1Ba_2Cu_3O_7-\delta$. Wegen des beschränkten Feldbereichs unserer Experimente wurde der vollständig kritische Bereich für $H \parallel c$-Achse nicht erreicht, so dass eine abschliessende Studie der Anisotropie der Relaxationsrate nicht möglich war.
INTRODUCTION

In irreversible type-II superconductors in the mixed state pinning allows to build up a flux-line density gradient that screens the material from the penetration of large magnetic fields (H > Hc1). As a result, a certain amount of flux is trapped inside the material, when the magnetic field H is decreased to zero. In this situation the pinning force Fp equals the driving force F arising from the mutual interactions of the flux lines in the flux-density gradient. Thus, at T = 0 no flux motion should occur and the currents inside the superconductor should be persistent. However, at T > 0 thermal energy will allow flux lines to hop over the pinning energy barriers, even if F < Fp. The resulting phenomenon is flux-creep and is responsible for the decrease of the persistent currents inside the superconductor. It was first experimentally observed in 1962 by Kim, Hempstead, and Strnad and then theoretically interpreted by Anderson in terms of thermally-activated flux motion.

In classical superconductors thermally-activated flux-creep is a small effect, so that currents can be considered persistent for any practical purpose. In high-Tc superconductors relaxation effects are much stronger, as demonstrated by the experimental observation of large relaxation rates at temperatures as low as 4 K.

In order to account for these effects, Yeshurun and Malozemoff proposed a model of thermally-activated flux-creep, where the magnitude of the relaxation effects is explained in terms of the small values of the coherence lengths that characterize the high-Tc superconductors. The short coherence lengths result in low pinning energies that, combined with higher temperatures, determine the larger relaxation rates observed in the high-Tc's in comparison to classical superconductors. These effects led to the conclusion that in the high-Tc's pinning is essentially provided by a large number of weak pinning centres. There is now some experimental and theoretical evidence that these weak pinning centres may be represented by oxygen vacancies in the CuO2 layers. Following this assumption another kind of approach to flux-creep was developed, based on the theory of collective pinning developed by Larkin and Ovchinnikov in the 70's. The result is the theory of collective creep by Feigel'man, Geshkenbein, Larkin, and Vinokur (1989), that is based on the idea that a large number
of randomly distributed weak pinning centres produces elastic distortions of the flux-line lattice. By calculating the free energy change associated with these elastic distortions the response of the flux-line lattice to a field gradient was derived.

However, both kinds of models fail to account for the fact that in high-$T_C$ superconductors large relaxation effects subsist down to very low temperatures (in the mK range). This gives evidence that non-thermally activated mechanisms are effective at low temperatures. Recently, several models have been developed, based on the original idea by Mitin that at low temperatures flux lines can tunnel under energy barriers. The theory of quantum collective creep (QCC) by Blatter, Geshkenbein, and Vinokur (1991) is one of them. It was proposed in order to explain the experimental results obtained by several groups on high-$T_C$, Chevrel Phase, heavy-fermion and organic superconductors showing relaxation rates that extrapolate to some finite constant value at low temperatures.

Relaxation effects of the remanent magnetization of classical (NbTi, PbIn), Chevrel Phase PbMo$_6$S$_8$, high-$T_C$ ($Y_1Ba_2Cu_3O_{7.8}$, $Y_1Ba_2Cu_4O_{8+x}$ and $Sr_{0.2}La_{1.8}CuO_4$) and organic [(BEDT-TTF)$_2$Cu(SCN)$_2$] superconductors are the main topic of this thesis.

In a first series of measurements at 4.2 K the magnitude of the relaxation effects in these materials was determined. To that purpose isothermal dc magnetization and time relaxation curves of the remanent magnetization $M_{rem}$ were measured as function of field ($H \leq 500$ Oe). Assuming a logarithmic time dependence, the values of the initial normalized relaxation rates and of the corresponding activation energies at 4.2 K were obtained. The values of the lower critical field $H_{c1}$ were determined from the magnetization curves. In the case of the $Y_1Ba_2Cu_4O_{8+x}$ and of the (BEDT-TTF)$_2$Cu(SCN)$_2$ specimens (single crystals) the measurements were performed in different field orientations in order to study the effects of the layered structure on the magnetic properties. In all cases the field dependence of the remanent magnetization was found to be in agreement with the predictions of the Bean critical state model. Using an anisotropic extension of the same model we estimated the values of the different critical current components (in- and out-of-plane) at 4.2 K.

In a second series of measurements, the temperature dependence of the time relaxation of $M_{rem}$ was investigated (for 5 mK < $T$ < 8 K) on two
single crystals of (BEDT-TTF)$_2$Cu(SCN)$_2$ with applied fields parallel and perpendicular to the strongly superconducting layers. From the analysis of the magnetization curves values of the lower critical field $H_{c1}$ and of the remanent magnetization $M_{rem}$ were determined. The extremely low values of $H_{c1}$ obtained for $H$ parallel to the layers confirm the idea that this material behaves as stack of superconducting sheets with very weak coupling in-between. The temperature dependence of the lower critical field $H_{c1}$ for $H$ perpendicular to the layers is highly unconventional and exhibits an upward curvature. This result can be explained by the occurrence of proximity-induced superconductivity between the superconducting layers of the material at low temperatures. The temperature dependence of $M_{rem}$ was found to be in agreement with the predictions of the Bean critical state model. An anisotropic extension of latter model allowed us to estimate the values of the different critical current components (in- and out-of-plane). The values of the normalized relaxation rates for both field orientations give clear evidence of non-thermally activated flux motion at low temperatures. The theory of quantum collective creep predicts the right order of magnitude of the decay rates at low temperatures and the right dependence on the field orientation as well. At much higher temperatures thermally activated flux-creep occurs and typical values of the activation energies were calculated.

An analysis of the time relaxation law was also performed on the relaxation curves of the remanent magnetization of (BEDT-TTF)$_2$Cu(SCN)$_2$ in order to test the validity of the predictions of the theory of collective creep. The relaxation curves showing deviations from the logarithmic time dependence could be well fitted by a power law.

This work is organized in the following way:

— Chapter II contains an outline of some models and theories of the magnetic properties of irreversible type-II superconductors in the mixed state. They are: Bean's critical state model, Anderson theory of flux-creep, theory of collective creep, quantum collective creep (QCC) theory and vortex-glass model.
— Chapter III describes the two experimental set-ups and the experimental methods used in the analysis of the remanent magnetization data as function of field, temperature and time.

— In chapter IV we report measurements of the low-field magnetic properties of single crystals of (BEDT-TTF)$_2$Cu(SCN)$_2$. A brief description of the samples is given in section IV.1. In section IV.2 we present the analysis and the discussion of the remanent magnetization and time relaxation data measured at 4.2 K as function of field and field orientation. The third section (§ IV.3) of this chapter contains the analysis of the same type of data measured as function of temperature and field orientation. The results are discussed and compared with data on high-T$_C$ superconductors. We will also present (§ IV.4) the results of ac susceptibility measurements performed as function of temperature on two single crystals in different field orientations.

— Chapter V presents an analysis of time relaxation effects of the remanent magnetization on a single crystal of Y$_1$Ba$_2$Cu$_4$O$_{8+x}$. The relaxation data were measured at 4.2 K as function of field in three different field orientations.

— Chapter VI contains a brief overview of the values of the relaxation rates obtained at 4.2 K from low-field magnetic relaxation data on classical, Chevrel Phase, organic and high-T$_C$ superconductors.
II. MODELS OF THE CRITICAL STATE AND FLUX MOTION

The main purpose of this chapter is to sketch some basic theoretical concepts that will be used in the analysis of the magnetization data. Since in our case the samples consisted essentially of type-II superconductors, we will deal with the magnetic properties of type-II superconductors in the mixed state.

One of the key features of the mixed state is its irreversibility. It was shown that the concept of pinning is fundamental in order to understand this phenomenon. In absence of pinning flux lines are able to move freely inside the superconductor. In this case thermodynamical equilibrium can be reached at every point of a magnetization curve. Anything inside the specimen that will interact with the motion of flux lines, will cause an irreversibility of the magnetization. The irreversible part of the magnetization results from the pinning forces that allow to build up flux density gradients in response to external field changes. On the other hand, a density gradient of mutually interacting flux lines gives rise to a driving force that tends to move the flux lines towards the zones of lower flux density. It is clear, that such a configuration will evolve towards an equilibrium situation, characterized by the equality of the pinning force \( F_p \) and the driving force \( F \) everywhere in the sample. However, there are several mechanisms that modify this "equilibrium" situation. At finite temperatures, thermal energy will allow flux lines to move by hopping over the pinning barriers, even if \( F < F_p \). At \( T = 0 \), no thermal energy is available. Nevertheless, it will be shown in this work that flux lines can also tunnel under energy barriers, so that flux motion is possible even at \( T = 0 \).

The critical state model by Bean [1,2] is one of the phenomenological models, that have been proposed in order to account for the magnetic properties of hard type-II superconductors in an external magnetic field. In the first section of this chapter, we will outline some predictions of this model for isotropic materials and for different geometries, that will be useful for the analysis of the magnetization data. Extensions of this model
for anisotropic materials and the influence of sample geometry will also be discussed. The following three sections of this chapter will deal with models of flux motion. We will start by describing Anderson's theory of thermally activated flux-creep [3], based on the critical state model. Section 3 will deal with the problems of flux pinning and flux-creep in superconductors, where pinning is provided by a large number of weak and randomly distributed pinning centres. The collective creep theory proposed by Feigel'man et al. [4,5,6,7] describes flux motion at finite temperatures in the high-$T_c$ superconductors and is based on the original theory of collective pinning developed by Larkin and Ovchinnikov [8,9]. Section 4 will deal with the extension by Blatter et al. [10,11] of the collective creep theory to the quantum tunneling regime, i.e. the region around $T = 0$. The last section of this chapter will outline the basic features of the vortex-glass model by Fisher et al. [12,13,14,15], that suggests the existence of a new thermodynamic phase in the mixed state of high-$T_c$ superconductors. This model also makes predictions about the time dependence of the magnetization and suggests a universal time relaxation law for the high-$T_c$ superconducting oxides.

II.1 Bean's Critical State Model

The critical state model by Bean [1,2] describes the response of a hard type-II superconductor to an external field (or current) change. One of the basic concepts in the critical state model is the existence of a limiting value $J_c$ for the current density that can flow through a superconducting specimen. The value of $J_c$ depends on the magnetic field, but at low fields one can assume that it is field independent. After a field change, shielding currents are induced in the specimen up to the critical value $J_c$. When this value of current is reached the flux starts penetrating deeper into the material, where more shielding currents can be induced.

We will now apply this model to the geometry of an infinite slab of thickness $d$ with an external applied field $H$ parallel to the surface of the slab. The response of the superconducting slab to an applied field is given by
rot \( \mathbf{B} - \mu_0 \varepsilon_0 \frac{\partial \mathbf{E}}{\partial t} = \mu_0 \mathbf{J} \) \hspace{1cm} (II-1)

with \( \mathbf{E} = 0 \) in a superconductor.

Assuming that the thickness of the slab is along the x-axis and that the applied field is along the y-axis one obtains \( \partial \mathbf{B}/\partial x = \mu_0 \mathbf{J} \), where \( \mathbf{J} \) is the current induced in the superconductor in the z-direction by the applied field. This means that the problem becomes one-dimensional, so that with the assumption made above \( (\mathbf{J} = \mathbf{J}_c) \), we obtain:

\[
\frac{\partial \mathbf{B}}{\partial x} = \mu_0 \mathbf{J}_c .
\] \hspace{1cm} (II-2)

The calculation of the average value of the magnetization \( \bar{M} \) takes place through \( \bar{B} = \mu_0 (\mathbf{H} + \bar{M}) \) and

\[
\bar{B} = \frac{1}{d} \int_{-d/2}^{d/2} B(x) \, dx
\] \hspace{1cm} (II-3)

where the origin of the x-axis \( (x = 0) \) is set at the midpoint of the slab thickness. For the average magnetization one obtains:

\[
\bar{M} = \frac{1}{\mu_0 d} \int_{-d/2}^{d/2} B(x) \, dx - \mathbf{H} .
\] \hspace{1cm} (II-4)

The field distribution \( B(x) \) is calculated by taking into account the boundary conditions at the surfaces of the slab \( [B(d/2) = B(-d/2) = \mu_0 \mathbf{H}] \) and by integrating Eq. (II-2). One finally gets

\[
B(x) = \mu_0 \mathbf{H} - \mu_0 \mathbf{J}_c (d/2 - |x|) \quad \text{for} \quad 0 \leq (d/2 - |x|) \leq \Delta .
\] \hspace{1cm} (II-5)

The quantity \( \Delta = \mathbf{H}/\mathbf{J}_c \) is the Bean macroscopic penetration depth for the applied field \( \mathbf{H} \) (assuming \( \mathbf{H}_{c1} = 0 \)). In fact, the induced currents flow in a sheath of maximum thickness \( \Delta \) from the surface. It is noteworthy that the Bean penetration depth \( \Delta \) depends on the applied field. This implies that the measured magnetization curve will depend on the dimensions of the sample. It is useful to define a parametric field \( \mathbf{H}^* = \mathbf{dJ}_c/2 \), as the value of \( \mathbf{H} \) for which the two field fronts penetrating the specimen from both sides meet at the centre of the slab. This means that applied fields bigger than \( \mathbf{H}^* \) penetrate the sample completely.

This model does not include any demagnetization effects. However, the effect of the lower critical field \( \mathbf{H}_{c1} \) can be taken into account. In this case there is no field penetration on a macroscopic scale until the value of the
local field $B(x)$ has reached $\mu_0 H_{c1}$. The shielding currents flow in a layer of thickness $\lambda_L$ (London penetration depth), which is assumed to be much smaller than the thickness of the slab. Even when the field starts penetrating the sample ($H_{c1} < H < 2 H_{c1}$) there is still a portion of the sample around the centre, where $B = 0$. Complete penetration is reached when the Bean penetration depth $\Delta = (H - H_{c1})/J_c$ is equal to $d/2$, i.e. when $H = H^* + H_{c1}$. If the field is reduced by $\delta H$ after having reached a maximum value $H_i$, a current $J_c$ flowing in the opposite direction along the $z$-axis will be induced. This current will flow in a layer of thickness $\Delta = \delta H/(2J_c)$ from the surface of the slab.

We will now apply this model to the case of the remanent magnetization $M_{\text{rem}}$ as function of the cycling field $H_i$ at constant temperature. According to the value of $H_i$ one obtains four different configurations of the distribution $B(x)$ inside the specimen (Fig. II.1). They correspond to the following cases:

a) $H_{c1} \leq H_i \leq 2 H_{c1}$:

$$M_{\text{rem}} = \frac{(H_i - H_{c1})^2}{dJ_c}$$

(II-6)

b) $2 H_{c1} \leq H_i \leq H_{c1} + \frac{d}{2} J_c$:

$$M_{\text{rem}} = \frac{(H_i^2 - 2H_{c1}^2)}{2dJ_c}$$

(II-7)

c) $H_{c1} + \frac{d}{2} J_c \leq H_i \leq dJ_c$:

$$M_{\text{rem}} = (H_i - \frac{H_i^2}{2dJ_c} - \frac{dJ_c}{4})$$

(II-8)

d) $H_i \geq dJ_c$:

$$M_{\text{rem}} = \frac{dJ_c}{4}$$

(II-9)

From these field distributions one clearly sees that the remanent magnetization does not depend on the applied field only in the field regime d), where $M_{\text{rem}}$ is constant. We will refer to this regime as the fully (or completely) critical regime. From the saturation value of $M_{\text{rem}}$ one can estimate the value of $J_c$.

It is also straightforward to see [Fig. II.1,d)] that in the fully critical
Fig. II.1  Field distribution $B(x)$ at different temperatures and constant cycling field $H_i$, calculated on the basis of Bean's critical state model. The shaded areas correspond to the remanent magnetization $M_{\text{rem}}$. Qualitatively, one obtains the same field distributions as function of applied field at constant temperature.
regime the time relaxation of the remanent magnetization will occur outwards, i.e. the trapped flux will relax only towards the outside of the specimen. The situation described in Fig. II.1,a) is somewhat different, because of the central region of the sample, where $B = 0$. The effect of $H_{c1}$ is approximated by the vertical slope at $x = \pm \left[ d/2 - (H_i - H_{c1})/J_c \right]$, so that also in this case (as in Fig. II.1,d)) the trapped flux relaxes towards the outside of the specimen. The situation is quite different in the cases depicted in Fig. IV.1,b) and c), where there is also a part of the field profile that relaxes inwards, i.e. towards the central plane of the slab. This relaxation cannot be measured in our experiments, because the gradiometer cannot detect flux motion taking place within the specimen (§ III.2.3). For this reason, we will now calculate the part $M_{\text{rem}}^{\text{out}} = f \cdot M_{\text{rem}}$ of the remanent magnetization, that decays outwards. This quantity will be used to normalize the relaxation data in the next chapters. From the field distributions shown in Fig. II.1, we obtain:

a) $H_{c1} \leq H_i \leq 2H_{c1}$:

$$M_{\text{rem}}^{\text{out}} = M_{\text{rem}} = \frac{(H_i - H_{c1})^2}{dJ_c} \quad (II-10)$$

b) $2H_{c1} \leq H_i \leq H_{c1} + \frac{d}{2} J_c$:

$$M_{\text{rem}}^{\text{out}} = \frac{M_{\text{rem}}}{2 - 4 \left( \frac{H_{c1}}{H_i} \right)^2} = \frac{H_i^2}{4dJ_c} \quad (II-11)$$

c) $H_{c1} + \frac{d}{2} J_c \leq H_i \leq dJ_c$:

$$M_{\text{rem}}^{\text{out}} = \frac{M_{\text{rem}}}{2 - \left( \frac{dJ_c}{H_i} - 2 \right)^2} = \frac{H_i^2}{4dJ_c} \quad (II-12)$$

d) $H_i \geq dJ_c$:

$$M_{\text{rem}}^{\text{out}} = M_{\text{rem}} = \frac{dJ_c}{4} \quad (II-13)$$

As stated before, $M_{\text{rem}}^{\text{out}}$ is different from $M_{\text{rem}}$ only in the two field regimes b) and c), where $M_{\text{rem}}^{\text{out}}$ follows a parabolic field dependence. The value of the fraction $f$ always lies between 0.5 and unity. Since all the measured (magnetization) data refer to average values of the
magnetization of the specimen, we will simply use the symbol M throughout this work.

This model gives a satisfactory account of the dependence on the applied field $H_i$ of the remanent magnetization of isotropic type-II superconductors for the geometry described above. In order to analyse magnetization data of high-$T_c$ or organic superconductors we have to take into account anisotropy effects. A further aspect that should also be taken into account is represented by demagnetization effects affecting the data (on single crystals) taken with an applied field perpendicular to the basal plane of the specimens. These two aspects will be briefly discussed in the next paragraphs.

Recently, Gyorgy et al. [16] have proposed an extended version of the Bean model in order to take into account anisotropy effects of the critical current. They consider a superconducting parallelepiped in the fully critical state. The external field is applied perpendicularly to the base of the parallelepiped [Fig. II.2,a)], which has dimensions $t$ (thickness) and $L_\perp$. The latter is the length of the parallelepiped transverse to the field, while $L_\parallel$ is the length parallel to the field. Assuming that the plane $(L_\perp \times L_\parallel)$ of the parallelepiped corresponds to the ab-plane of a high-$T_c$ single crystal (Fig. II.2), one can define $J_{c,c}^{c,ab}$ and $J_{c,c}^{c,ab}$ as the critical current components parallel to $t$ and $L_\perp$, respectively. For an applied field perpendicular to the ab-plane [Fig. II.2,b]) there is a critical current component $J_{c,c}^{c,ab,c}$ flowing in the ab-plane. Here we have used the notation of Cronemeyer et al. [17], where the first and the second superscripts denote the direction of the induced current and of the applied field, respectively. This notation follows from the Lorentz force formula $\mathbf{F}_L = \mathbf{J} \times \mathbf{B}$, so that it becomes clear that there are actually two anisotropic components $J_{c,c}^{c,ab}$ and $J_{c,c}^{c,ab,c}$ of the critical current in the ab-plane, depending on the direction of the applied field. In the two cases the Lorentz force has different directions, namely perpendicular ($J_{c,c}^{c,ab}$) and parallel ($J_{c,c}^{c,ab,c}$) to the ab-planes. In fact, when the Lorentz force tends to move the vortices through the strongly superconducting planes, the pinning force should be very large. This is due to the very short coherence length in the $c$-direction that causes a modulation of the order parameter in this direction, so that the layered structure itself works as pinning centre. This has been taken [18] as the
Fig. II.2 Schematic representation of the different critical current components contributing to the remanent magnetization of a single crystal of high-Tc (or layered) superconductor as function of the field orientation. For \( H \parallel c \) there is a single current component that flows within the ab-planes. For \( H \parallel ab \) there are two current components parallel and perpendicular to the layers.

Gyorgy et al. [16] have calculated the profile of the remanent magnetization (fully critical state) inside the sample (Fig. IV.3) for the finite geometry described in Fig. II.2. Campbell and Evetts [19] have shown that in the isotropic case the "critical state equation" \( F_p = \text{rot} \mathbf{H} \wedge \mathbf{B} = \mathbf{J}_c \wedge \mathbf{B} \) is identical to the one determining the maximum height of a sandpile on the surface under consideration (with dimensions \( t \times L_x \)). The corresponding profile is very similar to the one shown in Fig. II.3, but for the fact that the slope of the profile is the same in every direction.

In the fully critical state and for \( H_i \parallel ab \) the value of the remanent magnetization \( M_{\text{rem}} \) is given by:

\[
M_{\text{rem}} = \frac{tJ_{c,ab}^{ab,ab}}{4} \left( 1 - \frac{tJ_{c,ab}^{ab,ab}}{3L_{\perp}J_{c,ab}^{e,ab}} \right) \quad \text{for} \quad \frac{tJ_{c,ab}^{ab,ab}}{L_{\perp}J_{c,ab}^{e,ab}} < 1 \quad (\text{II-14})
\]
Fig. II.3 Profile of the irreversible magnetization inside a high-Tc (anisotropic) superconducting sample in the fully critical state for the case $tJ_{c,ab} > L_\perp J_{c,ab}$. The values of the critical current components are given by the slopes of the profile: $h/k = J_{c,ab}$ and $2h/L_\perp = J_{c,c,ab}$.

$$M_{\text{rem}} = \frac{L_\perp J_{c,ab}^c}{4} \left( 1 - \frac{L_\perp J_{c,ab}^c}{3tJ_{c,ab}^{ab}} \right) \quad \text{for} \quad \frac{tJ_{c,ab}^{ab}}{L_\perp J_{c,ab}^c} > 1 \quad (\text{II-15})$$

It is clear from Eq. (II-15) that in the case of strong anisotropy, i.e. $J_{c,ab}^c \ll J_{c,ab}^{ab}$, this formula reduces to one for an infinite slab in the isotropic case, unless the aspect ratio $L_\perp/t$ of the crystal is bigger than the ratio $J_{c,ab}^{ab}/J_{c,ab}^c$ [Eq. (II-14)]. However, due to the typical dimensions of high-Tc or organic single crystals, the current component dominating the critical state in most cases [Eq. (II-15)] is in the direction perpendicular to the ab-plane. Then the important dimension is the transverse length $L_\perp$ and not the thickness, as one would expect in the case of a slab parallel to the field.

In high-Tc compounds typical values of the ratio $J_{c,ab}^{ab}/J_{c,ab}^c$ are of the order of 3 to 5, so that it is clear that the anisotropic component $J_{c,ab}^{ab}$ (intrinsic pinning) will dominate the critical state only in the case of specimens with a large ($\geq 3-5$) aspect ratio $L_\perp/t$, i.e. in the case of very thin single crystals or thin films. Otherwise, since Eqs. (II-14) and (II-15) contain two critical current components, it is actually necessary to measure at least two specimens with different aspect ratios (in the same field orientation) in order to determine both $J_{c,ab}^{ab}$ and $J_{c,ab}^c$. 
For applied fields $H_i$ perpendicular to the ab-plane, one obtains

$$M_{rem} = \frac{L \cdot J_{c}^{ab,c}}{4} \left(1 - \frac{L_\perp}{3L_\parallel}\right) \quad \text{for } L_\perp < L_\parallel,$$

(II-16)

where the critical state is dominated by the in-plane component $J_{c}^{ab,c}$ of the critical current. In this case the Lorentz force is parallel to the ab-plane, so that $J_{c}^{ab,c}$ should actually have a lower value than $J_{c}^{ab,ab}$. Moreover, in the case of thin platelets ($H \| t$) flux lines are parallel to the applied field only in the zones at the centre and at the edges of the specimen. In most of the basal plane of the platelet the flux lines are parallel to the surface of the sample, i.e. approximately perpendicular to the applied field. In these zones the critical current component $J_{c}^{ab,ab}$ is likely to give a significant contribution to the critical state. The extent of this contribution is not well established up to now, but it is clear that the values of $J_{c}^{ab,c}$ obtained from (II-16) should be considered as an upper limit.

Recently, theoretical calculations by Däubling and Larbalestier [20] have shown that at low fields self-field effects have a strong influence, leading to a significant curvature of the flux lines. In their model, one considers a thin disk-shaped isotropic superconductor (cylinder geometry, radius $R$, thickness $t$ and $R \gg t$) with field applied perpendicular to the plane of the disk ($H \perp z$-axis). With this geometry the induced currents $J$ flow in the plane of the disk. The problem is solved by calculating the field distribution due to the induced current $J$. The obtained field (axial and radial components) is then added to the externally applied field. The problem is solved self-consistently, so that $J = J_{c}(H)$ at every point, where $H$ is the total field. The field dependence of $J_{c}$ is assumed to be of the Kim-type, i.e. $J_{c}(H) \approx \alpha/(H + H_0)$. From the calculation one obtains an axial and a radial field component $H_r(r,z)$ and $H_t(r,z)$, respectively.

The calculations show that $H_t(r,z)$ is maximum (almost constant!) at the surface of the disk and of the order of $tJ_{c}/2$. This field component reverses sign between the upper and the lower surface and is zero in the mid-plane. The axial component $H_r(r,z)$ at the centre of the disk is also of the order of $tJ_{c}$ and is weakly dependent on $z$. This means that the field trapped (or shielded) at the centre of the disk is of the order $tJ_{c}$. Moreover, in the case of thin disks ($R \gg t$) it is the field gradient $\partial H_r(r,z)/\partial z$ along the thickness that determines the critical state, being bigger than the
one along the radius (by a factor $\sim R/t$).

Latter prediction has been confirmed by more extensive calculations by Conner and Malozemoff [21]. However, in the case of a field-dependent $J_c$ (e.g. Kim-type) the value of the gradient $\partial H_z(r,z)/\partial z$ is found to be much smaller than the expected low-field limit ($J_c = \alpha/H_0$). So, the main result is that the remanent magnetization depends approximately linearly on the radius $R$ even in the case of field-dependent $J_c$. However, because of self-fields, the value of $J_c$ obtained at low fields ($H \leq tJ_c/2$) is much lower than the expected one ($J_c = \alpha/H_0$). The prediction obtained with the conventional model is recovered at high fields ($H \gg tJ_c$), where the variation of $B$ (and $J_c$) over the radius of the disk is small. In this case flux lines are almost parallel to the direction of the applied field, so that the critical state depends on the gradient $\partial H_z(r,z)/\partial r$, that can be approximated by a straight line [slope $= J_c(H)$].

In conclusion, the determination of $J_c$ from magnetization measurements at low fields ($H \leq tJ_c/2$) should yield lower values of $J_c$, because of self-field effects. For the same reason, the determination of the field dependence of the critical current from magnetization measurements in this field regime would give a dependence weaker than the real one.

Estimations [20] of the magnitude of self-fields at 4.2 K for a typical $Y_1Ba_2Cu_3O_{7-8}$ single crystal yield values of the order of 1-3 T. However, the magnitude of these effects has to be confirmed by more experimental data, even if Yeshurun et al. [22] have found some evidence in favour of this model in magnetization data of Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O single crystals of different size.

### II.2 Anderson's Flux-Creep Theory

This model [3] considers the dynamics of vortices in a type-II superconductor in the critical state, where the gradient of the field is determined by the critical current ($\nabla B = \mu_0J_c$). For convenience, we will mainly follow the description of this model given by Beasley, Labusch and Webb [23].
In the critical state model (at \( T = 0 \)) flux lines inside a superconductor can move only if the driving force density \( F \) (Lorentz force) exceeds the pinning force density \( F_p \). The application of a large magnetic field to a hard type-II superconducting sample builds up a density gradient in the distribution of mutually interacting flux lines. This gives rise to a driving force on the flux lines. The array of flux lines will relax in such a way that the driving force equals the pinning force everywhere. In such a state, if the pinning force is known, the currents flowing in the specimen will be uniquely determined. Moreover, in such an arrangement motion of single flux lines is energetically unfavourable, due to mutual repulsive interactions between them. Collective motion of flux lines is therefore more likely to take place. We will call flux bundle (volume \( V \)) the cluster of neighbouring flux lines, that is expected to act collectively during flux motion.

Anderson [3] pointed out that at a finite temperature flux motion is possible even if \( F < F_p \), because of thermal energy. According to his model flux bundles will hop over pinning barriers at a rate \( v = v_0 e^{-U/kT} \), where \( U \) is an effective activation energy and \( v_0 \) is an attempt frequency. Of course, the rate of thermally activated flux-creep is strongly enhanced by the driving force caused by an applied field. Indeed, after a field change, when \( F \) falls well below \( F_p \), the rate of motion of the fluxoids diminishes drastically. The activation energy is thus a decreasing function of \( |VB| \). (From here on we will assume that the magnetic field \( B \) is along the longitudinal z-axis of a cylindrical superconducting sample.) If one takes the activation energy \( U \) as an average value of the pinning energy in a washboard-like potential, then one can write

\[
U = U_0 - |F| V X = U_0 - J_c B V X \quad \text{(II-17)}
\]

where \( U_0 \) is the value of the activation energy for thermally activated flux-creep in absence of flux density gradients, while the second term is the decrease of the height of the barrier due to a field gradient. The effect of a gradient (external field or current) is to tilt the washboard potential and therefore to decrease the activation energy \( U \). The length \( X \) represents the width of the pinning barrier and \( V \) denotes the volume of the flux bundle.

In order to calculate a flux-creep rate at a finite temperature, it is
useful to define a flux-flow density $D$ as:

$$D = -(\nabla B/|\nabla B|) B w v_0 e^{-U(B,|\nabla B|)/kT} \tag{II-18}$$

where $w$ is the average hopping distance of the flux bundle for a thermally activated jump. Conservation of flux then requires that

$$\frac{dB}{dt} = -\nabla \cdot D. \tag{II-19}$$

This equation has been solved by Anderson and Kim for the geometry of a superconducting hollow cylinder. Beasley et al. [23] solved it for a bulk superconducting cylinder. For the radial component $D_r$ of the flux-flow density Eq. (II-19) implies:

$$\frac{\partial B}{\partial t} = -\frac{1}{r} \frac{\partial}{\partial r} (r D_r). \tag{II-20}$$

The total flux change $\Phi$ through the specimen can be calculated from

$$\frac{\partial \Phi}{\partial t} = -2\pi \rho D_r(\rho, t) \tag{II-21}$$

where $\rho$ is the radius of the cylindrical specimen. The corresponding equations can be solved in the case of $k_B T/U << 1$ and the solution given is a first order approximation of an expansion in powers of $k_B T/U$. The total flux through the specimen at a time $t$ is given by:

$$\Phi(t) = \Phi(t_0) \pm 2\pi \rho f(\rho) \ln(t/t_0) \tag{II-22}$$

where $t_0$ is an arbitrary reference time, while $f(\rho)$ is a radial function. The $\pm$ signs refer to the sign of the gradient $\nabla B$, i.e. to increasing and decreasing external fields, respectively. A logarithmic time dependence was calculated by Anderson [3] for the case of a hollow cylinder and is consistent with the result of this calculation for the case of a bulk cylindrical specimen. By taking into account the boundary conditions at the surface and assuming that flux motion is not controlled by surface energy barriers, one obtains:

$$\frac{\partial \Phi}{\partial \ln t} = \pm 2\pi \rho f(\rho) = \pm \frac{\pi}{3} k_B T \rho^3 \left(\frac{\partial U}{\partial |\nabla B|}\right)_{t_1, \rho}^{-1} (1 \pm \delta) \tag{II-23}$$

where $\delta$ is a factor much smaller than unity and the subscripts indicate that the corresponding quantity has been evaluated at the surface ($r = \rho$) and at a time $t_1$ in the critical state. In the range $U << U_0$, Eq. (II-23)
yields
\[ \frac{\partial \Phi}{\partial \ln t} = \frac{k_B T}{U_0} \pi \rho^2 \frac{\mu_0}{3} \rho J_c . \] (II-24)

Inserting \( \overline{M} = \rho J_c / 3 \) and \( \Phi = \mu_0 \rho^2 \pi \overline{M} \) into Eq. (II-24), we obtain:
\[ \frac{1}{\overline{M}} \frac{\partial \overline{M}}{\partial \ln t} = \frac{k_B T}{U_0} . \] (II-25)

The final result is that the normalized logarithmic creep rate is equal to the ratio of the thermal energy to the activation energy. Equation (II-25) shows clearly that only for \( k_B T \ll U_0 \) the relaxation rate is small enough, so that one can consider the sample in a truly critical state. In fact, under this condition the amount of flux involved in the relaxation process is very small, so that the balance between the driving and the pinning force inside the sample can be achieved.

II.3 Theory of Collective Flux-Creep

This theory of thermally activated flux-creep by Feigel'man, Geshkenbein, Larkin, and Vinokur [4,5,6] is based on the idea of pinning by a large number of weak and randomly distributed pinning centres. The problem is treated using the formalism of the collective pinning theory developed by Larkin and Ovchinnikov [8,9] in the 70's.

It is well known that in a homogeneous type-II superconductor at sufficiently high fields, a periodic structure of flux lines develops. This structure is the equilibrium configuration of the flux lines inside the material and corresponds to a triangular lattice (Abrikosov lattice). The presence of inhomogeneities (e.g. structure defects) affects this configuration by creating elastic distortions of the flux-line lattice (FLL). The free energy change \( F \) associated with these distortions [8] in the case of a magnetic field \( B \) along the z-axis is
\[ F = \int d^3r \left[ \frac{(C_{11} - C_{66}) (\text{div} \mathbf{u})^2}{2} + C_{66} \frac{(\nabla \cdot \mathbf{u})^2}{2} + C_{44} \frac{(\partial u/\partial z)^2}{2} + U_{\text{pin}}(\mathbf{u},r) \right] \] (II-26)
where \( \mathbf{u}(\mathbf{r}) \) is the two-dimensional displacement vector of the FLL, while \( C_{11}, C_{44}, C_{66} \) are the elastic compression (bulk), tilt, and shear moduli, respectively. \( U_{\text{pin}}(\mathbf{u}, \mathbf{r}) \) is the random potential describing the interaction of the FLL with the defects. The random potential arising from the weak pinning centres is supposed to be short-ranged, so that it falls off rapidly at a typical distance \( r_p \). In the case of defects with dimensions smaller than \( \xi \), the distance \( r_p \) is of the order of \( \xi \).

By minimizing the free energy change due to the elastic distortions of the FLL, Larkin [8] calculated the quantity \( (\mathbf{u}(\mathbf{R}) - \mathbf{u}(0))^2 \), that describes the deviation of the structure from a periodic array. This quantity is proportional to \( W \) (mean square value of the random force of the defects) and increases with \( R \), so that for small values of \( W \) short-range order exists, but long-range order is destroyed. One can therefore define a pinning volume \( V_c \), as the volume inside which short-range order exists. One defines the volume \( V_c = R_c^2 L_c \) by the condition \( |\mathbf{u}(\mathbf{r})| \leq \xi \), so that within this volume vortices are arranged almost periodically. \( R_c \) and \( L_c \) are the transverse and longitudinal dimensions (with respect to the magnetic field) of the volume \( V_c \), that is collectively pinned as a whole. The critical current density \( J_c \) can be expressed [9] in terms of the dimensions of the volume \( V_c \), namely

\[
J_c \approx (W/V_c)^{1/2} B^{-1} \approx \frac{W^{1/2}}{R_c L_c^{1/2} B}.
\]

At values of current \( J < J_c \) [4] the FLL is in a metastable state and transitions to other (more favourable) metastable states take place through thermal activation across free energy barriers of the order \( U(J) \). In the case of collective pinning the energy \( U(J) \) is of the order of the elastic energy of the hopping flux bundle (volume \( V_B \)) [3]. In order to get some insight into the dependence of the activation energy \( U \) on the current, we will now consider different regimes of currents and fields.

For values of current \( J \) close to \( J_c \) the hopping distance is \( u_{\text{hop}} \approx \xi \). At low fields (\( B \sim H_{c1} \)), the flux bundle volume is approximately equal to the coherence volume \( V_c \). At higher fields (\( B \gg H_{c1} \)) the hopping of a single flux bundle \( V_c \) would involve a large compression in the direction of the hopping. In order to minimize this compression several bundles \( V_c \) will
hop simultaneously, so that the effective flux bundle moving is made of a large number \[ (C_{11}/C_{66})^{1/2} \] of subbundles with volume \( V_c \). The volume of the "superbundle" is \( V_B = R_{\perp} R_l L \), where \( L \), \( R_\perp \), and \( R_l \) are the dimensions of the superbundle along the magnetic field, transverse and parallel to the direction of hopping, respectively. The energy barrier \( U_c \) for a correlated hop is approximately equal to the sum of the energy barriers for the subbundles.

The situation described above (\( J = J_c \)) corresponds to the typical region of flux-creep measurements in classical type-II superconductors. In this case the activation energy is large and the temperature is low, so that flux-creep is weak and the current value is close to \( J_c \). The effect of flux-creep is much larger in the high-\( T_c \) superconductors, so that measurements take place at values of current that are often considerably lower than \( J_c \).

When a sufficiently strong magnetic field is applied to a superconducting sample, the critical state [1] develops inside the specimen within a short time scale. The value of the current at a later observation time \( t_{\text{obs}} \) is smaller, due to flux-creep, and is determined by the condition

\[
U(J(t_{\text{obs}})) = k_B T \cdot \ln(t_{\text{obs}}/\tau_0)
\]  

(II-28)

where the term \( \ln(t_{\text{obs}}/\tau_0) \) is typically of the order of 10-30.

In the theory of thermally activated flux-creep [3] it is usually assumed that \( (J_c - J) \ll J_c \) and that \( U(J) \approx U_c(1 - J/J_c) \), so that one obtains a logarithmic time dependence of the current: \( J(t) = J_c [1 - (k_B T/U_c) \ln(t/\tau_0)] \). Several assumptions can be made on the dependence of \( U(J) \) in the low-current regime. Anderson [3] assumed that the energy barrier grows linearly with decreasing values of \( J \), so that \( U(J \approx 0) \) is a constant. One of the main results of the collective creep theory is to show that the activation energy increases as \( U \propto J^{-\alpha} \) at low currents.

Qualitatively, the power law dependence of \( U(J) \) can be understood in the following way. The effect of a current on the flux-line lattice is to make some other metastable state more favourable than the actual one. These states are determined by the balance between the gain in the Lorentz force energy and the elastic (pinning) energy of the flux-line lattice. At \( J = J_c \) this balance is fulfilled by neighbouring states, that the flux bundle can attain by hopping over a distance \( u_{\text{hop}} \) of the order of \( \xi \).
while at current values $J << J_c$ the hopping distance of the flux bundle becomes much larger. It can be shown (§ II.5), that the elastic energy involved in the displacement of a segment of flux line scales as a positive power of the minimal length of the segment displaced. At low currents the minimal segment length is inversely proportional to the current, so that one can express the activation energy as $U(J) = U_c(J/J_c)^\alpha$.

Inserting this result into Eq. (II-28), that determines the value of the current, we obtain in the asymptotic limit ($J << J_c$)

$$J(t) = J_c \left( \frac{U_c}{k_B T \ln(t/\tau_0)} \right)^{1/\alpha}.$$  

(II-29)

It turns out [4] that $\alpha$ depends on the characteristics of the FLL, so that it depends both on temperature and magnetic field. According to the values of these quantities, one obtains several regimes of pinning and flux-creep.

Keeping in mind that this theory is valid in the region of low-to-intermediate temperatures and fields and for hopping distances $u_{hop}$ much smaller than the FLL constant $a_{\Phi_o}$, one obtains essentially three regimes:

i) $J_1 \sim J_c(L_c/a_{\Phi_o})^{7/5} < J < J_c$:
low $T$ and low magnetic field $B$ [$J_c(B) \approx \text{const.}$] and $R_c < a_{\Phi_o}$, so that the critical current is determined by collective pinning of single vortices. Pinning is characterized by the length $L_c$ ($\xi < L_c < a_{\Phi_o}$), that is the length of the segment of flux line collectively pinned (independent of field). One obtains:

$$\alpha = 1/7, \quad U(J) \approx U_c(j_o/j_c)^{1/7},$$

$$U_c \sim H_c^2 \xi^3 (\xi/L_c), \quad J_c \sim j_o (\xi/L_c)^2 \sim (H_c/\lambda)(\xi/L_c)^2$$

$$J(t) = J_c \left( \frac{U_c}{k_B T \ln(t/\tau_0)} \right)^{7}.$$  

(II-30)

ii) $J_2 = j_1(a_{\Phi_o}/\lambda)^2 < J < J_1$:
flux-creep of small flux bundles with transverse size $R_\perp$ ($\sim R_c$) smaller than $\lambda$ ($a_{\Phi_o} < R_\perp < \lambda$). The size in the direction of hopping is larger: $R_\parallel \sim R_\perp(R_\perp/a_{\Phi_o}) \sim R_\perp(C_{11}/C_{66})^{1/2} \sim L \sim L_c$. One obtains:

$$\alpha = 3/2, \quad U(J) \approx U_1(j_1/j_c)^{3/2},$$

$$U_1 = U_c(a_{\Phi_o}/L_c)^{1/5} < U_c$$. 
\[ J(t) = J_1 \left( \frac{U_1}{k_B T \ln(t/\tau_0)} \right)^{2/3} \]  \hspace{1cm} (II-31)

iii) \( J < J_2 = J_1(a_\phi/\lambda)^2 \) : the flux bundles are much larger than \( \lambda \). One gets:
\[ \alpha = 7/9, \quad U(J) = U_2 (J_2/J)^{7/9} \]
\[ U_2 = U_1 (\lambda/a_\phi)^3 > U_1 \]
\[ J(t) = J_2 \left( \frac{U_2}{k_B T \ln(t/\tau_0)} \right)^{9/7} \]  \hspace{1cm} (II-32)

The theory of collective flux-creep described above has also been adapted to the case of layered superconductors [24,25]. In these materials the dimensionality and the characteristic of the FLL depend on the strength of the interlayer coupling, so that qualitatively one obtains the same dependence for \( U(J) \) and \( J(t) \) as above, but with different values of \( \alpha \).

We will now discuss how the results of the collective flux-creep theory can be applied to magnetization or current relaxation experiments in order to get reliable information about pinning [5]. The main problem involved in this kind of measurements on high-\( T_C \) superconductors is that flux-creep is so strong that very often the measured values of current are well below the true value of the critical current \( J_C \). Another consequence is the fact that the logarithmic relaxation rate \( |d\ln M/d\ln t| \) cannot be directly related to one value of the activation energy, because the actual value of the pinning barrier (at a certain stage of the relaxation process) depends on the actual value of the current.

It was previously shown that at low currents the activation energy depends strongly on current as \( U(J) = U_0 (J_c/J)^\alpha \), where \( U_0 \) is the characteristic energy barrier at low currents. This \( U(J) \) dependence gives rise to a nonlinear current-voltage characteristic \( V \propto \exp(-(A/J^\alpha)) \), and so to a zero linear resistance: \( \rho_{\text{lin}} = \left( \frac{dV}{dJ} \right)_{J \to 0} = 0 \). This is also one of the main features of the vortex-glass state [12].
For the relaxation of currents arising from some thermally activated process, the relevant energy barrier is related to the "persistent" current $J(t)$ in the way described by Eq. (II-28). In the long-time limit one obtains for the asymptotic form (i.e. for $J \ll J_c$) of the relaxation law:

$$J(t) = J_c \left( \frac{U_0}{\frac{k_B T}{U_c} \ln(t/\tau_0)} \right)^{1/\alpha}.$$  (II-33)

It was also shown that at the initial stage $[(J_c - J) \ll J_c]$ of the relaxation process the Anderson formula

$$J(t) = J_c \left( 1 - \frac{k_B T}{U_c} \ln(t/\tau_0) \right)$$  (II-34)

should be valid [$U_c \approx U(J \sim J_c)$]. It is then possible to write an interpolation formula for the whole relaxation process:

$$J(t) = J_c \left( 1 + \alpha \frac{k_B T}{U_c} \ln(t/\tau_0) \right)^{-1/\alpha}.$$  (II-35)

A comparison between (II-33) and (II-35) in the corresponding limit ($t \to \infty$, i.e. $J \ll J_c$) shows that $U_c = \alpha U_0$. The normalized relaxation rate $S = -d \ln J(t)/dt$ can then be obtained from Eq. (II-35). It yields

$$S = \frac{k_B T}{U_c + \alpha \frac{k_B T}{U_c} \ln(t/\tau_0)}.$$  (II-36)

It is clear that defining an activation energy as $U_a = k_B T/S$, implies that the apparent activation energy $U_a$ should increase linearly with $T$ at higher temperatures.

In the case of single-vortex creep the theoretical value of $\alpha$ is $1/7$ and for $\alpha (k_B T/U_c) \ln(t/\tau_0) \ll 1$, Eq. (II-35) can be approximated by

$$J(t) = J_c \exp\left( -\frac{k_B T \ln(t/\tau_0)}{U_c} \right) = J_c \left( \frac{t}{\tau_0} \right)^{k_B T/U_c}. $$  (II-37)

According to Anderson's theory of flux-creep, the time $\tau_0$ corresponds to the attempt frequency $\nu_0$ (§ II.2) for hopping of a flux bundle over an energy barrier, so that $\tau_0$ is a microscopic time, typically [26] of the order of $10^{-5}$-$10^{-12}$ s. However, recent results [5] point to bigger values of $\tau_0$ ($10^{-6}$-$10^{-3}$ s). The value of the microscopic time $\tau_0$ is still an open question and further work on this issue is needed.
II.4 Theory of Quantum Collective Flux-Creep

We will now outline the theory of quantum collective creep (QCC-theory) by Blatter, Geshkenbein and Vinokur [10]. We will first describe the basic features of this theory for the case of isotropic superconductors and currents close to the critical one.

The tunneling rate $\gamma$ of flux lines under energy barriers is given by the effective Euclidean action $S^\text{eff}_E$ of the tunneling process, according to $\gamma \propto \exp(- S^\ast_E/h)$ [cf. § II.2, flux-creep rate for thermally-activated motion $v \propto \exp(-U(J)/k_BT)$]. The Lagrangian determining the classical dynamic equations of the vortex is

$$L[u] = \int dz \, \frac{M}{2} (\partial_t u)^2 - \mathcal{F}[u]$$

where $\mathcal{F}[u]$ is the free-energy functional described by

$$\mathcal{F}[u] = \int dz \, \frac{\epsilon}{2} (\partial_z u)^2 + U_{\text{pin}}(z,u).$$

Here, $M$ is the vortex mass per unit length, $u(z,t)$ is a two-dimensional field describing the displacement of the vortex from its equilibrium position ($B \parallel z$-axis), $\epsilon$ is the line tension of the vortex and $U_{\text{pin}}$ denotes the pinning potential.

In order to estimate the vortex mass, Blatter et al. [10] assumed that the main contribution is of electronic origin and that it is given by the local change of dispersion within the vortex core. The number of electrons exposed to this change is approximately $N(0)\pi \xi^2 \Delta$, where $N(0)$ is the density of states at the Fermi energy $\varepsilon_F$, $\Delta$ is the energy gap and $\xi$ is the coherence length. By multiplication with the value of the effective mass change $m\Delta/\varepsilon_F$ per electron, one obtains an estimate of the vortex mass $M \approx M_{\text{el}} = (3\pi/2)m \eta^2 (\Delta/\varepsilon_F)^2 = (2/\pi^3)mk_F$ ($n$ is the electron density, $m$ is the effective mass and $k_F$ is the Fermi wave vector). This estimate agrees with the results obtained by Suhl [27], who used time-dependent GL-theory.

In the framework of the collective creep theory and in the case of single-vortex pinning a segment of length $L_c$ of a single flux line is
collectively pinned. The length $L_c$ is determined by minimization of the free-energy functional $\mathcal{F}$ described by Eq. (II-39) and is independent of field: $L_c \approx \xi (J_0/J_c)^{1/2}$ (previous section). Within the collective pinning theory the rate of thermally activated flux-creep is determined by the saddle-point solution of Eq. (II-28).

> In order to obtain the rate of creep due to quantum tunneling one has to find the saddle-point solution of the Euclidean action of the process, that is given by

$$S_E = \int dt \left\{ \int dz \left[ \frac{M}{2} (\partial_t u)^2 + \mathcal{F}[u] \right] \right\}.$$  \hfill (II-40)

Blatter et al. [4] calculated the action $S_E$, using the method of the dimensional estimates. It actually turns out that quantum creep in $d$ dimensions can be mapped onto the problem of classical creep in $d+1$ dimensions, so that the time axis can be considered as an additional variable in the minimisation problem.

The length $L_c$ corresponds to the geometrical part of the solution and has been determined above. The tunneling time $t_c$ can be estimated by equating the elastic and the kinetic energy density in Eq. (II-40). This yields $\varepsilon_t (\xi/L_c)^2 \approx M(\xi/t_c)^2$, and one obtains $t_c \approx L_c (M/\varepsilon_t)^{1/2}$. The corresponding action is

$$S_E \approx t_c U_c \approx L_c (M/\varepsilon_t)^{1/2} \varepsilon_t (\xi/L_c)^2 L_c \approx \xi^2 (M\varepsilon_t)^{1/2}. \quad \hfill (II-41)$$

This equation shows that in the limit of weak fields and vanishing dissipation the action $S_E$ is independent of $L_c$ and therefore of the strength of the pinning potential.

Dissipative motion of vortices will now be included in the model. This is done by inserting an extra term (non-local in time) in Eq. (II-40). The corresponding action is then denoted by $S_E^{\text{eff}}$ (effective action). After Fourier transformation, one obtains

$$S_E^{\text{eff}} = \int \frac{d\omega}{2\pi} \int \frac{dq}{2\pi} \left\{ \frac{1}{2} \left[ (M + \eta/|\omega|) \omega^2 + \varepsilon_t q^2 \right] |u(q,\omega)|^2 + U_{\text{pin}}(q,u) \right\}$$ \hfill (II-42)

where $\eta$ is the viscous drag coefficient given by $\eta = \Phi_0 H_c c^2/(c^2 \rho_n)$, with $\rho_n$ denoting the normal-state resistivity. Equation (II-42) shows that the
inclusion of dissipation in the model corresponds to the substitution of the mass \( M \) with an enhanced mass \( M_{\text{eff}} = M[1 + \eta/(M\omega)] \).

The saddle-point solution of Eq. (II-42) produces a larger tunneling time \( t_c \), that is solution of the quadratic equation \( M_{\text{eff}}(\omega_c) \omega_c^2 = \epsilon_0 q_c^2 \), where \( \omega_c = 2\pi/t_c \) and \( q_c = 2\pi/L_c \). Flux motion in a dirty superconductor is usually overdamped, so that the factor \( \eta/(M\omega_c) \) is expected to be much bigger than unity. The mass enhancement factor can also be written as \( (1 + 2/[(1 + v)^{1/2} - 1]) \), with \( v = 4M\epsilon_0 q_c^2/\eta^2 \). The parameter \( v \) can be expressed in terms of the ratio \( J_c/J_0 \) \((\sim 10^{-2})\) of the critical and the depairing current, of the normal-state resistivity \( \rho_n \) \((-100 \mu\Omega cm)\), the coherence length \( \xi \) \((\sim 15 \AA)\) and the Fermi wave vector \( k_F \) \((\sim 0.5 \AA)\). The values of the parameters given in parenthesis are appropriate for high-\( T_c \) superconductors. One obtains

\[
v = (32/3\pi^4)\left(\frac{e^2 \rho_n \hbar \xi}{c^2} (k_F \xi)^4 \frac{J_c}{J_0}\right) = 0.1, \text{ i.e. } \frac{\eta}{(M\omega_c)} \sim 40.
\]

This result indicates that flux motion in these materials can be treated in the strong dissipation limit. In this case the tunneling time is given by \( t_c \approx L_c^2 \eta/\epsilon_1 \) and the effective action becomes

\[
S_{\text{eff}}/\hbar \approx \frac{\hbar}{e^2} \frac{\xi}{\rho_n} \left( \frac{J_0}{J_c} \right)^{1/2}.
\]

(II-43)

According to this result quantum tunneling is more likely to occur in superconductors with short coherence length \( \xi \) and high normal-state resistivity \( \rho_n \). Since \( \hbar/e^2 \) is equal to 4.1 k\( \Omega \) and \( J_c/J_0 \) is typically of the order of \(-10^{-2}\) in high-\( T_c \) superconductors, one can expect relaxation rates \((1/M_0)\mathrm{d}M/\mathrm{d}t \approx \hbar/S_{\text{eff}} \) of the order of 1 % due to quantum tunneling in the case of a ratio \( \rho_n/\xi \) of the order of 1 k\( \Omega \). Using the above values of \( \xi \) and \( \rho_n \), one obtains a ratio of \( \rho_n/\xi \approx 0.6 \) k\( \Omega \), showing that in principle the regime of quantum tunneling is observable in the high-\( T_c \) superconductors.

This model will now be extended to the case of larger applied fields, where collective pinning is effective on a flux bundle of volume \( V_c^b \) (denoted by \( V_c \) in the previous section). The crossover to this regime takes place when the pinning length \( L_c \) becomes comparable with \( a_{\Phi_c} \), i.e. when the interactions between vortices become relevant.
As before, the equilibrium solution is obtained by minimization of the free-energy functional described by Eq. (II-39). After substitution of \( M \) and \( \eta \) with the corresponding densities \( M/(a\phi_o)^2 \) and \( \eta/(a\phi_o)^2 \), the functional \( F \) takes the form described by Eq. (II-26) [with \( u(z,t) = u(r) \)]. Since in a static configuration the vortices adjust themselves to the pinning potential by shear and tilt deformations alone, the minimal solution of Eq. (II-39) corresponds (previous section) to the equilibrium between tilt \([C_{44}(\xi/Lc)^2]\), shear \([C_{66}(\xi/Rc)^2]\) and pinning \([\xi(W/Vc^b)^{1/2}]\) energy densities. This equilibrium situation defines the pinning volume \( V_c^b \). It was previously shown that for current densities \( J \leq J_c \) several \([\sim (C_{11}/C_{66})^{1/2}]\) bundles hop together in order to reduce the compression energy. The resulting superbundle is characterized by dimensions \( R_\perp, R_\parallel, \) and \( L \) bigger than \( a\phi_o \) (but \( < \lambda \)), where \( R_\perp \approx R_c, R_\parallel \approx (C_{11}/C_{66})^{1/2} R_\perp \approx L \approx L_c \). The critical current, however, is still given by the energy barrier of a single bundle \([U_c^b = \xi(WV_c^b)^{1/2}]\).

Since the tunneling process is characterized by several flux bundles tunneling simultaneously, it is clear that the tunneling time \( t_c^b \) is that of a single flux bundle with volume \( V_c^b \) tunneling under an energy barrier \( U_c^b \). In the limit of strong dissipation, one obtains \( t_c^b = R_\perp^2 \eta/(a\phi_o^2 C_{66}) \). The effective action for the tunneling of a superbundle is obtained by summing the contributions of every single flux bundle taking part in the tunneling process. This yields

\[
S_{E}^{\text{eff}} = t_c^b U_c^b \left( R_\parallel/R_\perp \right) \tag{II-44}
\]

where \( R_\parallel/R_\perp = (C_{11}/C_{66})^{1/2} \) is approximately equal to the number of flux bundles forming the superbundle. The corresponding tunneling rate is smaller than in the case of single-vortex tunneling. In the discussed current regime \( (J \leq J_c) \), an increasing magnetic field involves stronger interactions between flux lines; as a consequence the pinning volume \( V_c^b \) increases, the FLL becomes more rigid and the tunneling rate is eventually suppressed.

We will now describe an extension of this theory by Blatter and Geshkenbein for anisotropic superconductors \([28,11]\) in low or moderate magnetic fields (single-vortex pinning).
This model considers an anisotropic superconductor characterized by a mass anisotropy ratio \( \Gamma^{-1} = \epsilon^2 = m/M < 1 \), where \( m \) and \( M \) denote the effective masses in the directions parallel and perpendicular to the superconducting layers (ab plane), respectively. One defines a coordinate system, where the z-axis is parallel to the c-axis of the superconductor and the magnetic field lies in the yz-plane. The applied magnetic field is tilted by an angle \( \theta \) with respect to the superconducting layers. A second coordinate system is rotated around the x-axis (x = x'), so that the z'-axis coincides with the direction of the applied field.

The Lagrangian determining the dynamics of the vortex is

\[
L[u] = \frac{1}{2} \int dz' \left[ M^2(\theta)(\partial_t u_x)^2 + M^2(\theta)(\partial_t u_y)^2 \right] - F[u] \tag{II-45}
\]

where the free-energy functional \( F[u] \) is given by

\[
F[u] = \frac{1}{2} \int dz' \left[ \epsilon^2(\theta)(\partial_z u_x)^2 + \epsilon^2(\theta)(\partial_z u_y)^2 + 2U_{\text{pin}}(z', u) \right]. \tag{II-46}
\]

Here \( M^2(\theta) = \epsilon_0 M \) and \( \epsilon^2(\theta) = \epsilon_0^2/\epsilon_\theta \) denote the vortex mass and the elasticity of the vortex for the case of in-plane motion \([u = (u_x, 0, 0)]\). The corresponding quantities in the case of out-of-plane motion \([u = (0, u_y', 0)]\) are \( M^2(\theta) = M/\epsilon_\theta \) and \( \epsilon^2(\theta) = \epsilon_0^2/\epsilon_\theta^3 \). \( M \) is the effective mass for the case of a field parallel to the c-axis and \( \epsilon_0 \) is \( (\Phi_0/4\pi \lambda_{ab})^2 \) is the corresponding elasticity \( (\lambda_{ab} = \text{London penetration depth}) \). The anisotropy parameter \( \epsilon_\theta \) has the form \( \epsilon_\theta^2 = \epsilon^2 \cos^2 \theta + \sin^2 \theta \), while the characteristic lengths of the pinning potential \( U_{\text{pin}} \) are \( \zeta \) and \( \epsilon_\theta \xi \) for in-plane and out-of-plane motion, respectively. The mean individual pinning force \( f \) of one defect is the relevant one for in-plane motion, while the enhanced force \( f/\epsilon_\theta \) is effective in the case of out-of-plane motion.

The action of the tunneling process is given by \( S_E = \int dt L_E \), where the free-energy term \( F[u] \) is added to the kinetic energy term in \( L_E \). The effective action is obtained similarly to the isotropic case, i.e. by addition of a term containing the viscous drag coefficients. In the case of Ohmic dissipation the latters are given by \( \eta^2(\theta) = \epsilon_\theta \eta \) and \( \eta^2(\theta) = \eta/\epsilon_\theta \), with \( \eta = \Phi_0/2(2\pi \xi^2 \rho_n) \), where \( \rho_n \) is the planar normal resistivity extrapolated at \( T \to 0 \) and \( \xi \) is the coherence length in the layers. The solution for the effective Euclidean action is found again with the method of the dimensional estimates.
The first step consists in solving the classical problem of flux-creep in anisotropic superconductors by minimization of the free-energy functional $F$. The minimal solution corresponds to the balance of the kinetic and the pinning energies inside the collective pinning volume $V_c = L_c^2 \xi^2 e^\theta$. Inside this volume the pinning forces add up only with the square root of the number of pins, because of the competition between elastic and pinning energy. It turns out that the pinning length $L_c$ and the energy $U_c$ do not depend on the direction of motion of the flux lines.

For a magnetic field aligned to the c-axis of the specimen, one obtains $L_c = (\varepsilon_0^2 \epsilon^4 / f^2 n)^{1/3}$ and $U_c = (f^2 n \xi^2 L_c)^{1/2} \xi$, where $n$ is the density of pins. It is noteworthy that $L_c$ can also be expressed as $L_c = \varepsilon \xi (J_o/J_c)^{1/2}$. For a field enclosing an angle $\vartheta > m/M$ with the layers, the collective pinning length is $L_c = L_c / \varepsilon^\theta$, while the collective pinning energy does not depend on the angle $\vartheta$ ($U_c \approx U_c$).

In the case of layered superconductors in a field tilted by a small angle $\vartheta < m/M$ the vortex lines cannot be considered rectilinear objects [28]. They can rather be represented as chains of 2D "pancake" vortices connected by interplanar Josephson-like vortices. Within this ($\vartheta < m/M$) regime several "subregimes" of pinning and creep can occur depending on the value of the angle $\vartheta$.

From now on we will restrict to the case of applied fields tilted by "large" angles $\vartheta > m/M$ with respect to the superconducting layers. In this case the results for anisotropic and layered superconductors are the same [28].

As a second step one has to estimate the tunneling time of the process. This is done by equating the kinetic energy of the tunneling process and the elastic energy involved in the relaxation. The effective action of the process is approximately given by $S_E^{\text{eff}} \approx t_c U_c$ and is independent of the angle $\vartheta$. In the two limits of zero and strong Ohmic damping, one obtains, respectively:

$$S_E \approx \varepsilon \xi^2 (\varepsilon_0 M)^{1/2}$$  \hspace{1cm} (\Pi-47)

$$S_E^{\text{eff}} / \hbar \approx \frac{\hbar}{2 \rho_n} \frac{\varepsilon_0 \xi}{J_c} \left( \frac{J_c}{J_0} \right)^{1/2}$$  \hspace{1cm} (\Pi-48)
A comparison of Eq. (II-48) with Eq. (II-43) shows that the tunneling rate in the anisotropic (or layered) case is enhanced by a factor $1/e$ with respect to the isotropic one.

The previous results are valid in the limit $T \to 0$, while at finite temperatures the effective action is reduced. The temperature correction to the effective action was calculated by Grabert et al. [29] for the case of macroscopic quantum tunneling (MQT) in SQUIDs. They obtain a correction term that is determined by the kind of dissipation involved, but is only weakly dependent on the pinning potential (through the tunneling time). In the case of strong Ohmic damping the result is

$$S_E^{\text{eff}}(T) \approx S_E^{\text{eff}}(0) \left[ 1 - \left( \frac{k_B T_c}{\hbar} \right)^2 \right].$$  \hspace{1cm} (II-49)

The crossover between the flux-creep regimes of quantum tunneling and thermal activation takes place at the temperature $T_{qc}$, that can be estimated from the condition $\delta S_E^{\text{eff}}(T)/S_E^{\text{eff}}(0) \approx 1$. Keeping in mind that $S_E^{\text{eff}}(0) \approx t_c U_c$, it yields

$$T_{qc} \approx \frac{\hbar}{k_B t_c} \approx \frac{\hbar U_c}{k_B S_E^{\text{eff}}(0)}$$  \hspace{1cm} (II-50)

so that the crossover temperature is inversely proportional to the tunneling rate at $T = 0$ and thus to the tunneling time. For $k_B T/(\hbar/t_c) \ll 1$, i.e. $T \lesssim T_{qc}/3$, equation (II-49) can be expanded and one obtains

$$-\frac{\partial \ln M_{\text{rem}}}{\partial \ln t} \approx \frac{\hbar}{S_E^{\text{eff}}(T)} \approx \frac{\hbar}{S_E^{\text{eff}}(0)} \left[ 1 + \left( \frac{k_B T_c}{\hbar t_c} \right)^2 \right]$$ \hspace{1cm} (II-51)

so that the normalized relaxation rate should follow a $T^2$-dependence in this temperature region ($T \lesssim T_{qc}/3$).

### II.5 Vortex-Glass Model

This model proposed by Fisher [12] suggests the existence of a new thermodynamic phase in the mixed state of bulk disordered type-II superconductors. This phase is characterized by the absence of long-range...
order and by zero linear resistance. It is therefore argued that it represents a true superconducting phase. We will now proceed to outline the main features of this model, relying essentially on the description given by Fisher et al. in Refs. 12, 13, 14 and 15.

Fisher considers the isomorphism existing between the FLL in a bulk type-II superconductor in an applied field and a system of two-dimensional bosons at $T = 0$. This allows to relate the ground states of the boson system to the thermodynamical phases of the superconductor. When a magnetic field penetrates the superconductor the important degrees of freedom are represented by the flux lines that thread the sample. Representing the FLL as an array of 2D bosons ($T = 0$) allows to study the effect of disorder in the mixed state of a bulk type-II superconductor applying ideas and techniques from the domain of disordered superfluids.

For magnetic fields larger than $H_{c1}$ there is a competition between thermal fluctuations on one side and pinning disorder plus flux-line interactions on the other. The former tends to enhance the vortex-line density fluctuations, while the latters contribute to minimize such fluctuations. At high temperatures thermal fluctuations are dominating and will result in an unpinned vortex-liquid phase. This phase is likely to provide a good description of the high-$T_c$ superconductors in the range just below the $H_{c2}(T)$-line, but, due to large fluctuations, above the irreversibility line, below which pinning of the flux lines occurs. At low temperatures and low fields pinning disorder and flux-line interactions will dominate thermal effects, so that a pinned phase occurs. It is argued that this phase is a true superconducting phase with zero linear resistivity, that will be referred to as vortex-glass phase or vortex-glass superconductor. This phase is limited on one side by the boundary to the Meissner phase and on the other by the so-called $T_g(H)$-line (Fig. II.4).

The physical picture underlying the vortex-glass phase involves flux lines at low temperatures (in the mixed state) creeping through an environment of dense randomly located pinning centres. In absence of an applied current, "vortex-loop" excitations of size $L$ cost an energy, that increases as $\sim L^{\zeta}$ ($0 < \zeta \leq 1$). This scaling law can be used, as shown by Feigel'man et al. [4,6] within the framework of the collective creep
theory (§ II.3), to deduce the response of a single vortex line to an applied current. Vortex-loop excitations can be viewed roughly as a representation of the transverse displacement of a segment (length $L$) of a flux line. With an applied current, vortex-loop excitations of minimum size $L_{\text{min}}$ are no longer metastable, due to the energy gain from the Lorentz force. The nucleation of vortex-loop excitations requires passing over energy barriers $U(L_{\text{min}})$, which typically scale as a power in $L_{\text{min}}$. Since the minimum size $L_{\text{min}}$ is proportional to an inverse power of $J$, the energy barrier scales as $U(J) \sim (1/J)^\mu$, with $\mu \leq 1$.

Motion of flux lines will proceed via nucleation (and growth) of vortex-loop excitations of size $L_{\text{min}}$ at a rate proportional to $\sim e^{-U(J)/T}$. The resulting voltage is then given by

$$\frac{\partial J}{\partial t} \sim V(J) \sim \exp(- (Jc/J)^\mu) \ . \quad (\Pi-54)$$

It is clear that this state has zero linear resistance and in this sense it is a true superconducting state, in contrast with the picture delivered by conventional flux-creep theory. The value of $\mu (\leq 1)$ is obtained from the scaling law of the elastic energy of the flux lines, so that $\mu$ is a universal exponent characterizing the $T = 0$ vortex-glass ground state.
A metastable current (or magnetization) will decay via $\partial J/\partial t \sim V(J)$, so that integration of Eq. (II-54) gives in the asymptotic limit

$$J(t) = J_c \left[ \ln\left(\frac{t}{t_0}\right) \right]^{-1/\mu}, \quad \text{for } t \to \infty$$  \hspace{1cm} (II-55)

where $t_0$ is a microscopic time. At short times, i.e. $t \geq t_0$, the relaxation of the current is dominated by vortex-loop excitations of fixed size and by a fixed barrier height $U_0$, while in the asymptotic limit the size $L_{\text{min}}$ of the dominant loop excitations varies with the current and the relaxation follows an inverse power law of $\ln(t/t_0)$. It is possible to describe the whole relaxation process by the interpolation formula

$$J(t) = J_c \left( 1 + \frac{\mu k_BT}{U_0} \ln(t/t_0) \right)^{-1/\mu}.$$  \hspace{1cm} (II-56)

These results [Eqs. (II-55) and (II-56)] are analogous to the ones obtained within the collective flux-creep theory [Eqs. (II-33) and (II-35)], if one assumes that $\mu = \alpha$. At short times in comparison with the crossover time $t_{cr} = t_0 \exp(U_0/k_B T)$ the denominator of Eq. (II-56) can be expanded and one recovers Anderson's formula [Eq. (II-34)].

In practice, the experimental time observation window is restricted, so that the long-time limit $t \geq t_{cr} = t_0 \exp(U_0/k_B T)$ can be accessed only by increasing the temperature. This is realized for temperatures higher than $T^* := U_0/(k_B \cdot \ln(t_{obs}/t_0))$. For high-$T_c$ superconductors typical values of $U_0$ obtained at low temperatures are of the order of 20 meV $\sim$ 200 K. Taking $t_0 \sim 10^{-12}$ - $10^{-10}$ s and $t_{obs} \sim 10^2$ - $10^3$ s yields $\ln(t_{obs}/t_0) \sim 30$, and one obtains values of $T^*$ of the order $\sim$ 10 K. It is clear that in high-$T_c$ superconductors the long-time limit is realized already at moderate temperatures. In this regime the normalized decay rate $S = - \frac{d\ln J}{d\ln t}$ is given by

$$S = \left[ \mu \ln(t/t_0) \right]^{-1}.$$  \hspace{1cm} (II-57)

This result predicts a normalized relaxation rate dependent on the universal exponent $\mu$ and only logarithmically on the observation time and on $t_0$. Since the latters should not vary very much from system to system, Eq. (II-57) predicts an essentially constant relaxation rate above $T^*$. Below $T^*$ the normalized relaxation rate calculated from Eq. (II-56) increases linearly with temperature [Eq. (II-36), with $\mu = \alpha$].
The value of the relaxation rate $S$ for $T > T^*$ depends primarily on $\mu$, so that Eq. (II-57) assumes the role of a universal law for the normalized relaxation rate of the magnetization (or current). The corresponding value of $S$ can be estimated by taking $\mu = 1$, $t = 1000$ s and $t_0$ of the order of $10^{-10}$ s, and one obtains $S = 0.033$. Malozemoff and Fisher [13] have analyzed results on $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_7$ samples obtained by several groups and found that in several cases there is an intermediate temperature region characterized by an approximately constant relaxation rate $S$ of the order of 0.02-0.035. Nevertheless, this universal interpretation of the relaxation effects is hard to reconcile with the variety of relaxation data on high-$T_C$ superconductors published up to now.
III. EXPERIMENTAL ARRANGEMENT AND METHODS

The only experimental apparatus existing in our laboratory at the time of the discovery of high-$T_c$ superconductors was a He$^3$-He$^4$ dilution refrigerator with a SQUID measuring system. This experimental arrangement had actually been built for very sensitive magnetic measurements at very low temperatures and low fields [30,31]. It was designed to run for long periods of time (several months) at low temperatures. In this arrangement it is not possible to change the samples without warming up the whole machine to room temperature.

In order to gain some insight into the problem of magnetic relaxation in high-$T_c$ materials, we decided to develop another version of this measuring system using a SQUID sensor at constant temperature (4.2 K). This would allow us to test samples of different superconductors in a relatively short amount of time.

In the first two sections of this chapter we will describe the two experimental arrangements used throughout this work, i.e. the dilution refrigerator and the so-called 4K-Magnetometer. Since most of the data that will be presented here have been taken in the 4K-Magnetometer, we will describe the dilution refrigerator only very briefly. For a more complete description see [30,31,32]. The third section of the chapter describes the experimental procedures characterizing our zero-field-cooled (ZFC) measurements of the remanent magnetization and of its time relaxation. The origin of the background signals of the 4K-Magnetometer will be discussed in the fourth section. The results of extensive measurements of the background signals will also be presented and analysed. The fifth section contains a few remarks about the units used in this work. In section 6 of this chapter we outline some of the typical problems encountered in the process of fitting the theoretical predictions of the previous chapter to the experimental data.
III.1 Dilution Refrigerator

The cooling down of part of the specimens studied in this work was done by using a He³-He⁴ dilution refrigerator model DRI-420 by S.H.E. Corporation (now BTi, Biomagnetic Technologies Inc., 9727 Pacific Heights Blvd, San Diego, CA 92121). In this cryostat the temperature can be stabilised anywhere between 5 mK and 9 K. The measuring system is based on SQUID sensors and is very similar to the one that will be described in section III.2.3.

III.2 4K SQUID Magnetometer

III.2.1 Cryogenic Environment

The magnetometer can be inserted directly from room temperature into a liquid helium storage dewar [33] and be kept at constant temperature in the helium bath (4.2 K). The main advantage of this configuration is the ease of operation, because the measuring probe (Model SP Magnetometer SQUID Probe, S.H.E. Corporation [34]) with the sample can be extracted and reinserted in few minutes. This reduces the time needed for cooling down or warming up the specimens enormously. Moreover, in this configuration the specimens are directly immersed in liquid helium, which guarantees best thermal contact and temperature stability, since the vapour pressure inside the dewar undergoes but very small changes. On the other hand, all materials involved in the process, samples included, have to be able to withstand repeated thermal cycling (§ III.3) between 4.2 K and a temperature above the critical one of the sample (if necessary room temperature). A minor drawback of our arrangement is the fact that measuring probes (Fig. III.1) with an overall diameter exceeding about 18 mm cannot be inserted into this dewar.
III.2.2 Coil Arrangement

The coil arrangement consists of two coaxial cylindrical coils. Each one is wound on a non-magnetic support of Delrin. The coil supports are mounted one around the other and the inner one is attached to lower end of the SQUID probe (figure above).

The inner coil is the pick-up coil, wound as a second-order gradiometer with three groups of turns made by 3-6-3 windings [Fig. III.2,a)]. The two outer groups of windings are wound oppositely to the central one, so that in principle this device is sensitive only to the second spatial derivative of the magnetic field. The loops of the gradiometer are made of 0.13 mm thick superconducting NbTi wire (0.005" NbTi superconducting wire, S.H.E. Corporation) and have a diameter of about 6.3 mm.

Around the gradiometer is a second support (Fig. III.2,b) with the dc field coil, that consists of 8 layers of superconducting NbTi-Cu wire [35] for a total of 2859 (± 55) windings. This coil is 34.0 mm long and gives a...
maximum field (along its longitudinal axis) at the centre of $950 \pm 17$ Oe for a current of 1 A flowing through it ([36], § 2.2). The field coil is centred at the middle of the gradiometer (Fig. III.2), so that for an ideally homogeneous field distribution the total flux threading the gradiometer should be zero. In reality the field at the longitudinal axis of the coil is homogeneous only over a small range (1 mm, $\Delta H/H < 1\%$) around the middle. At a distance of about 10 mm from the centre, the field component along the longitudinal axis has already lost about 6.3 % of its maximum value [36]. This results (§ III.4) in a nonzero total flux through the gradiometer, even without sample.

The specimens are usually placed on a Delrin sampleholder, that can be screwed into the pick-up coil support from the bottom. This allows to put specimens exactly in the centre of the gradiometer by simply placing them
at the appropriate height in the sampleholder. This configuration has a
great flexibility, because sampleholders convenient to the shape and the
type of the specimen investigated (powdered, sintered, single crystal etc.)
can be produced in a reasonably short amount of time.

For instance studying the anisotropy of magnetic properties requires
measuring the same sample in different orientations to the applied mag¬
netic field. This can be a problem in the case of small single crystals, that
have to be handled with extreme care in order not to be cracked or bro¬
ken. In our case this was achieved by placing the crystal on a small glass
disk with some grease, so that the crystal would stick to the disk, even in a
vertical position. The sample can then be measured in the appropriate ori¬
entation by using sampleholders that would keep the glass disk (with spec¬
imen) in the desired position. In this arrangement the removal and change
of position of the specimen do not involve any further handling of the
crystal itself, once it has been placed on the disk.

The space available for the specimens [Fig. III.2,a)] is limited by the
inner diameter ($\theta = 5.0$ mm) of the pick-up coil support. Thus, the
overall dimension of the samples in a plane parallel to the loops of the
gradiometer cannot exceed about 4 mm. In the direction perpendicular to
the loops the maximum length of the specimen is determined by the dis¬
tance between the central and the outer coils of the gradiometer (for a
second order gradiometer). In an ideal configuration for maximum sensi¬
tivity the sample has to be placed in the centre of the gradiometer. The
length of the specimen should exceed the length of the central coil of the
gradiometer, in order to ensure an almost homogeneous flux distribution
through this coil. On the other hand, the flux distribution in the outer
coils of the gradiometer should not be affected by the presence of the
sample in the central one. This means that they should be far apart enough
from the centre of the pick-up coil. In our case [Fig. III.2,a)] this distance
is 10 mm and samples with a length up to 8-10 mm can be measured
without loss of sensitivity. Indeed, measurements on cylindrical samples
have shown that the pick-up coil signal is independent of the length of the
sample (at constant cross section) for specimens whose lengths range be¬
tween about 4 and 10 mm.
III.2.3 Measuring System

All the data contained in this work have been taken using SQUIDs as detectors. In a typical arrangement the SQUID is inductively coupled [37] to a superconducting flux transformer, that in our case consists of an input coil and a second-order gradiometer. The input coil is placed inside the niobium body of the SQUID sensor and is connected to two niobium screw terminals, where the self-built gradiometer is attached. The total inductance of the flux transformer is $L_{\text{tot}} = 2.63 \mu\text{H}$. With a sensitivity at the SQUID sensor input of $1.09 \times 10^{-7} \text{A}/\Phi_0$ (Test Report of SQUID sensor No. 271986-07), we obtain a value of the flux-transformer ratio ($\Delta\Phi_{\text{pick-up}}/\Delta\Phi_{\text{SQUID}}$) of 139.

Four copper wires (insulated) were put inside the stainless steel tubing of the measuring probe and then attached to copper terminals on a small plate (Fig. III.1) inside the housing of the SQUID sensor. The leads from the field coil were then soft-soldered to these terminals. On the room temperature end of the probe a connector was mounted on the stainless steel tubing just below the rf head [37]. The dc field coil is then energized by means of an external power supply ($\S$ III.3.1).

The flux changes through the gradiometer are detected by the SQUID and the connected electronics (S.H.E. Model 30 SQUID Control Unit). Any change of the field distribution (including the magnetization of the sample) inside the gradiometer induces a current that is fed to the SQUID via the flux transformer. The output signals are then amplified [37] by the SQUID control unit, that delivers an output voltage proportional to the change of magnetic flux through the SQUID. Three different amplifications ($x_1$, $x_{10}$, $x_{100}$) are possible and 1 Volt at the output of the SQUID control unit corresponds to 48.1, 4.81, 0.481 $\Phi_0$ at the SQUID, respectively ($\Phi_0 = h/2e = 2.068 \times 10^{-15} \text{Tm}^2 = 2.068 \times 10^{-7} \text{Gcm}^2$). However, there is a limit (slew rate) to the amount of flux per unit time that the SQUID’s electronic can measure. Thus, the sweep of the external field has to be performed at a convenient speed.

The possibility of measuring the flux variation in the SQUID in units of fluxquanta is given by the so-called digital flux counter ("DFC", also by S.H.E. Corporation, [38]). This device is connected to the SQUID control unit and gives a digital display of the flux changes in the SQUID directly in units of $\Phi_0$. The effective variation of magnetic flux through the
gradiometer is obtained by multiplying the displayed value by the flux transformer ratio ($\Delta \Phi_{\text{pick-up}}/\Delta \Phi_{\text{SQUID}}$).

In this work it was assumed that the loops of the gradiometer are close enough to the specimen to detect any flux variation taking place at its surface, anytime flux is expelled from (or penetrates into) the specimen. This fact was confirmed by several measurements on cylindrical samples made by Nb, Nb-Ti, Ta, Pb, Pb-In wires and polycrystalline Chevrel Phase. All samples were 10 mm long (Chevrel 7.2 mm long) and between 0.5 and 2 mm thick ($\phi_{\text{pick-up}} = 6.30$ mm). In all cases the measured initial slope $\chi_{dc}$ of the dc magnetization curve corresponds to about 80% of the theoretical value at the sample. This was calculated assuming ideal complete screening of the external field by the sample throughout the central coil of the gradiometer and no screening in the outer groups of turns (previous section).

It has to be said that the experimental arrangement does not include any magnetic shielding for the sample and the gradiometer. Only the SQUID sensor is shielded by a niobium cylinder [34] around the SQUID housing. It is clear that this configuration does not allow to use the total sensitivity of the sensor itself, because of external noise. This is the main reason for choosing a second-order gradiometer. The result is that the output signal of the SQUID control unit after cooling down the probe (with sample) to 4.2 K can be stabilized within 0.5 $\Phi_0$.

III.3 Experimental Procedures

Basically two kinds of data were taken with the 4K-Magnetometer: isothermal dc magnetization curves and time relaxation curves of the magnetization. Most of the results that will be presented in this work are obtained from such data. We therefore describe the corresponding experimental procedures and set-ups in some more detail now.

One important feature concerning the measurements is the general procedure used to prepare the samples before every measurement. In order to get reliable zero-field cooled data one needs to be sure that no flux is trapped inside the specimen. This is achieved by warming up the specimen well above its critical temperature. To do this one can lift the SQUID
probe above the liquid helium level and observe the triangle pattern [37] of the SQUID disappear, when the sensor becomes normal around 9.2 K. In the case of superconducting specimens with a much higher \( T_c \), one simply takes the SQUID probe out of the dewar and and keeps it a few minutes at room temperature, before reimmersing it (dry!) in the liquid helium.

The cooling down of the specimens takes place in the earth field (0.1-0.3 Oe), that does not give any significant contribution to the relaxation signal. This can be verified by checking the output signal of the SQUID control unit. Any measurable amount of trapped flux in the specimen (or in the field and secondary coils, § III.4) would give rise to a corresponding relaxation, that can be detected as function of time. This was not the case, even if the output signal of the SQUID could be stabilized only within certain limits, because of some noise (see previous section) in the system.

III.3.1 Isothermal dc Magnetization Curves

Isothermal dc magnetization curves were taken by energizing the field coil with a low-noise power supply and by measuring the corresponding flux change in the pick-up coil. The flux change is directly proportional to the magnetization of the sample. This kind of measurement was performed using a digital flux counter (previous section), that displays the measured flux variation directly in fluxquanta at the SQUID.

The magnetization curves are taken stepwise up to a maximum field value \( H_i \) and then back to zero. The whole process is controlled by a computer that reads the values of the current in the field coil and of the flux changes in the SQUID after every step. The current value is determined by measuring the voltage drop over a calibrated shunt resistor (1 \( \Omega \)) by means of a precision voltmeter (Keithley Model 181 Nanovoltmeter). The reading of the current value by the computer takes place through an interface (model GPIB IEEE-488). A data acquisition card (DT 2801 by Data Translation) connected to the BCD output of the digital flux counter enables the computer to read the value of the flux changes at the SQUID.

The magnetization curve is driven by a program that controls it by means of a set of parameters that regulate the current sweep. Latter is performed by applying a voltage to a small electric motor, that turns the
potentiometer of the power supply. By choosing appropriate voltages and
time intervals one can set the sweep rate and the sweeping time (or step-
width) differently in two ranges of the magnetization curve. The limit
between the two ranges, the maximum shunt voltage value and the sta-
bilization time after every step are also given as starting parameters to the
program. The details of the routines that build the computer program
have been given by Pollini [31].

In order to sweep the field and keep track of the corresponding flux
variation with a SQUID, one needs extremely low-noise power supplies.
Even if the magnetization curve is taken stepwise, every single sweep has
be run very smoothly. Any abrupt variation of the current in the field
coil induces flux changes that may exceed the slew rate of the SQUID
control unit (or of the DFC) and be measured only partially. We have
used two very low-noise non-commercial power supplies delivering cur-
rent values between 0 and 100 mA and between 0 and 500 mA, respec-
tively. The former enabled us to run very smooth magnetization curves,
while the latter has greater stability over long periods of time.

In the case of superconductors exhibiting moderate relaxation effects,
the measurement of a magnetization cycle of the sample up to a field $H_i$
allows us to determine the value of the remanent magnetization $M_{\text{rem}}(H_i)$
at the beginning of the relaxation measurement ($t \approx 1$ s). The measure-
ment of the time relaxation (next section) only gives the flux change with-
in a time interval ($1 \text{ s} \leq t \leq 10^5 \text{ s}$), but no absolute value of the flux at any
time. For technical reasons the two measurements are taken separately.

III.3.2 Decays of the Remanent Magnetization

The dynamics of the magnetic relaxation processes can be rather fast [39],
so that it is convenient to record the dc output of the SQUID control unit,
rather than the signal from the DFC. The former has a much bigger slew
rate (previous section) than the latter [27,30].

The specimens are cooled down to helium temperature in zero field
($\S$ II.3) and then the external field is swept up to a maximum value $H_i$.
After a stabilization time of a few seconds the corresponding value of
current in the field coil is read and the field is then swept down to zero.
The recording of the dc output of the SQUID control unit as function of
time is started just before the applied field reaches zero. The field sweep
lasts typically 50-60 s in both directions. At this speed, the field sweep is much too fast for the slew rate of the SQUID electronics. The measuring system starts working again when the external field is close to zero. This is the reason why this kind of measurement does not give any indication about the absolute value of the magnetic flux in the sample.

The SQUID output voltage reading by the computer takes place through the same interface that reads the BCD output of the DFC (DT 2801 by Data Translation). The time scale of the experiment is given by the clock of the computer. A program takes readings of the output voltage at appropriate times. During the first 22 s a reading is taken every 55 ms (~ 400 readings), while afterwards the program takes the points at time intervals, that would appear approximately constant on a logarithmic time scale. This means that the time interval $\Delta t$ between readings increases as function of time (for $t > 22$ s). In our case $\Delta t = (t - t_{\text{start}})/50$ and this gives $\log (t_{i+1}) - \log (t_i) = 0.0087$. The routine is also programmed to take an extra point every time the SQUID output voltage changes by more than 0.5 V. This is done in order to detect any sudden flux change due to external disturbances or anything else. During the measurement it is possible to check the relaxation curve (on a logarithmic time scale) by plotting the measured points on the screen any moment (after the first 90 s). The computer routine can also be stopped anytime (for more details, see [31]).

III.4 Background

As already mentioned (§ III.2.2), the non-ideally homogeneous field distribution in the field coil gives a nonzero total flux through the gradiometer. At low fields this signal increases linearly with the applied field. The magnitude of the linear slope can be roughly calculated by simply taking into account the different field values in the central and outer coils building the gradiometer. For an exact calculation one should actually take into account the radial dependence of the field component along the axis of the coil. Moreover, the field components off the axis are completely neglected. Measurements and calculations of the field at the longitudinal axis of the field coil [36] have shown that the field value diminishes by about
6.3 % over a distance of about 10 mm from the middle of the field coil. Knowing that the field coil is centred around the gradiometer (Fig. III.2), this corresponds to the distance between central and outer coils of the gradiometer. Assuming an ideal geometry for the gradiometer, it results in a slope of $3.23 \times 10^6 \Phi_0/\text{A}$. The measured value is about $2.66 \pm 0.03 \times 10^6 \Phi_0/\text{A}$, which is in good agreement with the theoretical estimate. Since this slope is only due to geometrical factors, its value did not vary much ($\pm 1\,\text{-}2\,\%$) during the different series of measurements.

At higher fields ($H > 100 \text{ Oe}$) there is also some flux that starts penetrating into the superconducting material (NbTi wires), that builds the gradiometer and field coil. Therefore, the total flux through the gradiometer (as function of field) starts deviating from a linear function. At the end of a magnetization cycle (even without sample) there is then a finite amount of flux trapped inside the gradiometer and the field coil, that will thereafter decay as function of time. For this reason extensive measurements of the remanent magnetization and of its time relaxation have been originally carried out (as function of the applied field) on the experimental apparatus without sample.

The results of the measurements of the background signals were also compared with data taken with pieces of the corresponding NbTi wire ($\phi = 0.13 \text{ mm}$, $0.005" \text{ NbTi wire, S.H.E. Corporation}$). The cylindrical wires used as sample were measured with their axes perpendicular to the applied field ($D = 1/2$, $D =$ demagnetization factor). The experimental data in Fig. III.3 show very similar features for both the NbTi wires and the background. The weakest relaxations were measured for values of $H_i$ between 50 and 100 Oe in both cases. The values of the logarithmic slopes $|\partial M_{\text{rem}}/\partial \log t|$ of the decay curves increase approximately as $H_i^3$ up to $H_i \approx 800 \text{ Oe}$. At higher fields, $|\partial M_{\text{rem}}/\partial \log t|$ increases less strongly and eventually saturates. It is interesting to notice that measurements [36] on wires of the same type ($0.005" \text{ NbTi, S.H.E.}$) oriented parallel to the field do not show any relaxation for applied fields $H_i$ smaller than 200 Oe. It is then clear that the relaxations measured below 200 Oe are actually due to demagnetization effects, since the loops (wires) forming the gradiometer and the field coil are perpendicular to the field ($D = 0.5$).

For $H_i > 200 \text{ Oe}$ the normalized decay rates $|M_{\text{rem}}^{-1} \cdot (\partial M_{\text{rem}}/\partial \log t)|$
Fig. III.3 Magnetization data at 4.2 K of NbTi wires (full symbols) and background of the 4K-Magnetometer (open symbols) as function of the applied field \( H_i \). Triangles correspond to the logarithmic slopes \( \delta M_{\text{rem}}/\delta \log t \) of the relaxation curves, whereas circles denote the remanent magnetization data. The units are \( \Phi_0 \) at the SQUID.

(see Fig. III.4) of the NbTi wires and the background are quite small (~ 5 x 10^{-3}) and practically independent of field. Below 200 Oe the values of the normalized rates are higher in both cases, probably due to the fact that the flux is trapped mainly at the surface and at the corners because of demagnetization effects. For the background data the effect is much more dramatic, essentially due to the very small values of \( M_{\text{rem}} \) measured in this field regime (Fig. III.3). Therefore, we will rely more on the data obtained at higher fields (\( H_i > 200 \) Oe).

Measurements [40] of the background signals were also performed with coil arrangements including the same gradiometer, but with superconducting field coils of different dimensions (same wire). The results have shown that the high-field region (\( H_i > 200-300 \) Oe) is dominated by the relaxation of the flux trapped inside the field coil wires.

Measurements repeated after different intervals of time (weeks) showed
that the background signals are reproducible within a few percents. The values of $|\partial M_{\text{rem}}/\partial \log t|$ and $M_{\text{rem}}$ measured with specimens thereafter have been corrected with average values of the corresponding quantities measured for the background.

In a later version of the same experimental set-up the problem of the background was actually solved. The linear slope of the background signal was reduced (by a factor $\sim 8$) by changing the diameter of the different groups of turns of the gradiometer. In order to get the total flux through the gradiometer closer to zero, it is enough to reduce the diameter of the loops in the central region of the gradiometer. This is due to the fact that the magnetic field is maximal at the centre of the field coil, i.e. at the centre of the gradiometer (Fig III.2). On the other hand, the amount of flux trapped by the gradiometer and the field coil depends mainly on the material used (superconducting wire) and on the quantity of it. This problem was solved by building a field coil with copper wire (Duramit,
that does not give almost any contribution to the background signals. As a result the irreversibility signal due to the coils was reduced by more than a factor of 10.

Nevertheless, this kind of coil (copper wire) has other disadvantages, like power dissipation. In our case this factor is not critical, because the currents used are small (< 1 A) and the constraints on the coil dimensions (§ III.2.2) result in a low resistance of the field coil ($R_{coil} \simeq 3\Omega$ at 4.2 K). All these factors and the fact that most of the measurements are made in zero-field (§ III.3.2) keep the boiling rate of liquid helium during measurements within reasonable limits.

All data presented in this work correspond to the net signals of the specimens, i.e. the background signals were either negligible or they have been discounted from the measured signals.

III.5 Remarks on Chosen Units

The magnetization $M$ given in the formulas corresponds to the physical quantity defined in the international unit system (SI). However, the quantity measured in our experiments is the magnetic flux change through the SQUID, which is only proportional to the magnetization of the sample. For this reason all the values of magnetization in this work are given in arbitrary units, that are actually fluxquanta ($\Phi_0$) at the SQUID.

All the values of the magnetic field $H$ given in this work are expressed in Oe and refer to the field value in the field coil, not at the specimen, i.e. the field values have not been corrected with demagnetization factors. If such a correction is made, it will be explicitly mentioned in the text.

Taking into account the choice of Oersted as unit for the field $H$ (and $dJ_c$), we have introduced a proportionality factor between the magnetization in arb. units and the calculated magnetization. This yields

$$M[\Phi_0] = K\cdot M[\text{Oe}] \quad (\text{III-1})$$

where $K$ is given in arb. units/Oe = $\Phi_0$ (SQUID)/Oe. The value of $K$ can actually be calculated, but it depends on some geometrical and technical factors. In the case of small samples with irregular shape (e.g. single
crystals) these factors are difficult to determine exactly [41]. As a result the value of $K$ can be calculated only with an accuracy of a factor two or three, so that we have decided to give the results of the magnetization measurements in arbitrary units.

We will now briefly describe how the factor $K$ can be estimated, because its value will be also obtained from the fits to the remanent magnetization data. The measured quantity is the magnetic flux $\Phi = B \cdot A$, so that the measured signal depends on the surface (cross section) $A_s$ of the specimen in the plane of the loops of the central coil of the gradiometer (§ III.2.2). Moreover, the output signal is proportional to the number of loops $N_{loop}$ of the gradiometer, that actually measure the flux change due to the sample, i.e. is dependent on the length of the sample. In the case of samples shorter than the central coil of the gradiometer, the factor $N_{loop}$ can only be estimated. This is due to the fact that a short superconducting sample causes an inhomogeneous deformation of the field distribution inside the pick-up coil, so that the corresponding flux change can only be estimated using $\Delta \Phi = B A_s N_{loops}$. Taking into account the flux-transformer ratio $(\Delta \Phi_{\text{pick-up}}/\Delta \Phi_{\text{SQUID}})$ (§ III.2.3) one obtains:

$$K = \frac{1}{(\Delta \Phi_{\text{pick-up}}/\Delta \Phi_{\text{SQUID}})} A_s [m^2] N_{\text{loop}} \times 2.068 \times 10^{-11} [\text{Oe.m}^2/\Phi_0]$$  (III-2)

It is clear that the factor $K$ will be different for every sample. For this reason values of $M$ given in the same arbitrary units for two different samples cannot be compared without taking into account the right proportionality factor.

### III.6 Experimental Methods

Purpose of this section is to discuss the typical problems encountered in the process of fitting some of the theoretical predictions of the previous chapter to the experimental data. We will first discuss some problems related to the time relaxation analysis and go on to the analysis of the remanent magnetization as function of field and temperature.
III.6.1 Analysis of Time Relaxation Data

Many relaxation curves of the remanent magnetization show deviations (cf. § VI) from a pure logarithmic law [Eq. (II-22)]. This results in a gradual saturation of the relaxation process towards an equilibrium value of the magnetization at long times. For such experimental data the logarithmic law can be used only as a "local" approximation in a restricted time window (e.g. Fig. IV.9).

In order to be able to make a comparison with relaxation data showing a logarithmic time dependence, we calculate the values of the logarithmic rate $\frac{dM_{\text{rem}}}{d\log t}$ at short times. This approximation is correct in view of Anderson's flux-creep theory [3], because (for $H_i > 2H^*$) the state at the beginning of the relaxation experiment ($t \approx 1\text{ s}$) is close to the critical state. This allows us to use Eq. (II-25) in order to calculate values of the activation energy $U_0$. We define the initial logarithmic slope $S$ of the relaxation curve in the following way: $S = \left. \frac{dM_{\text{rem}}(t)}{d\log t} \right|_{t = 1\text{ s}}$. Thus, $S$ corresponds to the maximum value of the logarithmic slope in the time window of the experiment ($1\text{ s} \leq t \leq 10^5\text{ s}$). All the values of $S$ given throughout this work refer to this quantity.

In order to characterize relaxation data showing non-logarithmic time dependences more precisely, we have tried to fit them with several relaxation laws. This kind of analysis yields some typical problems, that will be discussed now. As an example we consider a power-law time dependence of the kind

$$M_{\text{rem}}(t) = M_{\text{rem}}(\infty) + b(t/t_0)^{-\beta} \quad (\text{III-3})$$

where $b = M_{\text{rem}}(t_0) - M_{\text{rem}}(\infty)$, $M_{\text{rem}}(\infty)$ is the equilibrium value of the magnetization at $t \to \infty$ and $t_0$ corresponds roughly to the beginning of the relaxation measurement.

In our case $t_0$ is actually set equal 1 s, because some uncertainty in the determination of the time origin of the relaxation process hinders us from taking reliable relaxation data at $t < 1\text{ s}$. On the other hand, it is clear that a certain amount of relaxation takes place at times shorter than 1 s, but this amount is usually rather small in comparison to $b$, so that as a first approximation we will neglect it. Our choice of $t_0$ implies that the quantity $b = M_{\text{rem}}(1) - M_{\text{rem}}(\infty)$ gives the total amount of relaxation between
the beginning of the experiment at \( t = 1 \text{ s} \) and the time, when saturation occurs \( (t \to \infty) \). Furthermore, we assume that \( M_{\text{rem}}(t_0 = 1 \text{ s}) \approx M_{\text{rem}} \), where \( M_{\text{rem}} \) is the value of the remanent magnetization obtained from isothermal dc magnetization curves (§ III.3.1).

It is noteworthy that from the values of the fit parameters \( M_{\text{rem}}(1) \), \( M_{\text{rem}}(\infty) \), and \( \beta \) one can calculate the value of the initial logarithmic rate \( S \). By expanding Eq. (III-3) in the range \( t/t_0 \approx 1 \), one obtains that

\[
-\frac{\partial M_{\text{rem}}(t)}{\partial \log t} \bigg|_{t \approx t_0} = \beta \cdot b \cdot \ln 10.
\]

Our choice of \( t_0 (= 1 \text{ s}) \) implies that \( \beta \cdot b \cdot \ln 10 \approx S \).

The determination of the fit parameters \( \beta \), \( M_{\text{rem}}(1) \) and \( M_{\text{rem}}(\infty) \) was done by fitting the measured data (on linear scales) with a program (proFit 4.0 by Quantum Soft, P.O. Box 6613, 8023 Zurich). The function to be fitted \([:= f(x)]\) can be written in the program almost in the same form as given above, so that \( M_{\text{rem}}(1) \), \( M_{\text{rem}}(\infty) \) and \( \beta \) become the fitting parameters. A time shift \( \Delta t \) was added as an extra parameter, because of the uncertainty in the time origin. However, the values obtained for this parameter are small (< 1-2 s) and do not influence much the values of the other parameters. The program provides two fitting algorithms (Monte-Carlo and Levenberg-Marquardt, [42]) for minimizing the quantity \( \chi^2 \), that corresponds roughly to the average square deviation between the values of \( f(x_i) \) and the data points \( y(x_i) \). The program actually allows the user to interact with the fitting procedure in order to accelerate it. This can be done by defining an initial set of values of the parameters, estimated from the measured data. It is also possible to check the dependence of the parameters from each other by fitting only some of them and keeping the others fixed.

In our case, all the relaxation data showing appreciable deviations from a logarithmic law, were fitted using the method just described, which is actually quite precise. Nevertheless, the interpretation of the results (parameters) of the fit and of the corresponding statistical errors is not always trivial.

It should be borne in mind (§ III.2.3) that the values of magnetic flux measured during the time relaxation experiments are shifted by an arbitrary constant value. Thus, the values of \( M_{\text{rem}}(\infty) \) and \( M_{\text{rem}}(1) \) obtained as fit parameters are also arbitrary. We adjust them by setting
\[ M_{\text{rem}}(1) = M_{\text{rem}}, \] where \( M_{\text{rem}} \) denotes the value of the remanent magnetization determined from the isothermal magnetization curve.

### III.6.2 Analysis of Remanent Magnetization Data as Function of Field or Temperature

We first discuss the analysis of remanent magnetization data as function of the applied field \( H_i \). The values of \( M_{\text{rem}}(H_i) \) measured at constant temperature can be fitted using the functions described by Eqs. (II-6) to (II-9), that were obtained within the framework of the critical state model by Bean. \( H_{c1}, dJ_c \) and the proportionality factor \( K \) introduced in section III.5 are the fitting parameters. After inserting the factor \( K \) into Eqs. (II-6) to (II-9), it becomes clear that the parameters \( K \) and \( dJ_c \) can be determined independently of each other only in the field regime described by Eq. (II-8). Since in the case of small single crystals the value of \( K \) can be only roughly estimated (§ III.5), one can rely only on the data points corresponding to the field regime of Eq. (II-8) to determine its value. The determination of the other fit parameters is less critical, since they can be obtained from fits to the data in all the field regimes.

The analysis of remanent magnetization data as function of temperature (at constant applied field \( H_i \)) is very similar to one described above. The first step is to insert the factor \( K \) and the temperature dependent quantities \( H_{c1}(T) \) and \( J_c(T) \) in Eqs. (II-6) to (II-9). Typically, the functions \( H_{c1}(T) \) and \( J_c(T) \) contain the parameters \( H_{c1}(0), J_c(0) \) and \( T_c \), that (together with \( K \)) become the fitting parameters of the resulting functions \( M_{\text{rem}}(T) \) for the different field regimes. The determination of the fit parameters \( K \) and \( dJ_c(0) \) independently of each other involves the same problems discussed in the previous paragraph.
IV. MAGNETIC RELAXATION EFFECTS IN (BEDT-TTF)$_2$Cu(SCN)$_2$ SINGLE CRYSTALS

Measurements of the remanent magnetization and of its time relaxation as function of applied field $H_i$ and temperature were carried out on several single crystals of (BEDT-TTF)$_2$Cu(SCN)$_2$. The data at 4.2 K as function of field were taken in the 4K-Magnetometer (§ III.2) on two different crystals, while the data as function of temperature were measured on two other single crystals inside a dilution refrigerator (§ II.1). In both cases the two samples were put inside the corresponding experimental set-ups with different orientations to the applied field in order to study the anisotropy of some magnetic properties.

In the first section of this chapter, we will briefly describe some structural and physical properties of this organic superconductor. This section also contains a list of the measured specimens with their dimensions. The second section will deal with the field dependence of the remanent magnetization and of its time relaxation behaviour at 4.2 K. The temperature dependence of the same phenomena will be analysed in the third part of the chapter. This chapter also includes a short section (§ IV.4) with the results of the ac susceptibility measurements performed on both samples as function of temperature.

IV.1 Specimens

(BEDT-TTF)$_2$Cu(SCN)$_2$ is an organic superconductor with a transition temperature $T_c = 10.4$ K [43]. BEDT-TTF is an organic molecule with formula C$_{10}$S$_8$H$_8$ (bis(ethylenedithio)tetrathiafulvalene). The structure is monoclinic [43] with lattice parameters $a = 16.248$ Å, $b = 8.440$ Å, $c = 13.124$ Å, $\beta = 110.3^\circ$ at 298 K. The spacing between the bc-planes is given by $s = a \cdot \sin\beta = 15.2$ Å. The BEDT-TTF molecules form dimers, which place themselves almost perpendicularly to each other. The almost two-dimensional array forming the conducting layers of the material along the bc-plane comes about through short intra- and interdimers sulphur contacts. The conducting layers are sandwiched (along the $a^*$-axis)
by insulating sheets of Cu(SCN)$_2$ anions. Therefore, the resistivity is highly anisotropic with $\rho_{a^*}:\rho_b:\rho_c = 600:1:0.8$ and $\rho_b = 0.025-0.100$ $\Omega$cm at room temperature [44]. Due to the stacking pattern of the material, the crystals grow generally as thin platelets with the shape of elongated and distorted hexagons. The plane of the crystals corresponds to the $bc$-plane. Moreover, crystal growth takes place predominantly in the $b$-direction, so that the longest dimension of the platelet is usually parallel to the $b$-axis. Since all the organic superconductor samples measured in this work are single crystals of the same compound (BEDT-TTF)$_2$Cu(SCN)$_2$, we will refer to them simply as "crystal 1", "crystal 2", etc.
All specimens were provided by B. Hilti, Ciba-Geigy AG, Basel. They will be described in some more detail now.

Crystal 1
This sample is a thin platelet with rather smooth surfaces and dimensions of about $1.15 \times 0.74 \times 0.03$ mm$^3$. This single crystal was measured in an external field applied perpendicular to the bc-plane. Taking the ellipsoidal shape as approximation one gets for the demagnetization factor $D$ a value of about $0.95 \pm 0.02$. Unfortunately, due to the repeated thermal cycling procedure (§ III.3) between measurements, this crystal cracked into several pieces. This resulted in average in-plane dimensions of the pieces that are about 4-5 times smaller than the original one ($D \leq 0.85$).

Crystal 2
This single crystal has the shape of an elongated hexagon, where the longest dimension corresponds to the b-axis. It is also a very thin platelet and has dimensions of about $3.30 \times 1.55 \times 0.20$ mm$^3$. It was measured with a magnetic field applied parallel to the b-axis. The corresponding demagnetization factor $D$ calculated with the assumption of an ellipsoidal shape is about $0.10 \pm 0.01$. The crystal could withstand the measurements, but for a thin crack along the b-axis in the middle of the crystal.

Crystal 3
Same single crystal as crystal 2, but the applied field was perpendicular to the bc-plane. In this case the corresponding demagnetization factor (ellipsoidal approximation) is about $0.74 \pm 0.05$. Again, during the measurements several cracks developed in the crystal, so that finally there were 4-5 crystal pieces. Their average dimensions were about 50% of the original ones.

Crystal 4
Thin platelet with dimensions $2.10 \times 1.45 \times 0.15$ mm$^3$. Measured with fields $H_i$ applied perpendicularly to the bc-plane. Assuming an ellipsoidal shape, one obtains a demagnetization factor $D = 0.87 \pm 0.03$. 
Crystal 5
Single crystal with dimensions 1.75 x 0.63 x 0.19 mm$^3$. The measurements were performed with magnetic field parallel to the bc-plane. The corresponding demagnetization factor is $D = 0.21 \pm 0.03$.

IV.2 Magnetic Relaxation Effects of the Remanent Magnetization at 4.2 K

IV.2.1 Remanent Magnetization as Function of the Field $H_i$

Measurements of the remanent magnetization were carried out on crystals 1 and 3 with applied field perpendicular to the bc-plane. Crystal 2 was measured with the bc-plane parallel to the external field. Thus, we will first compare the results obtained for crystals 1 and 3. Thereafter, we will extend the comparison to crystal 2 in order to characterize anisotropy effects.

Measuring the remanent magnetization at a constant temperature corresponds (§ III.3.2) to running an isothermal dc magnetization curve up to a maximum field $H_i$ and then down to $H = 0$. From now on we will call $H_i$ the "applied field". Note that all the values of $H_i$, $dJ_c$ and $H_{c1}^*$ given in this work (in Oe) correspond to the field at the field coil (§ III.5). If demagnetization correction are made, they will be explicitly mentioned in the text.

A typical magnetization curve of crystal 3 at 4.2 K is shown in Fig. IV.2. Both crystals (1 and 3) show essentially the same features, i.e. a relatively short Meissner region followed by a broad maximum around 25 Oe and pronounced irreversibility. At higher fields (40-60 Oe) the magnetization value is almost independent of field. The return curve at high fields has about the same slope as the initial part of the magnetization curve, indicating a considerable amount of pinning by the sample.

The value of $H_{c1}^*$ ($H_{c1}^* = H_{c1}(1-D)$, $D =$ demagnetization factor) was determined from the first macroscopic deviation of the slope $-\partial M/\partial H$ of the magnetization curve from the initial value $-\partial M/\partial H\mid_{H \to 0}$ ([31], § 5.2). The values of $H_{c1}^*$ determined in this way correspond approximately to the
first penetration of flux in the sample on a macroscopic scale (cf. § VI). For both crystals (1 and 3) we get about the same value of \( H_{c1}^{*} \), namely 6.5 ± 0.5 Oe. Nevertheless, local demagnetization effects causing flux penetration in the corners at fields lower than \( H_{c1}^{*} \) are to be expected, especially in the case of single crystals. Indeed, in the case of the remanent magnetization data (Fig. IV.3) the first measurable points are obtained with fields \( H_{i} \) slightly lower than the obtained value of \( H_{c1}^{*} \).

The estimate of the demagnetization factors (ellipsoid approximation) for both samples is quite difficult, due to the irregular form of the crystals and to cracks (§ IV.1) occurring during the thermal cycling procedure. We obtain values of \( 1/(1-D) \) between 7 and 10, so that the value of \( H_{c1}^{\perp} \) at 4.2 K lies between 40 and 65 Oe for both crystals. A comparison with the value obtained by Tokumoto et al. [45] at 4.5 K, after correction with the estimated demagnetization factor of their specimen, gives satisfactory agreement (\( H_{c1}^{\perp} \approx 57 \) Oe).

The remanent magnetization data of crystals 1 and 3 measured at 4.2 K as function of the applied field \( H_{i} \) are quite similar (Fig. IV.3). The first data points (crystal 1) lie around 6 Oe and they are followed by a steep
Fig. IV.3 Remanent magnetization data at 4.2 K as function of the applied field $H_i$ for the field orientation $H_i \perp bc$ [crystal 1 (○) and 3 (⧫)]. The data points for the field orientation $H_i \parallel bc$-plane [crystal 2 (▲)] are also shown for comparison (see later).

increase of $M_{\text{rem}}$. At higher fields the remanent magnetization increases less strongly and saturation sets in.

According to the Bean model ([2], § II.1) for an infinite slab, the field where the saturation occurs, corresponds to $H_i = 2H^* = dJ_c$ ($d$= thickness of the slab). This model is actually valid for an infinite slab parallel to the applied field, but we assume (§ II.1) that it can be at least qualitatively applied to our configuration ($H \perp bc$-plane), if one takes into account the right geometry. The result of the fit (§ III.6.2) with the Bean model to the $M_{\text{rem}}(H_i)$ data of crystal 3 is represented by the solid line in Fig. IV.4. The line is given by the fits in the different field regimes [Eqs. (II-6) to (II-9)] with the following set of parameters: $H_{c1}^* = 6.8$ Oe, $dJ_c = 45$ Oe and $K = 1200$. Note that the calculated (§ III.5) value of $K$ ($K \approx 1650$, with $N_{\text{loop}} = 1$ and $A_s = 4.73$ mm$^2$) corresponds within 30 % to the one of the fit parameter.

In the case of single crystals the determination of $H_{c1}^*$ (for both field
Fig. IV.4  Remanent magnetization of crystal 3 as function of the applied field $H_i$ ($H_i \perp bc$). The solid line represents the fit obtained on the basis of the Bean critical state model.

orientations) as parameter of the fit to the $M_{rem}(H_i)$ data is very difficult because of local demagnetization effects, that are not taken into account by the Bean model. As a result of flux penetration in the corners at low fields ($H < H_{c1}$), one gets more trapped flux than is predicted by the model. Thus, the resulting value of $H_{c1}^*$ (from the fit) is usually smaller than the real one. Indeed, the method of determination of $H_{c1}^*$ based on the first deviation of the slope of the magnetization curve has proved to be more reliable and to provide realistic values of $H_{c1}^*$.

We can try an estimate of the critical current from the value of the fit parameter $dJ_c = 2H^*$. In this case the value of $J_c^{bc}$ (anisotropic component of $J_c$ in the bc-plane, assuming that $J_c^{bc} = J_c^{bc,*}$, cf. § II.2) at 4.2 K can be given only as an order of magnitude, because of the imprecise determination of $d$ (§ IV.1) and of the unfavourable geometry. For $H_i \perp bc$-plane $d$ corresponds roughly to the smallest in-plane dimension of the crystals. For crystals 1 and 3 the obtained values of $dJ_c$ are 32 and 45 Oe,
respectively. These data are qualitatively consistent with the theoretical predictions, since crystal 1 has smaller (in-plane) dimensions than crystal 3. The obtained [Eq. (II-16)] value of $J_c^{bc}(4.2 \text{ K})$ is about 300 A/cm$^2$ for both specimens.

We will now discuss remanent magnetization data obtained for crystal 2, which was measured in a magnetic field applied parallel to the bc-plane. The magnetization curve (next figure) looks quite different from the previous one (Fig. IV.2). The initial part is very steep and a narrow maximum is reached already around 6-7 Oe. At higher fields the magnetization becomes almost field independent. The shape of the curve shows some irreversibility, although less pronounced than in the other field orientation ($H_i \perp bc$-plane). Also the absolute values of the magnetization (in Oe, cf. § III.5) are very small, if compared to the corresponding values for the same crystal (§ IV.1) in the other field orientation. The value of magnetization at the maximum of the curve is about 50 times bigger for field perpendicular to the bc-plane than for field parallel to it.

An exact determination of $H_{ci}^*$ (same method as above) is not possible here, due to some scattering of the initial points of the magnetization

![Magnetization curve of crystal 3 (H || bc) at 4.2 K for $H_i = 68$ Oe.](image)

**Fig. IV.5** Magnetization curve of crystal 3 (H || bc) at 4.2 K for $H_i = 68$ Oe.
curve. An estimate of the value of $H_{c1}^{\|}$ gives an upper limit of about 0.5 Oe. From this value of $H_{c1}^{\|} \approx H_{cl}^{\|}$ ($D = 0.03$) and from the width of the magnetization curve one could expect that moderate values of applied field ($\sim 20$ Oe) would be enough to produce the fully critical state inside the sample. This is clearly confirmed by the remanent magnetization data of crystal 2 (Fig. IV.3), since the value of $M_{\text{rem}}$ is practically independent of the applied field $H_1$.

These data were fitted with Eqs. (II-8) and (II-9) for $M_{\text{rem}}$ (Bean model) and the obtained values of the fit parameters are: $dJ_c = 4.8$ Oe and $K = 78$. We determine the value of the critical current component perpendicular to the layers at 4.2 K using $M_{\text{rem}} = K \cdot (L_1/4) J_c^{a*}$, where $L_1$ is the transverse length of the specimen with respect to the field ($\S$ II.1). With $L_1 = 1.55$ mm, we obtain $J_c^{a*}(4.2 \, \text{K}) \approx 25$ A/cm$^2$.

In order to estimate the value of $J_c^{a*}$ more precisely, it is necessary to take into account the fact that in this case ($H \parallel$ bc-plane) the induced currents flow in both directions, perpendicular and parallel to the bc-plane. We will use the extended Bean model proposed by Gyorgy et al. [16] ($\S$ II.1) to calculate the anisotropic components of the critical current. The dependence of the current components in the bc-plane on the direction of the applied field (Lorentz force) will be neglected, i.e. we will assume that $J_c^{bc, bc} = J_c^{bc, a*} : = J_c^{bc}$. It turns out $[(J_c^{bc, bc})/(L_1 J_c^{a*})] < 1$ that we have to use Eq. (II-14), that corresponds to the case where the critical state is dominated by the in-plane component $J_c^{bc, bc}$ of the current. Using the assumption made above, we can substitute $J_c^{bc, bc}$ with the value of $J_c^{a*}$ obtained before. This is also reasonable in view of the fact that latter value probably overestimates [33] the effective value of $J_c^{bc, a*}$, because of the curvature of the flux lines in that orientation ($H_1 \perp$ bc). With $L_1 = 1.55$ mm and $K = 78$, we obtain $J_c^{a*}(4.2 \, \text{K}) \approx 18$ A/cm$^2$. The fact that the values of $J_c^{bc}$ and $J_c^{a*}$ differ by more than a factor of 10 at 4.2 K indicates a strong current anisotropy in this material.

The difference between the values of the lower critical field for the two field orientations is rather striking, since the two values differ by more than a factor of 100. A possible explanation for this situation has been given by Kes et al. [46], who proposed a model for highly anisotropic layered superconductors. They argue that, when the coupling between layers is very weak, the 3D superconducting behaviour breaks down
below a certain temperature $T_{\text{co}}$, so that a description in terms of 2D superconducting layers coupled by Josephson junctions is more appropriate. In this regime (2D) the superconducting order parameter is large within the bc-planes, but almost zero in-between. For fields parallel to the bc-planes screening is therefore very weak and the value of $H_{c1}$ is extremely small. In this case vortex cores can effectively fit between the layers. As a result of this situation the superconductor behaves as if it were magnetically "transparent" to fields parallel to the layers.

The crossover temperature $T_{\text{co}}$ between the 3D and the 2D regime was estimated by Kes et al. within the framework of the Lawrence-Doniach model [47]. They obtained (with $\xi_{bc^*} = \frac{\xi_{bc}}{\Gamma^{1/2}}$):

$$\Gamma(1 - t_{\text{co}}) = 2\left[\frac{\xi_{bc}(0)}{s}\right]^2$$

where $t_{\text{co}} = T_{\text{co}}/T_c$, $\xi_{bc}(0)$ is the GL coherence length in the bc-planes at $T = 0$, $\Gamma$ is the anisotropy parameter and $s$ is the layer spacing. The value of $\Gamma$ can be calculated using known values of $\xi$ or $\lambda$ [48,49,50] in the directions parallel and perpendicular to the bc-plane and one obtains a value of $\Gamma$ of the order of 400. With $s = 15.2$ Å and $\xi_{bc}(0) = 150$ Å one gets $t_{\text{co}} = 0.5$. From the midpoint of the 10% to 90% diamagnetic transition (§ IV.4) of our crystal we get $T_c = 9$ K, so that the value of $T_{\text{co}}$ is around 4.5 K. This could explain our results from the magnetization curves with $H \parallel \text{bc}$, that yield values of the magnetization and of $H_{c1}^{\text{ll}}$ that are anomalously small.

The idea that our organic samples behave as layered superconductors with rather weak interlayer coupling allows us to explain another peculiarity of the experimental data presented in this work. It is noteworthy that for crystal 2 the value of $K$ obtained from the fit ($K = 78$) is much smaller than the calculated one ($K = 700$, with $N_{\text{loop}} = 6$ and $A_s = 0.34$ mm$^2$). It will become clear throughout this work that a discrepancy of the order 10-15 between the calculated and the fitted value of $K$ always occurs in the case of single crystals (organic and Y-Ba-Cu-O) measured in a field applied parallel to the layers. Since the typical in-plane dimensions of the samples exceed the length of the central coil of the gradiometer (§ III.5) the estimate of the factor $N_{\text{loop}}$ cannot be very imprecise. Thus, the problem must lie in the estimate of the effective cross section $A_s$ of the sample in the plane of the loops of the gradiometer. Such a big
discrepancy can be explained by the fact that regions of depressed order parameter between the layers reduce the screening of the specimen drastically, when it is exposed to a magnetic field parallel to the layers. Kes et al. [46] have calculated that a dimensional crossover should take place also in the Y-Ba-Cu-O compounds at temperatures of the order $T_c/2$. However, up to now no experimental evidence for this crossover has been found.

It is also noteworthy that in the case of single crystals measured in a field perpendicular to the layers the calculated values of $K$ correspond well ($\pm 25\%$) to the ones obtained from the fits.

**IV.2.2 Time Relaxation of the Remanent Magnetization as Function of the Field $H_i$**

Measurements of the time relaxation of the remanent magnetization ($\S$ III.3.2) at 4.2 K were carried out as function of field on the same single crystals as in the previous section. So again, we will first compare the results for crystals 1 and 3 ($H \perp bc$-plane) and then extend the comparison to crystal 2 ($H \parallel bc$-plane).

The values of the initial logarithmic slope $S = |\partial M_{\text{rem}}(t)/\partial \log t|_{t = \infty} \approx 1$ $S$ ($\S$ II.6.2) of crystals 1 and 2 (Fig. IV.6) exhibit a field dependence very similar to the one shown in Fig. IV.3 by the $M_{\text{rem}}(H_i)$ data. The data points for $H_i \perp bc$ show that, after an initial steep increase, the logarithmic slope $S$ saturates. The fields (for crystal 1 and 3), where the saturation occurs, correspond well to the values of $dJ_c = 2H^*$ (32 and 45 Oe, respectively) determined from the fit to the remanent magnetization data.

The field dependence of the logarithmic slope $|\partial M_{\text{rem}}/\partial \log t|$ was calculated by Chaddah and Ravikumar [51] using the Anderson flux-creep theory and the Bean model. In the case of field-independent $J_c$, they obtain (neglecting $H_{c1}$) that $S(H_i)$ follows a parabolic field dependence up to applied fields $H_i$ equal to $dJ_c$. For fields $H_i$ bigger than $dJ_c$ the logarithmic slope $S$ saturates. The same result can be obtained from Eqs. (II-10) to (II-13) for $M_{\text{rem}}^{\text{out}}(H_i)$, the part of the remanent magnetization that decays outwards. It is then straightforward to see that $S$ has the same field dependence as $M_{\text{rem}}^{\text{out}}$.

The values of $S$ obtained from the relaxation curves of crystal 3 confirm this prediction. The solid line in Fig. IV.7 is a fit to the data
and namely crystals, both for the almost the same R of values ratio (Fig. IV.6). The IV.3 (Figs. 5 and M) of behaviour described otherwise previouly the from expected because could as samples both for saturate R of values the penetration field in complete of regime the in occurring increase initial an After time of decade per 0.18 about and 0.05 between lierv.8) Fig. H if field applied the offunction as 3 and 1 crystal for K4.2 at obtained R of values typical The

s[l^t/lM]om/3log=S/M^2=R as R rate of decay normalized initial the can be calculated. We M^2 JJ. to equal is Mrem and state critical fully measured in sample there is caused by Hc1 than bigger Hi field with taken data the for none calculation. This data was calculated then were M£m(Hi) of values the for parameter the of values same the Hc1) (neglecting obtained we Bean the by given M^(H0 of dependence field the using obtained below). (see comparison for shown also are)

Fig. IV.6 Values of the initial logarithmic slope S of the relaxation curves at 4.2 K for H1 ⊥ bc: crystals 1 (○) and crystal 3 (○). The data points for H1 ∥ bc [crystal 2 (▲)] are also shown for comparison (see below).

obtained using the field dependence of M^out_rem(Hi) given by the Bean model. We obtained (neglecting Hc1) the same values of the parameters as for the fit to the M^out_rem(Hi) data.

Values of M^out_rem(Hi) were then calculated from the values of Hi corresponding to the data points M^out_rem(Hi) and from the obtained fit parameters Hc1 and dJc. This calculation is not necessary for the data taken with fields Hi bigger than dJc, because in this regime the sample is in the fully critical state and M^out_rem is equal to M^out_rem. We calculate the initial normalized decay rate R as R = S/M^out_rem = (1/M^out_rem) ∂M^out_rem/∂log t t = 1 s. The typical values of R obtained at 4.2 K for crystal 1 and 3 as function of the applied field Hi (Fig. IV.8) lie between 0.05 and about 0.18 per decade of time. After an initial increase occurring in the regime of incomplete field penetration the values of R saturate for both samples as could be expected from the previously described behaviour of M^out_rem and S (Figs. IV.3 and IV.6). The saturation values of R are almost the same for both crystals, namely 0.14 and
0.16 per decade of time for crystal 1 and crystal 3, respectively. These values are quite high and point to very strong relaxation effects in this material. Since the values of $R$ are calculated from the initial logarithmic slope $S$ (§ III.6.1) of the time relaxation curves, they should be taken as an upper limit for the effective normalized relaxation rate. The fact that at higher fields the value of $R$ seems to increase will be discussed after the analysis of the time relaxation law.

From the values of the normalized logarithmic rate $R$ we can extract values of $U_0$, the average activation energy of the relaxation process. Combining Eq. (III-25) with our definition of $S$, we obtain:

$$\frac{dM_{\text{rem}}(t)}{dt} = S \frac{R}{\ln 10} = \frac{k_B T}{U_0}$$  \hspace{1cm} (IV-1)

At 4.2 K our relaxation data yield values of $U_0$ between 4 and 6 meV for both samples (in the saturation regime). The data points at low fields
yield higher values of $U_0$, namely around 15 meV. For comparison we estimate the value of the activation energy from the fit to the relaxation data in Fig. IV.7 as well. Equation (IV-1) shows that the proportionality factor between $S$ and $M_{\text{rem}}^\text{out}$ is $(k_BT/U_0)\ln 10$. Taking the same value of $K$ as for the fit to the $M_{\text{rem}}(H_i)$ data (§ IV.2.1), we obtain $U_0 \approx 6$ meV. A similar calculation with the data of crystal 1 gives a value of $U_0$ of 4.5 meV, but in this case the fit does not approximate the data points in the low-field region ($H_i < dJ_c$) very well. The difference between these two methods for determining $U_0$ is that in one case the calculation of $U_0$ is done for every single data point separately, whereas in the other case one fits the data points (Fig. IV.7) to the field dependence of $M_{\text{rem}}^\text{out}$. From the proportionality factor $K$ obtained from the fit one calculates a value of $U_0$, that corresponds to the whole set of data.

The obtained values of the activation energy confirm preliminary results by our group [52,53], already pointing to the occurrence of very
strong relaxation effects in this compound. Kuznetsov et al. [54] have measured (after ZFC down to 4.2 K) field-on relaxation data on a single crystal (H \perp bc-plane). In the fully critical state (H = 60 Oe) they obtain an average value of U_0 of 7.2 meV, in good agreement with our results.

The bigger values of U_0 obtained by us at low fields can be interpreted in different ways. Geshkenbein and Larkin [55] have explained the existence of a peak in the normalized relaxation rate as function of temperature by the existence of two types of pinning centres. They assume that inside the specimen there are a relatively small number of strong pinning centres (activation energy U_1) and a large distribution of much weaker pinning centres (U_2 \ll U_1). This assumption can also be used to explain the behaviour of the normalized decay rate R as function of field. For small values of the field H_i there are few flux lines trapped by the sample, so that they are essentially trapped by the strong pinning centres. For higher values of H_i much more flux lines are trapped by the sample, so that they are pinned predominantly by the weak pinning centres. It is clear that in this field regime the latters would dominate the relaxation of the remanent magnetization, yielding lower values of the activation energy. On the other hand, higher values of U_0 are obtained for values of H_i that correspond to incomplete penetration of the sample. This could also be an indication that in this material the strong pinning centres are mainly represented by surface defects.

However, up to now, the origin of the pinning mechanisms in this material is not clear and, to our knowledge, the only report so far on structure defects has been given by Ravy et al. [56]. They argue that pinning in this material might take place through a succession of stacking faults both in the orientation of the (BEDT-TTF)_2 dimers and in the sequence of the Cu(SCN)_2 anions.

We will now proceed to discuss the results of the relaxation measurements performed with applied field parallel to the bc-plane (crystal 2). In this case the logarithmic slope S is almost independent of the applied field H_i (Fig. IV.6). The corresponding value of R = S/\(M_{\text{rem}}^{\text{out}}\) is about 0.03 yielding a value of the activation energy U_0 of the order of 25-30 meV. This value is about 5 times larger than the one obtained for H_i \perp bc-plane (cf. Fig. IV.12).

This difference in the activation energy should reflect the typical
situation encountered in layered materials, where flux motion perpendicular to the layers is inhibited, due to the high value of the condensation energy inside the layers. In fact, a previous calculation (§ IV.2.1) has shown that at the temperature of our experiments (4.2 K) the samples may be regarded as 2D superconductors with very weak coupling between the layers (bc-planes). At $T < T_{\text{c0}}$ and in the case of applied fields $H_j$ parallel to the bc-plane, vortices penetrate first in the regions between the layers, where the condensation energy is smaller. In this situation flux motion through the layers is energetically very unfavourable. It is therefore likely that relaxation processes will take place mostly through motion of vortices in a direction parallel to the layers.

However, this kind of motion might cause further interaction between the flux lines, since almost all of them are trapped between the layers. A first estimate of the average distance $a_0$ between flux lines for crystal 2 in the fully critical state, assuming an homogeneous distribution (Abrikosov lattice) through the sample, yields $a_0 \approx 13 \, \mu m$. In reality this calculation should be done by taking into account essentially the regions between the superconducting layers. This would further reduce the obtained value, giving in the end $a_0 \lesssim 10 \, \mu m$. These values of $a_0$ are about the same order of magnitude as the value of $\lambda$ in this compound, that is [50] about 30 $\mu m$ for field penetration perpendicular to the layers. This comparison shows that at 4.2 K the interaction through flux lines is still weak.

In conclusion, the analysis of the relaxation data at 4.2 K on single crystals of this organic compound points to strong anisotropy effects. The magnitude of these effects is much bigger for the obtained values of critical current and lower critical field than for the relaxation rate of the remanent magnetization. The results indicate that at 4.2 K the organic superconductor (BEDT-TTF)$_2$Cu(SCN)$_2$ behaves essentially as a layered superconductor with weak interlayer coupling.

We will now present the results of the analysis of the time relaxation law of the remanent magnetization. The time relaxation curves measured with $H_j \parallel \text{bc}$ seem to follow the logarithmic law quite well, even though in this case the time window of our experiments was somewhat limited ($1 \, s \leq t \leq 1-2 \times 10^4 \, s$). Moreover, due to the weakness of the signals measured a precise analysis of these relaxation curves was not possible.
The time relaxation curve shown in Fig. IV.9 definitely shows that this is not the case of the data on crystal 3 (and 1). All the time relaxation curves measured at 4.2 K with $H_i \perp bc$-plane show to some extent the same features, i.e. none of them is strictly logarithmic in the whole time window of the experiments ($1 \leq t \leq 10^5$ s). For instance, for crystal 3 and an applied field $H_i = 98$ Oe (Fig. IV.9) a deviation from the logarithmic law is clearly visible and takes place already at short times. It is straightforward to see that the logarithmic law can be used only as a "local" approximation of the time relaxation curve in a restricted time window.

The relaxation curves measured with crystal 1 and 3 ($H_i \perp bc$) could be well fitted by a power law time dependence of the kind

$$M_{\text{rem}}(t) = M_{\text{rem}(\infty)} + b \left[ (t + \Delta t)/t_0 \right]^{-\beta} \quad \text{(IV-2)}$$
where \( b = M_{\text{rem}}(t_0) - M_{\text{rem}}(\infty) \), \( M_{\text{rem}}(\infty) \) is the equilibrium value of the magnetization at \( t \to \infty \) and \( t_0 \) corresponds roughly to the beginning of the relaxation measurement. Note that in our case \( t_0 \) was actually set equal 1 s and that the obtained values of the fit parameter \( \Delta t \) were always smaller than 1 s (§ III.6.1). With our choice of \( t_0 = 1 \) s the quantity \( b \) corresponds to the part of the remanent magnetization \( M_{\text{rem}}(1 \text{ s}) = M_{\text{rem}} \), that decays between the beginning of our relaxation experiments \( (t = 1 \text{ s}) \) and the time, when the saturation of the relaxation process sets in.

This time relaxation law \( [\Delta M(t) \propto (t/t_0)^\beta] \) was observed experimentally in spin-glass materials and was found to be an approximation for a limited time window of the stretched-exponential law [57,58]. Recently, the power law time dependence was also obtained by Feigel'man et al. [5] within the framework of the collective flux-creep theory for the limit of single-vortex creep (§ II.3).

The values of \( b \) as function of \( H_i \) for crystal 1 (Fig. IV.10) exhibit a behaviour very similar to the one shown by the \( M_{\text{rem}}(H_i) \) data. At low

![Graph showing the relationship between relaxation and applied field](image)

**Fig. IV.10** Values of the total amount of relaxation \( b = M_{\text{rem}}(\infty) - M_{\text{rem}}(1 \text{ s}) \) of crystal 1 (○) as function of the applied field \( H_i \) (\( H_i \perp \text{bc} \)) obtained at 4.2 K from the fit parameters \( M_{\text{rem}}(\infty) \) and \( M_{\text{rem}}(1 \text{ s}) \). The values of \( M_{\text{rem}} \) (●) measured with crystal 1 are also plotted for comparison.
fields $b$ increases steeply, then levels off to reach saturation and starts to fall down at higher fields ($H_i \geq 200 \text{ Oe}$). The fact that the field dependences of $b$ and $M_{\text{rem}}$ are quite similar is quite reasonable, since $b$ is essentially a fraction of $M_{\text{rem}}$. In order to discuss the behaviour of $b$ at higher fields it is convenient to analyse first the field dependence of another fit parameter, namely of $\beta$.

The field dependence of $\beta$ is similar to the ones showed in Fig. IV.10. The only difference takes place at high fields ($H_i \geq 200 \text{ Oe}$), where $\beta$ suddenly increases. The values of $\beta$ range from 0.02 at low fields, to about 0.12 in the saturation regime and they finally reach 0.27 around 500 Oe. The parameter $\beta$ describes the curvature of the $M_{\text{rem}}(t)$ curve on a log-lin scale, so that a large (typically $\sim 0.1$) value of $\beta$ implies deviations from the logarithmic law, that will occur already at very short times. Thus, the saturation of the relaxation process is reached at shorter times for bigger values of $\beta$. In the opposite limit ($\beta \leq 10^{-3}$) saturation is reached at very long times ($t \rightarrow \infty$) and the relaxation curve has an (approximately) logarithmic time dependence. Any increase of $\beta$ will therefore correspond to a decrease of $b$, and viceversa. The data on crystal 1 in the high-field

![Graph showing the field dependence of $\beta$ for different applied fields.](image)

**Fig. IV.11** Values of the fit parameter $\beta$ (power-law exponent) as function of the applied field $H_i$ ($H_i \perp bc$) for crystal 1 (○) and 3 (◊).
region \((H_i \geq 200 \text{ Oe})\) of Figs. IV.10 and IV.11 clearly demonstrate this effect. It is noteworthy that in this case the quantity \(S (= \beta \cdot b \cdot \ln 10)\) is essentially constant (Fig. IV.6).

From the values of \(\beta\) and \(b\) one can calculate the quantity \(\beta \cdot b \cdot \ln 10\), that should be equal to \(S\). A comparison shows a very good agreement between the values of these two quantities for crystal 3, since they differ by only 2-3\%. In the case of crystal 1 the difference is somewhat bigger, but still below 15-20\%.

The decrease of remanent magnetization at high fields (Fig. IV.10) may be understood in terms of dependence of the critical current on the applied field \(H_i\). In fact, one could expect that for fields \(H_i\) high enough the value of current induced during the magnetization cycle at \(H = H_i\) becomes essentially smaller than \(J_c\) (the value of current corresponding to the plateau of \(M_{\text{rem}}\)). In this case also the value of \(J_c\) (or \(M_{\text{rem}}\)) measured thereafter at \(H = 0\) would be smaller than \(J_c^*\). This is also confirmed by the decrease of the quantity \(b\), that is essentially a fraction of \(M_{\text{rem}}\). However, the fact that \(b\) seems to decrease faster than \(M_{\text{rem}}\) is somewhat surprising. In fact, for \(H_i > 2H^*\) the difference \(M_{\text{rem}} - b = M_{\text{rem}}(\infty)\) corresponds to the amount of pinning centres in the specimen that are strong enough to trap a flux line forever \((t \to \infty)\). One would then expect \(M_{\text{rem}}(\infty)\) to be essentially constant in the high-field regime, so that \(b\) should decrease with the same rate as the remanent magnetization. The reason for this discrepancy is not clear, but it should be borne in mind that the determination of \(b\) as a fit parameter is not trivial, since the saturation of the relaxation process takes place at very long times \((t \sim 10^{12}\text{ s})\) even in the case of large values \((\sim 0.10 - 0.15)\) of \(\beta\).

From the data points in Fig. IV.10 one can estimate the ratio \(b/M_{\text{rem}}\) and see that in the saturation regime \((H_i \geq 2H^*)\) it is between 0.5 and 0.7. The corresponding value for crystal 3 in the same field range is somewhat lower and lies around 0.5. The quantity \(b/M_{\text{rem}}\) corresponds to the fraction of the trapped flux \(M_{\text{rem}}\), that is expelled by the specimen during the relaxation process. The values given above indicate once again that the value of the effectively persistent current \([\sim M_{\text{rem}}(\infty)]\) in this material is much lower than the value of \(J_c\) determined from the remanent magnetization.
The fact that the power law time dependence was also obtained within the framework of the collective flux-creep theory (§ II.3) allows us to reinterpret some of the fit parameters. According to this theory, the power law approximation is justified when the parameter \( \alpha \) is much smaller than unity, i.e. in the case of single-vortex creep (\( \alpha = 1/7 \)). This situation takes place typically at low fields and low temperatures, when the average distance between flux lines is large (\( \sim \lambda \)), so that interactions between them are weak or negligible. The value of the distance \( a^c \) previously calculated (this section) demonstrates that this is the case of the specimen measured with \( H_i \parallel bc \), even in the fully critical state. An analogous calculation for crystal 1 and 3 yields values of \( a^c \sim 2-4 \mu m \) that are of the same order of magnitude as the penetration depth \( \lambda \) (\( \sim 1 \mu m \), [50]) for fields perpendicular to the layers.

Mathematically, the power law approximation of the relaxation law is justified in the case \( \alpha(k_BT/U_c)(\ln(t/t_0)) \ll 1 \). For single-vortex creep the predicted value of \( \alpha \) is 1/7, whereas the values of \( k_BT/U_c \) correspond to the determined values of \( R/\ln10 \), that are typically 0.06-0.08 in the saturation regime. Inserting values of \( t_0 \) between \( 10^{-12} \) and \( 10^{-4} \) s (§ II.3) and values of \( t \) between 1 and \( 10^5 \) s, we obtain \( \alpha(k_BT/U_c)(\ln(t/t_0)) \sim 0.1-0.6 \). This calculation shows that from a mathematical point of view the power law approximation is not always correct in the typical time window of our time relaxation experiments.

Recent results [41] obtained in our group have also shown that the power law function described by Eq. (IV-2) cannot be used in its original form. It turns out that the fit parameters \( t_0 \) and \( M_{rem}(\infty) [b = M_{rem}(t_0) - M_{rem}(\infty)] \) cannot be determined independently. Qualitatively, the same problems arise when one tries to fit the time dependence of the relaxation data with other laws, like the ones described by Eqs. (II-33) and (II-35).

For these reason, we have set \( t_0 = 1 \) s. Of course, this does not allow us to draw any conclusion on the microscopic parameter \( t_0 \), but at least we can obtain some information about the strength of the relaxation by means of the parameters \( \beta \) and \( M_{rem}(\infty) \). The latter is actually proportional to the persistent current. Using \( t_0 = 1 \) s and \( t \) between 1 s and \( 10^5 \) s, we get \( \alpha(k_BT/U_c)(\ln(t/t_0)) \leq 0.1 \), so that in this case the power law approximation is correct in the whole time window of our relaxation experiments. According to collective creep theory, the exponent \( \beta \) of the power law fit is equal to the ratio \( k_BT/U_c \) [Eq. (II-37)], where \( U_c \) is equal to \( U_0 \), as
defined in the Anderson's theory of flux-creep. Note that $U_c$ stands for the typical energy scale of the relaxation process for currents close to the critical value $J_c$ (§ II.3), due to the fact that a linear extrapolation of $U(J)$ from $J = J_c$ to zero current does not change the order of magnitude of $U(J)$. On the contrary, the power law dependence of $U(J)$ (at $J \ll J_c$) predicted by the collective creep theory can shift the typical energy scale $U_0$ [Eq. (II-33)] of the low-current regime to energy values much bigger than $U(J \approx J_c) \sim U_c$.

The values of $U_c$ obtained from the power law fit to the relaxation data of crystal 1 (Fig. IV.12) are close to the values of $U_0$ previously obtained. Note that the determination of $U_c$ relies only on the fit of the relaxation data, so that it is independent on the remanent magnetization data.
IV.3 Magnetic Relaxation Effects of the Remanent Magnetization as Function of Temperature Between 5 mK and 8 K

In this section of the chapter we will present the results of measurements of the remanent magnetization and of its time relaxation (§ III.3) for crystals 4 and 5, that were measured with magnetic fields $H_i$ oriented perpendicular and parallel to the bc-plane, respectively. The measurements were performed as function of temperature in the range $5 \text{ mK} < T < 8 \text{ K}$ at a constant value of applied field $H_i$. For the measurements with fields perpendicular to the bc-plane the value of $H_i$ was 63 Oe, while the value of the field $H_i$ in the other orientation ($H \parallel bc$) was 17 Oe. In this section we will refer to the data on crystal 4 and 5 using "$H_i \perp bc$" and "$H_i \parallel bc$" only.

In the first part of this section we discuss the results obtained from the measurements of magnetization curves (for both field orientations), i.e. the values of remanent magnetization, of the lower critical field and of the critical currents as function of temperature. In the second part we will analyse the temperature dependence of the time relaxation of the remanent magnetization. The third part of this section contains the results of ac susceptibility measurements performed as function of temperature on both samples.

IV.3.1 Remanent Magnetization, Lower Critical Fields and Critical Currents

Isothermal dc magnetization curves up to a field $H_i = 63$ Oe ($H \perp bc$) were measured at temperatures between 5 mK and 8 K. Below 1 K the magnetization curves show very small deviations from the Meissner line and, therefore, almost no irreversibility.

From the first macroscopic deviation (§ IV.2.1) of the slope $-\partial M/\partial H$ of the magnetization curve from the initial value $-\partial M/\partial H|_{H \rightarrow 0}$, we have determined [59] the values of the lower critical field $H^{*}_{c1}$ at different temperatures. The most striking feature displayed (Fig. IV.13) by the obtained values is the upward curvature of the $H^{*}_{c1}(T)$-curve and the "kink" in the data at $T = 4.3$ K. In order to make a quantitative analysis of the
results, we have to correct them with the demagnetization factor $D$ of the crystal. For crystal 4 ($H \perp bc$) we obtained (§ IV.1) $D = 0.87 \pm 0.03$, and this yields a correction factor $1/(1 - D)$ of the order of $8 \pm 2$. A quantitative comparison (after correction with the estimated demagnetization factor of their crystal) with the data of Tokumoto et al. [45] in the range between 4.5 and 8 K gives [60] a good agreement ($\pm 20\%$). The obtained values of $H_{c1}^\perp$ become rather large at low temperatures, so that, after correcting with the demagnetization factor of our sample, we get a value of $H_{c1}(0) = H_{c1}^\perp(0)/(1 - D)$ of $380 \pm 100$ Oe. A comparison of this value with other published data is not possible, due to the lack of other measurements in the low-temperature region. The value of the lower critical field $H_{c1}^\perp$ at 4.2 K is of the order of $70 \pm 20$ Oe, in satisfactory agreement with the values previously (§ IV.2.1) obtained for crystals 1 and 3.

The origin of the upward curvature of the $H_{c1}^\perp$ data is not clear. It is well known that the determination of the lower critical field is difficult, due to surface pinning effects that may become relevant in the low-
temperature range. However, our method ([31], § 5.2) for determining $H_c^+$ has proved to be less sensitive to such effects than other methods (e.g. first deviation of $M(H)$ from the Meissner line). Moreover, the same kind of temperature dependence has been observed by Oshima et al. [48] for the upper critical field $H_{c2}$ in both field orientations ($H \perp bc$ and $H \parallel bc$). In the case of magnetic fields parallel to the layers (bc-plane) this behaviour can be explained [46,47,61] in terms of a dimensional crossover between a 3D anisotropic superconductor and a 2D one with Josephson coupling between the layers. However, our results refer to the case of a magnetic field perpendicular to the layers, where the upper and lower critical fields depend on in-plane quantities, like $\xi_b$, $\xi_c$ and $\lambda_b$, $\lambda_c$, respectively. Thus, both critical fields should not be affected by the occurrence of a dimensional crossover.

Recent data by Uemura [62] show that the muon spin relaxation rate ($\sigma \sim 1/A^2$) of aligned single crystals of (BEDT-TTF)$_2$Cu(SCN)$_2$ increases linearly below $T \leq 0.6T_c$ ($H \perp bc$-plane, field cooled). This is in clear contrast with the behaviour observed for high-$T_c$ superconductors, that show [62,63,64] temperature dependences of $\sigma(T)$ in agreement with BCS weak-coupling theory or with the predictions of the two-fluid model. Recently, Le et al. [65] have performed similar measurements ($\mu$SR) on single crystals of another member of the BEDT-family, namely the organic superconductor $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br. In the temperature range $T \leq 0.4T_c$ ($T_c = 12$ K), after field cooling, they also find a linear temperature dependence of $\sigma(T)$ (for $H \perp bc$-plane). Moreover, in the case of zero-field cooling, the relaxation rate $\sigma(T)$ is strongly enhanced at temperatures $T$ below \sim 5 K. A comparison with the field-cooled relaxation rate at the lowest temperatures ($T \leq 1$ K) gives an enhancement factor of the order of 40.

NMR experiments by Takahashi et al. [66,67] on polycrystalline samples of (BEDT-TTF)$_2$Cu(SCN)$_2$ show that below 3.2 K the field-cooled NMR intensity differs from the zero-field-cooled one. Moreover, the $^1$H-NMR relaxation rate shows an anomalous temperature dependence with a peak around 3.2 K.

An anomalous enhancement of the $\mu$SR rate after zero-field cooling has also been reported (e.g. Sternlieb et al. [63]) for the high-$T_c$ compound $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ in the form of thin film, single crystal, crushed crystal
and sintered specimen, suggesting that it is an intrinsic property of this material. It was found that pinning is effective only at low temperatures, so that above a well defined depinning (or melting) temperature $T_D$ flux lines are able to move almost freely inside the sample. The temperature $T_D$ is a decreasing function of the applied field. These (and previous [68,69]) results on this compound have been interpreted in terms of FLL melting or depinning of flux lines. Recent results obtained by Kopelevich et al. [70] on single crystals of $(\text{BEDT-TTF})_2\text{Cu(SCN)}_2$ with the vibrating reed technique point to strong similarities of the magnetic behaviour of the organic and the high-$T_C$ compound (Bi-Sr-Ca-Cu-O). However, extrapolations of $T_D$ to the field range of our experiments yield values of the depinning temperature very close to $T_C$.

Summarizing, our $H^*_C(T)$-data, as well as $\mu$SR, NMR and vibrating reed magnetometry data by other groups, point to a sharp change in the regime of pinning at a temperature well below $T_C$.

A theoretical approach by Koyama, Takezawa, and Tachiki [71,72] extends the Lawrence-Doniach model [47] to the case of superconductors composed by stacks of normal and superconducting layers coupled by proximity effect and predicts an upward curvature of the lower critical field (for $H \perp$ layers). This is explained by the fact that the cost of condensation energy for a flux line penetrating the superconductor increases below a certain temperature, due to proximity-induced superconductivity between the layers. Originally, this model was proposed as an explanation for the anomalous enhancement of $H^*_C$ observed at low temperatures in several high-$T_C$ compounds (Refs. 1-7 in [72]).

The observation of an exponential temperature dependence of the “irreversibility line” in Bi-Pb-Sr-Ca-Cu-O at low temperatures was interpreted by De Rango et al. [73] in terms of temperature dependence of the breakdown field characterizing a proximity system. Deutscher et al. [74] have pointed out that in fact the small values of the condensation energy typical of high-$T_C$ superconductors, are actually due to the short coherence lengths $\xi_c$ [$\xi_c(0) < s$] in the c-direction only. They interpret the irreversibility line in terms of “breakdown” or “decoupling” fields, where the interlayer coupling (of the Josephson-type) becomes negligible. They argue that the idea of breakdown field proposed by De Rango et al. can also be interpreted in terms of decoupling of the superconducting layers. In
Fig. IV.14 Values of $H_{c1}^\perp$ ($H_1 \perp bc$) as function of temperature determined from the first macroscopic deviation of the slope $-\partial M/\partial H$ of the magnetization curves. The solid line is a fit to the data of the type:

$$H_{c1}^\perp(T) = H_0 \exp(-aT),$$

with $a \approx 0.5 \, K^{-1}$ and $H_0 \approx 700 \, Oe$.

In that case the coupling between the layers is provided by proximity effects. It is noteworthy that our $H_{c1}^\perp$ data follow an exponential temperature dependence over a broad temperature range ($2 \, K \lesssim T \lesssim 7 \, K$). The behaviour of $H_{c1}^\perp$ depicted in the above figure is very similar to the one obtained by Mota et al. [75,30] for the breakdown fields of Cu/NbTi and Ag/Nb wires. This temperature dependence is related to the fact that in a N-S proximity system [76] the probability of finding a Cooper pair at a distance $|x|$ from the N-S boundary is essentially proportional to $\exp(-K|x|)$, where the length $K^{-1}$ is given by $K^{-1} = \hbar v_F/(2\pi k_B T)$. Here $v_F$...
is the Fermi velocity of the normal metal. Therefore, the condensation energy due to proximity-induced superconductivity between the layers increases exponentially as the temperature is lowered. This would explain the measured temperature dependence of $H_{c1}$. Moreover, this confirms the idea (cf. § IV.2.2) that at 4.2 K (BEDT-TTF)$_2$Cu(SCN)$_2$ behaves as a stack of superconducting sheets with very weak coupling in-between. On the other hand, we notice that the temperature dependence of $H_{c2}$ obtained by Oshima et al. [48] cannot be directly explained by this approach.

The magnetization curves measured with $H \parallel bc$ ($H_i = 17$ Oe) as function of temperature confirm the general features shown by the magnetization data of crystal 2 at 4.2 K (§ IV.2). The Meissner region is very short even at the lowest temperatures ($T = 5$ mK) and the shape of the magnetization curves does not change much below 1 K. At higher temperatures the magnetization curves are very similar to the one shown in Fig. IV.5, but for the fact that the irreversibility gets smaller as $T$ approaches $T_c$ (cf. Ref. 45). The typical magnetization values (in Oe) are very small and point to weak screening effects. This is also confirmed by the analysis of the magnetization curves that yields values of $H_{c1}^\parallel$ lower than about 1 Oe at all temperatures. An exact determination of the values of $H_{c1}^\parallel$ was not possible, due to some scattering of the initial points of the magnetization curves. For this reason, we can only give an upper limit of about 0.5 Oe for the value of $H_{c1}^\parallel(0) \approx H_{c1}(0)$ ($D = 0.21$).

This estimate shows that the anisotropy of the lower critical field at low temperatures is extremely high. It results from the anomalous enhancement of $H_{c1}^\perp$ at low temperatures on one side and from the very small values of $H_{c1}^\parallel$ on the other.

The obtained value of $H_{c1}^\parallel$ points again (§ IV.2) to a description of the organic specimens in terms of two-dimensional superconductors. The fact that $H_{c1}^\parallel$ is very small indicates that the coupling between the layers is extremely weak. Moreover, we do not observe any increase of $H_{c1}^\perp$, even at temperatures as low as 5 mK. This is an indication that at 5 mK the increase of superconducting volume due to proximity effects is not large enough yet to recover the three dimensional superconducting behaviour. This would also imply that even the "proximity-enhanced"coherence length $\xi_c(T)$ is smaller than the layer spacing $s$ at low temperatures. In this case the upward curvature shown by the $H_{c2}(T)$-data [48] can be
explained in terms of the layered structure of the material ($\xi_c \leq s$). However, this result can be obtained within the frameworks of both the extension of the Lawrence-Doniach theory for layered superconductors (with Josephson coupling) by Klemm et al. [61] and the theory of superconducting multilayers (with coupling by proximity effect) by Tachiki and Takahashi [77].

Summarizing, our data for the lower critical fields indicate that (BEDT-TTF)$_2$Cu(SCN)$_2$ behaves as a strongly layered superconductor in most of the temperature range below $T_c$. The upward curvature of the $H_{c1}^L(T)$ curve can be explained in terms of proximity-induced superconductivity between the layers. However, further work is needed in order to clarify what kind of mechanism provides the coupling between the superconducting layers.

We will now present the remanent magnetization data. Again, we will start with data measured with $H_i \perp bc$-plane and then go over to data measured with $H_i \parallel bc$-plane. Some technical details of the analysis will be omitted, but they can be found in the diploma thesis of J. Robadey [59].

The remanent magnetization data measured with $H_i \perp bc$ ($H_i = 63$ Oe) on crystal 4 exhibit (Fig. IV.15) a sharp maximum around 3 K. Just above the maximum, $M_{\text{rem}}(T)$ decreases very sharply and then a sudden change of slope takes place. This is a further indication that below this temperature ($T \approx 4$ K) flux pinning is much more effective. Above this "kink", $M_{\text{rem}}(T)$ decreases smoothly up to highest temperature of our measurements ($T = 8$ K). One could suppose that this smooth decrease of the data represents the temperature dependence of $J_c(T)$, since the latter is directly proportional to $M_{\text{rem}}$ in the fully critical state [Eq. II.9].

It turns out [59] that the $M_{\text{rem}}(T)$ data above the kink ($T \geq 4.1$ K) can be very well fitted (§ III.6.2) by a temperature dependence of the kind

$$J_c(T) \propto J_c(0)(1 - T/T_c)^{3/2}.$$  \hspace{1cm} (IV-3)

Notice that the temperature dependence of the depairing current $J_0$ given by the Ginzburg-Landau theory ([78], p. 118, 149) is:

$$J_0(T) \propto H_c(T)/\lambda(T)$$ \hspace{1cm} (IV-4)

where $$H_c(T) = \frac{\sqrt{2} \kappa}{\ln \kappa} H_{c1}(T).$$ \hspace{1cm} (IV-5)
Neglecting the weak temperature dependence of the GL-parameter $\kappa$, one obtains

$$J_0(T) \propto H_{c1}(T)/\lambda(T). \quad (IV-6)$$

Assuming that $J_0$ has the same temperature dependence as $J_c$, Eq. (IV-6) implies that the temperature dependence of the critical current at high temperatures can be determined by fitting the temperature dependences of $H_{c1}(T)$ and $\lambda(T)$.

In our case the temperature dependence of $\lambda$ has been obtained by ac susceptibility measurements (§ IV.4). The ac data can be well fitted by a GL temperature dependence, i.e. $\lambda(T) \propto \lambda(0)(1 - T/T_c)^{-1/2}$, whereas other temperature dependences, like $\lambda(T) \propto \lambda(0)[1 - (T/T_c)^4]^{-1/2}$ (Gorter-Casimir) or $\lambda(T) \propto \lambda(0)[1 - (T/T_c)^2]^{-1/2}$ (Kanoda et al. [79]), do not give satisfactory results.

It is noteworthy that the $H_{c1}^{\perp}$ data above 4.3 K can also (Fig. IV.16) be fitted by a linear function of the type: $H_{c1}(T) \propto H_{c1}(0)(1 - T/T_c)$. This result, together with the GL temperature dependence obtained for $\lambda(T)$, indicates that in this material the depairing and the critical current follow
In order to extend the fit to the \( M_{\text{rem}} \) data to the low-temperature region, we need a temperature dependence valid in the whole range below \( T_c \). As a first approximation, we assume that the temperature dependence described by Eq. (IV-3) is valid in the whole temperature range. It turns out (figure above), that the \( H_{c1}^\perp \) data in the temperature region below the kink can also be fitted by a linear temperature dependence, but with different values of the parameters \( H_{c1}(0) \) and \( T_c \). In view of the fact that the \( M_{\text{rem}}(T) \)-data also yield a kink at about the same temperature (\( T \approx 4.1 \) K) (and that at this temperature the sample is in the fully critical state), it seems very likely that the kink is related to the temperature dependence of \( H_{c1}^\perp(T) \). We therefore expect the fits to the \( M_{\text{rem}}(T) \)-data below 4.1 K to yield different values of the parameters than above the latter temperature. From now on we denote with "’" and """" the values of the parameters \( [dJ_c(0), H_{c1}(0) \) and \( T_c] \) obtained from fits to the data below and above the
temperature of the kink, respectively.

From the lower critical field data \( (H_i \perp bc) \), we obtain that \( H_{c1}(T) = H_{c1}(0)(1 - T/T_c) \), where

\[
H_{c1}'(0) = 48 \text{ Oe} \quad \text{for } T \leq 4.3 \text{ K}
\]

\[
H_{c1}''(0) = 15 \text{ Oe} \quad \text{for } T \geq 4.3 \text{ K}
\]

The fits to the \( M_{\text{rem}}(T) \)-data were done (§ III.6.2) by inserting the temperature dependences of \( J_c(T) \) and \( H_{c1}(T) \) in the equations describing the four field regimes of the Bean model [Eqs. (II-6) to (II-9)]. The fits obtained (Fig. IV.17) follow the experimental data very well and are described by the following parameters:

\[
dJ_c'(0) = 290 \text{ Oe} \quad \text{for } T \leq 4.1 \text{ K}
\]

\[
H_{c1}'(0) = 49 \text{ Oe} \quad \text{for } T \leq 4.1 \text{ K}
\]

The four regimes characterizing the Bean model are separated by the temperatures 0.95 K, 2.8 K, and 3.7 K, respectively (see Fig. IV.17). The value of the fit parameter \( K \) (§ III.5) was found to be equal to 390 Oe/\( \Phi_0 \). This value is about 30% smaller than the value (\( K \approx 560 \text{ Oe}/\Phi_0 \)) calculated using \( N_{\text{loop}} = 1 \), \( (\Delta \Phi_{\text{pick-up}}/\Delta \Phi_{\text{SQUID}}) = 260 \) and \( A_s = 3.0 \text{ mm}^2 \).

The value of \( H_{c1}'(0) \) obtained from the fit to the \( M_{\text{rem}}(T) \)-data is 49 Oe, in very good agreement with the value (48 Oe) determined from magnetization curves measured at low temperatures. Also the values of \( T_c \) obtained with these two methods are in good agreement with each other and with the value (\( T_c \approx 9 \text{ K} \)) obtained from ac susceptibility measurements (§ IV.4).

In order to compare the value of \( J_c \) from the fit with the previously calculated ones, we calculated the value of \( dJ_c''(4.2 \text{ K})/4 = M_{\text{rem}}(4.2 \text{ K}) \).

We obtain that \( M_{\text{rem}}(4.2 \text{ K}) = (114/4)(1 - (4.2/9.07))^{3/2} \text{ Oe} = 11.2 \text{ Oe} \).

Using this value of \( M_{\text{rem}} \) and inserting \( L_\perp = 1.45 \text{ mm} \) and \( L_\| = 2.1 \text{ mm} \) into Eq. (II-16), we obtain \( J_c(4.2 \text{ K}) \approx 320 \text{ A/cm}^2 \), in very good agreement with the values of \( J_c \) previously calculated (§ IV.2.1) from the data on crystals 1 and 3.

The value of \( J_c \) at zero temperature can be estimated from the value of the parameter \( dJ_c(0) \), assuming that the fitted temperature dependence is valid also at low temperatures. In this way, we get \( J_c(0) \approx 2000 \text{ A/cm}^2 \). A comparison with literature values is possible only at high temperatures.
Fig. IV.17 Remanent magnetization data for $H_i \perp bc$ ($H_i = 63$ Oe) as function of temperature. The solid line is a fit to the data obtained on the basis of the critical state model by Bean in four different temperature regimes.

(above 4.2 K), where most of the magnetic measurements are performed. Nozawa et al. [80] calculated a value of $J_c$ of 1060 A/cm² from the width of an hysteresis loop at 4.9 K, so that our values of $J_c$ at 4.2 K are about a factor 3 smaller. It is noteworthy that in our case the same value of $J_c$ (at 4.2 K) was obtained from measurements on different single crystals (crystal 1, 3 and 4) in different experimental set-ups, so that it should be quite reliable.

The temperature dependence of the remanent magnetization data taken with $H_i \parallel bc$ is completely different from the one shown in Fig. IV.15. The behaviour depicted in Fig. IV.18 is typical of a sample in the fully critical state. The scattering of the points is due to the fact that the measured signal is very small. As a result, a precise fit of the temperature dependence of $M_{\text{rem}}(T)$ and hence of $dJ_c(T)$ was not possible. We found that the qualitatively best fit to the data is a parabolic temperature dependence $[dJ_c(T) \propto dJ_c(0) \cdot (1 - (T/T_c)^2)]$. In order to determine $K$ and
Fig. IV.18 Remanent magnetization $M_{\text{rem}}$ as function of temperature for $H_{\parallel} \parallel \text{bc-plane}$ ($H_{\parallel} = 17$ Oe). The data were fitted with a temperature dependence of the kind: $M_{\text{rem}}(T) = M_{\text{rem}}(0)\times[1 - (T/T_c)^2]$. The obtained values of the fit parameters are $M_{\text{rem}}(0) = 52.8$ arb. units and $T_c = 9.3$ K.

$\Delta J_c$ independently (cf. § III.6.2), we calculate the value of $K$ using $N_{\text{loop}} = 4$ and $A_S = 0.32$ mm$^2$ and we obtain $K \approx 310$. However, in this field orientation the value of $K$ obtained from the fits is always much lower than the calculated one (§ IV.2.1). Since this crystal (crystal 5, § IV.1) is actually a piece of crystal 2, previously measured in the 4K-Magnetometer, we assume that the ratio between calculated and fitted value of $K$ is the same for both specimens. In this way we get $K = 35$ and by fitting the data to the parabolic temperature dependence, we obtain $T_c = 9.3$ K and $\Delta J_c(0) \approx 6.05$ Oe.

A first estimate of $J^*_{c}$ at 4.2 K using the formula for the isotropic case yields $J^*_{c}(4.2$ K $) \approx 22$ A/cm$^2$. In this case (cf. crystal 2) we can use Eq. (II-15) by inserting an estimated value of $J_{c}^{\text{bc,bc}} (\approx 300$ A/cm$^2$). This way, we obtain a value of $J^*_{c}(4.2$ K $)$ of about 34 A/cm$^2$. A comparison with crystal 2 shows that the values of $J^*_{c}$ calculated with the formula for the isotropic case do not differ much ($J^*_{c} \approx 22$ and 25 A/cm$^2$). On the other hand, we observe that the correction obtained using the anisotropic
formula shifts the values of $J_c^*$ for the two specimens in different directions. Due to the slight difference in the factor $(L_{\perp}/t)$ of the two samples, the ratio $((J_{c}^{bc,bc}/L_{\perp}J_{c,cb}^{*})^{bc})$ is slightly smaller (bigger) than unity for crystal 5 (crystal 2). It is therefore not clear which critical current component (in-plane or perpendicular) dominates the critical state of these two specimens.

Summarizing, a value of $J_c^*$ of about 25 A/cm$^2$ should correspond to the typical value of the critical current across the planes at 4.2 K. The corresponding value at 0 K differs only by 20% $[J_c^*(0) \approx 30$ A/cm$^2]$. Thus, we obtain an anisotropy ratio of the critical current at 4.2 K of the order of 10. At low temperatures ($T \rightarrow 0$) the obtained values of the anisotropic critical current components $J_c^{bc}$ and $J_c^*$ are 2000 and 30 A/cm$^2$, respectively. This yields an anisotropy ratio $(J_c^{bc}/J_c^*)$ of the order of 50-100, but one has to keep in mind that the value of $J_c^{bc}(0)$ was calculated using the temperature dependence obtained from the fit in the temperature regime above 3.7 K. The large anisotropy effects in the critical currents give further evidence that the coupling between the superconducting sheets of (BEDT-TTF)$_2$Cu(SCN)$_2$ is extremely weak. The fact that the current anisotropy ratio becomes larger as the temperature is lowered is due essentially due to the increase of the in-plane current component $J_c^{bc}$. This can be explained in terms of an increase of the condensation energy due to proximity effects at low temperatures. As a result of these effects the superconducting volume becomes larger as superconductivity increases in the regions between the layers. However, at the lowest temperatures of our measurements (5 mK) the magnitude of this effect is probably not large enough to restore three-dimensional superconductivity. Otherwise, an increase of $M_{rem}$ (and of $H_{c1}$) at low temperatures should have occurred in the field orientation denoted by $H_i \parallel bc$.

It is noteworthy that, although the measured single crystals are quite thin ($t \approx 200 \mu$m), the strong anisotropy of $J_c$ does not permit (cf. § II.2) us to distinguish between the two in-plane current components $J_c^{bc,bc}$ and $J_c^{bc,a*}$. As already pointed out, the value of $J_c^{bc}$ previously determined ($H_i \perp bc$) should be taken as an upper limit for $J_c^{bc,a*}$, since it is very likely that in this field orientation the current component $J_c^{bc,bc}$ due to intrinsic pinning also gives a significant contribution to the remanent...
magnetization. In our case, the determination of latter current component through Eq. (II-14) requires a somewhat higher aspect ratio than the one of crystal 2 [and crystal 5, (L1/L2) ~ 8]. However, the qualitative validity of the concept of intrinsic pinning has been demonstrated by our relaxation data, that give strong evidence that the layered structure of the superconductor acts as pinning centre for fields H || bc.

IV.3.2 Time Relaxation of the Remanent Magnetization: Flux-Creep by Thermal Activation and Quantum Tunneling

This section will be divided essentially in three parts. In the first one we will present the results of the analysis of the relaxation curves in the whole temperature range (5 mK < T < 8 K) for both field orientations. The values of S, i.e. of the initial logarithmic slope of the relaxation curves (§ III.6.1), will be given together with the corresponding values of the normalized decay rate R = S/M_out rem. In the second part of this section we will deal with the data at low temperatures, where non-thermally activated flux-creep occurs. The data will be interpreted within the framework of the quantum collective creep model (QCC) for anisotropic (and layered) superconductors by Blatter et al. (§ II.4). In the third part we will analyse the high-temperature data (T \geq 1 K) in terms of thermally activated flux-creep. We will also present the results of the analysis of the relaxation law performed on the data measured with H || bc-plane, that show noticeable deviations from the logarithmic time dependence.

Time relaxation curves were measured at the same temperatures as for the remanent magnetization data presented in the previous section for both field orientations. For applied fields H || bc the logarithmic slope S shows (Fig. IV.19) a temperature dependence similar to the one of the remanent magnetization (Fig. IV.15). The main difference takes place in the region just above the maximum. Between 3 and 4 K the value of S drops by about 50 %, while the remanent magnetization decreases by about 35 %. Above 4 K ("kink" temperature) the remanent magnetization decreases smoothly due to the temperature dependence of the critical current Jc. On the other hand, the values of S decrease abruptly above the maximum, seem almost to saturate around 6-7 K and finally drop again
above 7 K. It is also noteworthy that the relaxation data in the low temperature region of the above figure (T < 1 K) show an almost constant finite value as it is the case for the remanent magnetization.

The values of $S$ from the relaxation measurements performed with $H_i \parallel bc$ show a completely different behaviour (Fig. IV.20). The temperature dependence is essentially linear up to 6-7 K, where a maximum is reached. At higher temperatures the value of $S$ diminishes. Also in this field orientation (cf. Fig. IV.19) the values of $S$ at low temperatures do not extrapolate to zero, but tend to a constant value.

In order to calculate the normalized relaxation rate $R = S/M_{\text{rem}}$, we have to determine $M_{\text{out}}(T) = f \cdot M_{\text{rem}}(T)$, the part of the remanent magnetization that decays outwards (§ II.2). This can be done using the values of the parameters obtained from the fits to the $M_{\text{rem}}(T)$-curves.

For $H_i \parallel bc$, the $M_{\text{rem}}(T)$-data show that the applied field ($H_i = 17$ Oe) was strong enough to produce the fully critical state in the sample at all
temperatures. This corresponds to a value of the fraction $f$ equal to 1. For $H_i \parallel bc$, it turns out that the difference between $M_\text{rem}^{\text{out}}$ and $M_\text{rem}$ is relevant only in the region of the peak, namely between 2 and 3.5 K. In this temperature range the value of $f$ is about 0.7-0.8. However, it should be pointed out that in this region the measurement of $M_\text{rem}$ and therefore the determination of $M_\text{rem}^{\text{out}}$ are not trivial, due to very strong relaxation effects. This may cause errors (underestimates) of $M_\text{rem}$ and $M_\text{rem}^{\text{out}}$ of the order of 20-30%.

The temperature dependences of the normalized relaxation rates $R = S/M_\text{rem}^{\text{out}}$ are strikingly different (Fig. IV.21) in the two field orientations. The data taken with $H_i \parallel bc$ yield values of $R$ that increase smoothly with temperature up to 8 K. The data measured with $H_i \perp bc$ show a much stronger temperature dependence leading to a first peak around 3 K and to a second one around 7.5 K. At all temperatures the values of $R$ for fields perpendicular to the bc-plane are bigger than the corresponding values of $R$ for the other field orientation. There is however a common
feature, that is, the fact that at low temperatures both rates extrapolate to a finite value of the order of 1%. This behaviour is in clear contrast with the results of the Kim-Anderson theory (§ II.2) of thermally activated flux-creep that yields a relaxation rate extrapolating linearly to zero for $T \to 0$. It will become clear later, that the behaviour of $R$ at low temperatures can be understood within the framework of the QCC theory presented in section II.4.

In order to better characterize the low-temperature relaxation data, we plot (Fig. IV.22) the values of $R/\ln 10 = -\partial \ln M_{\mathrm{rem}}/\partial \ln t$ using logarithmic scales. This shows the low-temperature saturation of the relaxation rate for both field orientations very clearly. Below 200-300 mK the values of $R/\ln 10$ are practically constant and of the order of $2 \times 10^{-3}$ ($6 \times 10^{-3}$) for $H_i \parallel bc$ ($H_i \perp bc$). Values of $\partial \ln M/\partial \ln t$ around 1% at millikelvin temperatures had already been obtained by our group [81,53] from relaxation data on powdered Ba-La-Cu-O.
Mitin [82] obtained a similar value of the normalized rate at $T = 0$ by extrapolating relaxation data measured above 0.5 K on Chevrel Phase specimens. He was the first to propose an interpretation of the experimental data based on the idea of quantum tunneling of vortices under energy barriers. Experimental data showing constant values of relaxation rate at millikelvin temperatures were also obtained by our group [83] for the heavy-fermion compound UPt$_3$. Recently, further evidence of quantum tunneling of vortices was given by Fruchter et al. [84]. They obtained a constant value of $|\frac{\partial \ln M}{\partial \ln t}|$ of about 0.6 % for a Y-Ba-Cu-O single crystal in a field of 0.2 T parallel to the c-axis and temperatures between 100 mK and about 1 K. Their results for a higher field (1.7 T) yield a larger value of the normalized relaxation rate ($\sim$ 0.012-0.014). A value of $|\frac{\partial \ln M}{\partial \ln t}|$ of the same order of magnitude ($\sim$ 1 %) was also obtained by Griessen et al. [85] from an extrapolation of data obtained above 1 K for a Y-Ba-Cu-O thin film.

The larger values of rate obtained by Fruchter et al. at higher field
values \((H = 1.7 \text{ T})\) have been interpreted in terms of curvature of the flux lines around the crystal. They argue that at low fields \((H \parallel c\text{-axis})\) the flux lines are much more curved around the platelet (single crystal). In this case (cf. § II.1) there is a contribution to flux-creep also from the part of the flux lines that is practically parallel to the basal plane of the crystal, i.e. parallel to the ab-plane. At higher fields, due to larger penetration in the sample, the flux lines are much less curved, so that one can assume that the decay measured in this configuration is due essentially to the flux lines parallel to the c-axis. Thus, the results of Fruchter et al. imply that non-thermally activated flux-creep is stronger for fields applied perpendicularly (with respect to parallel) to the superconducting planes. This is also confirmed by our results on the organic superconductor \((\text{BEDT-TTF})_2\text{Cu(SCN)}_2\) shown in Figs. IV.21 and IV.22.

All these results suggest that in several families of superconductors (high-\(T_c\), organic, heavy-fermion, Chevrel Phase) below a characteristic temperature flux-creep proceeds mainly by quantum tunneling of vortices.

We will now interpret our results within the framework of the quantum collective creep theory. It was already shown (§ IV.2) that at 4.2 K and in the range of applied fields of our experiments the average distance between flux lines is of the order of the penetration depth \(\lambda\) (or larger). The data points in Fig. IV.15 show that the value of the remanent magnetization \((H_i \perp bc)\) at 4.2 K is about 60 % of the maximum value of \(M_{\text{rem}}\) at the peak. This shows that in the whole temperature range the interactions between flux lines are negligible, so that flux-creep is most likely to take place through motion of single flux lines. This case was treated by Blatter et al. (§ II.4) for anisotropic 3D superconductors with an applied field perpendicular to the layers, so that as a first step we estimate [60] the value of the relaxation rate (for \(H_i \perp bc\)) due to quantum tunneling at \(T = 0\) using Eq. (II-48).

The main problem lies in the estimate of the ratio \((J_0/J_c)\). The values of \(J^c_0(0)\) obtained in the previous section are of the order of 2000 A/cm\(^2\), while the value of \(J_0\) is approximately equal to \(H_c/\lambda\). We estimate \(H_c\) to be of the order of 1000 Oe (cf. Sugano et al. [86], \(H_c(0) \approx 600\) Oe) and by taking \(\lambda_\parallel \approx 1.4 \mu\text{m}[50]\) we get \(J_0 \approx 6 \times 10^6\) A/cm\(^2\). This yields a value of \((J_0/J_c)^{1/2}\) of about 55.
From the resistivity data of Oshima et al. [48], we obtain a value of the in-plane normal-state resistivity $\rho_n$ of about 100 $\mu\Omega \cdot \text{cm}$. With $\varepsilon = 1/20$ and $\xi_{bc} \approx 150 \, \text{Å}$, we finally obtain $S_{\text{E}}^{\text{eff}}/\hbar \approx 170$. This corresponds to a relaxation rate $R/\ln 10 = -\partial \ln M_{\text{out}}/\partial \ln t = \hbar/S_{\text{E}}^{\text{eff}} \approx 6 \times 10^{-3}$, in very good agreement with our experimental results for $H_i \perp bc$ (cf. Fig. IV.22).

In this case the collective pinning length is $L_c^e \approx e \xi_{bc}(J_0/J_c)^{1/2} \approx 400 \, \text{Å}$, which corresponds to approximately 25 layer spacings.

In order to calculate the value of the relaxation rate for $H_i \parallel bc$, we should be able to estimate the “misalignment angle” $\vartheta$ between the applied field and the basal plane (layers) of our sample (crystal 5). In fact, for $\vartheta > \varepsilon$ the QCC theory for layered superconductors [28] predicts a behaviour equal to the one of anisotropic 3D superconductors, i.e. a relaxation rate independent of the angle $\vartheta$. For angles $\vartheta < \varepsilon$ the behaviour is completely different and the effective Euclidean action of the tunneling process is strongly dependent on the value of $\vartheta$, so that four regimes as function of $\vartheta$ (for $\vartheta < \varepsilon$) occur. In our case we estimated ($\S$ IV.2) the value of $\varepsilon$ to be $1/20$, which corresponds to an angle of $0.05 \, \text{Rad} = 2.9^\circ$.

Unfortunately, we cannot determine exactly to what degree the sample (crystal 5, $H_i \parallel bc$) was aligned to the field. However, the theoretical results (QCC) indicate that in three of the four angle regimes mentioned above the value of the effective action $S_{\text{E}}^{\text{eff}}$ is diminished in comparison to the case of a 3D anisotropic superconductor. In these three regimes vortices cannot be considered rectilinear object and one can view them as chains of 2D pancake vortices connected by interplanar Josephson vortices. Only in one regime at very small angles $[\vartheta < \varepsilon(d/L_c)^{1/2}(\xi/\Lambda)^3]$ the effective action is enhanced by a factor $(\Lambda/\xi)^2$ in comparison to the value calculated above for $H_i \perp bc$. The quantities $\Lambda = d/e$ and $d/2$ ($d = \text{layer spacing}$) denote the typical dimensions of the vortex core (Josephson type) in the directions parallel and perpendicular to the layers, respectively. These vortices are practically aligned to the $bc$-planes and are intrinsically pinned against motion through the layers, whereas pinning with respect to motion along the layers is rather weak. The Josephson pinning length $L_c^J$ is bigger by a factor $(\Lambda/\xi_{bc})^2$ than the pinning length $L_c^e/\varepsilon$, that one obtains in the case of a 3D anisotropic superconductor in the same field orientation. The corresponding pinning energy is $U_c^J \approx U_c^e$. By taking $d = 15.2 \, \text{Å}$ and $\varepsilon = 1/20$, we get $\Lambda \approx 300 \, \text{Å}$. With $\xi_{bc} \approx 150 \, \text{Å}$, we obtain a
value of the factor \((\Lambda/\xi_{bc})^2\) approximately equal to 4.

This means that the relaxation rate due to quantum tunneling at \(T = 0\) should be four times smaller for fields \(H_i \parallel bc\) than for \(H_i \perp bc\). The experimental results shown in the low-temperature region (\(T \leq 300\, \text{mK}\)) of Fig. IV.22 give a factor of 3 between the tunneling rates for the two field orientations, in satisfactory agreement with the theoretical prediction. This would imply that the alignment of the crystal to the field (\(H_i \parallel bc\)) was actually very precise, namely better than \(\varepsilon(d/L_c)^{1/2}(\xi/\Lambda)^3 \approx 0.0012\, \text{Rad} \approx 0.1^\circ\). In our case it is clear that the quantitative validity of latter statement cannot be verified.

We estimate the Josephson pinning length \(L_c^J = (L_c^\xi/\varepsilon) \cdot (\Lambda/\xi_{bc})^2\) using the aforementioned values of the parameters and obtain \(L_c^J \approx 30000\, \text{Å}\).

We conclude, that the predictions of the QCC-theory (for \(T \to 0\)) for layered superconductors agree well with the results of our relaxation experiments at very low temperatures. In this region flux-creep can be interpreted in terms of motion of segments of single vortices, that tunnel under the energy barriers represented by the pinning centres of the material. The latters act collectively on the flux lines. At finite temperatures the QCC theory predicts a correction term to the effective action. For the case of ohmic dissipation and for \(\eta(\omega)\) finite in the limit \(\omega \to 0\), the correction term is proportional to \(T^2\) [Eq. (II-51)]. This correction should be valid at temperatures below \(T_{qc}\), the crossover temperature between the regimes of flux-creep by quantum tunneling and by thermal activation [Eq. (II-50)]. Above \(T_{qc}\) the classical linear dependence in \(T\) of the relaxation rate is expected.

The measured relaxation data can be quite well fitted (Fig. IV.22) by a temperature dependence of the kind: \(A + B \cdot T^2\). One obtains:

\[
\frac{R}{\ln 10} = \frac{S}{(M_{rem}^\text{out} \cdot \ln 10)} \approx 5.9 \times 10^{-3} + 7.9 \times 10^{-3} \cdot T^2 \quad \text{for} \quad H_i \perp bc
\]

\[
\frac{R}{\ln 10} = \frac{S}{(M_{rem}^\text{out} \cdot \ln 10)} \approx 2.1 \times 10^{-3} + 5.2 \times 10^{-4} \cdot T^2 \quad \text{for} \quad H_i \parallel bc.
\]

The values of the tunneling time \(t_c\) can be calculated from the coefficients \(A\) and \(B\) using \(t_c = (1/k_B) \cdot [BS_{\text{eff}}^E(0) / \hbar]^{1/2}\), with \(S_{\text{eff}}^E(0) = \hbar/\Lambda\) [Eq. (IV-51)]. We obtain values of \(t_c\) of about \(2.8 \times 10^{-11}\, \text{s}\) and \(3.8 \times 10^{-12}\, \text{s}\) for \(H_i \perp bc\).
and $H_i \parallel bc$, respectively.

It is straightforward to see that our relaxation data (Fig. IV.22) do not show the expected crossover to a linear temperature dependence at higher temperatures, since the data points follow the $T^2$-dependence up to temperatures much higher than predicted by the theory $[(k_B T_{tc}/h)]^2 \ll 1$, i.e. $T \leq T_{qc}/3$, cf. II.4).

Summarizing, the QCC-theory for layered superconductors gives a satisfactory account of the relaxation behaviour of the remanent magnetization of the organic superconductor $(\text{BEDT-TTF})_2\text{Cu(SCN)}_2$ at low temperatures. The theory predicts (in the limit $T \to 0$) the right order of magnitude for the relaxation rates experimentally observed at the lowest measuring temperatures ($T = 5 \text{ mK}$). The dependence of the measured relaxation rates on the field orientation can be both qualitatively and quantitatively explained. The theoretical predictions concerning the region of higher temperatures ($R \propto T^2$) are qualitatively verified, although the crossover to a thermally activated regime of flux-creep is observed only partially (see below) in our samples.

We will now analyse the region of higher temperatures. It is noteworthy, that the relaxation data measured with $H_i \parallel bc$ between 1.5 and 5 K (Fig. IV.21) can also be quite well approximated by a linear temperature dependence extrapolating to zero. This is in agreement with the estimate of the crossover temperature from the coefficients of the $T^2$-fit to the relaxation data. In this way, we obtain a value of $T_{qc}$ of about 2 K. From the values of $R$ in Fig. IV.21 ($H_i \parallel bc, T \geq 1.5 \text{ K}$), we have calculated values of the activation energy $U_c$ according to the Kim-Anderson theory. As explained in section IV.2.2, the activation energy will be denoted by $U_c$, following the notation of the collective creep theory. In the temperature range between 1.5 and 5 K the activation energy (Fig. IV.23) has an almost constant value of $36 \pm 2 \text{ meV}$, confirming the values obtained at 4.2 K for crystal 2 (Fig. IV.12). Above 5 K the activation energy decreases sharply and is equal to about 20 meV around 7-8 K. This effect can be understood in terms of temperature dependence of the condensation energy that reduces the critical current and the pinning potential at higher temperatures. A comparison with the remanent magnetization data of Fig. IV.18 confirms this idea.
The relaxation data measured with $H_i \perp bc$ at temperatures $T \geq 1$ K yield an unusual temperature dependence of the normalized relaxation rate (Fig. IV.21). However, Geshkenbein and Larkin ([55], cf. § IV.2.2) have shown that a slight generalization of the Kim-Anderson theory for flux-creep is enough to explain the occurrence of a peak in the temperature dependence of the relaxation rate. They assumed that pinning inside the sample is characterized by two typical pinning energies $U_1$ and $U_2$, with $U_1 \gg U_2$. Moreover, the number of weak pinning centres (energy $U_2$) is supposed to be much larger than the number of strong pinning centres (energy $U_1$). At low temperatures the main contribution to flux-creep is given by the weak pinning centres and the relaxation rate increases with temperature. As the temperature increases the weak pinning centres are slowly “switched off”, until the relaxation process is “dominated” by the strong pinning centres, so that the value of the creep rate becomes smaller.

The relaxation data on crystal $4$ ($H_i \perp bc$) can be accounted for by this
model and are in agreement with the results by Zavaritsky and Zavaritsky [87,88,89] on Bi$_2$Sr$_2$CaCu$_2$O$_y$, TlBa$_2$CaCu$_2$O$_y$ and YBa$_2$Cu$_3$O$_7$ single crystals. For the first two compounds they find a temperature dependence of the creep rate with two peaks, whereas the relaxation rate of latter compound exhibits a single peak. The first two compounds also show sharp changes of slope of the $M_{\text{rem}}(T)$-curve at temperatures slightly above the peaks of the relaxation rate as function of temperature. This is also the case of our $M_{\text{rem}}(T)$-data for $H_i \perp bc$, that yield a kink at about 4.1 K. The left side of the peak in the rate $R$ shown in Fig. IV.21 corresponds to the region, where flux-creep is dominated by the weak pinning centres. The crossover temperature $T_J$ to the regime of creep characterized by the strong pinning centres can be estimated to lie approximately half-way between the peak and the following minimum of the creep rate, so that in our case $T_J$ is about 4 K. For the region of the peak the values of $U_2$ obtained from the Anderson formula range between 2 and 4 meV. The temperature region following the minimum of the rate $R$ should correspond to flux-creep determined mainly by the strong pinning centres. In this region we obtain values of $U_1$ of the order of 10 to 18 meV. This gives roughly a factor of 5 between the activation energies of the strong and the weak pinning centres.

Unfortunately, due to the fact that below 3.7 K the sample is not in the fully critical state (§ IV.3.1), it is not completely correct to compare the values of $U_C$ obtained at low and high temperatures. However, the low values of $U_C (= U_2)$ seem to indicate that the mechanisms quoted in section IV.3.2 as a possible origin of the enhancement of $M_{\text{rem}}$ and $H_{c1}$ (for $H_i \perp bc$) at low temperatures do not change the strength of the pinning centres substantially. The drastic change of temperature dependence observed at $T \approx 4$ K in our $M_{\text{rem}}(T)$ and $H_{c1}(T)$ data, as well as in $\mu$SR and NMR data [62,63,65,66,67], can be interpreted as a consequence of the increase of the condensation energy, i.e. of the superconducting volume, that takes place as the temperature is lowered. This phenomenon seems mainly to affect the number of pinning centres available inside the sample, but not their pinning strength.

We will now briefly outline the results of the analysis of the time relaxation law (cf. § III.6.1). As in the case of the relaxation data taken at 4 K, strong deviations (cf. Fig. IV.9) from the logarithmic time dependence
are observed only in the relaxation curves taken with \( H_i \perp bc \) and at higher temperatures [60]. We have tried to fit such data using the relaxation laws predicted by the theory of collective creep [Eqs. (II-33) and (II-35)]. It turns out (D. Risold, Diploma work [41]) that fits are possible for any value of \( \tau_0 \) between 1 s and \( 10^{-12} \) s and that the values of some of the fit parameters are dependent on \( \tau_0 \). It is noteworthy that, qualitatively, all the fitting curves obtained follow the data points quite well.

The use of a power-law time dependence [Eq. (IV-2)] also allows to fit the data with any value of \( \tau_0 \) between 1 s and \( 10^{-12} \) s, but in this case the values of the other fit parameters are independent of \( \tau_0 \) (Fig. IV.25). Thus, the value of the attempt time \( \tau_0 \) cannot be determined from this kind of fits to our data, so that again (cf. § IV.2.2) we use Eq. (IV-2) after setting \( \tau_0 = 1 \) s.
Fig. IV.25 Values of $k_B T/U_c$, obtained from fits to the relaxation curves based on Eq. (11-35), as function of the value of $\tau_0$: (○) $\tau_0 = 10^{-4}$ s, (●) $\tau_0 = 10^{-8}$ s, (◇) $\tau_0 = 10^{-12}$ s, (+) for any value of $\tau_0$ and fit based on Eq. (IV-2).

The relaxation curves measured with $H_i \perp bc$ at temperatures between 1.5 and 8 K are well fitted (cf. Fig. IV.9) by the power-law time dependence. The obtained values of the fit parameters $\beta$ and $b$ yield values of $\beta \cdot b \cdot \ln 10$ that correspond within few percents to the values of the initial logarithmic slopes $S$ shown in Fig. IV.19. The values of $\beta$ show a temperature dependence (Fig. IV.26) very similar to the one of the normalized relaxation rate $R$, but for the fact that the peaks are shifted to slightly higher values of temperature. A similar behaviour was also observed by Zavaritsky [89] in relaxation data on high-$T_c$ single crystals. In his case the measured relaxation curves ($10 \text{ s} < t < 6-7 \times 10^3 \text{ s}$) are essentially logarithmic in time, but he observes strong deviations from the logarithmic law in a short temperature range just above the peak of the rate $R$. This is the crossover region ($T = T_j$), where flux-creep is not ruled by a single type of energy barrier.

In our case the relaxation curves are logarithmic only below 1 K. Above 1 K the curvature $\beta$ of the decay curves increases with the creep...
rate $R$ and reaches a maximum around 3.5 K, i.e. just above the maximum of $R$. At higher temperatures the values of $\beta$ go through a minimum at 6 K and increase again in the region of the relaxation peak corresponding essentially to the strong pinning centres. The minimum value of $\beta$ ($\approx 4 \times 10^{-3}$) obtained from the data at 6 K corresponds to a relaxation curve that is logarithmic in the whole observation range of our experiments ($t \leq 10^5$ s), as it is the case for the relaxation data at $T < 1$ K.

Summarizing, the analysis of the relaxation data with the power-law time dependence confirms the results obtained from the analysis based on the logarithmic approximation of the time dependence. The temperature dependences of $\beta$ and of the rate $R$ can be explained with the model proposed by Geshkenbein and Larkin and are in agreement with the experimental results by Zavaritsky and Zavaritsky.

Fig. IV.26 Values of the exponent $\beta$ (+) obtained from the power-law fit to the relaxation curves measured with $H_1 \perp bc$. The corresponding values of the normalized relaxation rate $R$ (○) are also shown.
IV.4 ac Susceptibility Measurements

The SQUID measuring system [30,31] installed in the dilution refrigerator (§ III.1) can also be used to measure the complex susceptibility \( \chi = \chi' + i \chi'' \) of the specimens.

In order to characterize our samples, we have measured the susceptibility of both samples (crystal 4 and 5, cf. § IV.1) as function of temperature between 5 mK and 9.2 K. The measurements were performed applying ac fields \( H_{ac} \) between 0.05 and 33 mOe. The ac fields used have a frequency of 80 (160) Hz and were applied parallel (perpendicular) to the bc-plane of crystal 5 (crystal 4). Thus, the field orientations are the same as for the dc fields in section IV.3.

The susceptibility \( \chi \) of crystal 4 was measured both in zero field and with an applied dc field (\( H \perp bc \)) of 407 Oe (after zero-field cooling). Both \( \chi'(T) \)-curves show quite sharp transitions, although the zero-field data do not include the full diamagnetic transition, due to the fact that the measuring system (§ III.1) works only up to 9.2 K [\( T_c(NbTi) = 9.2 \) K]. The \( \chi' \)-data in Fig. IV.27 show that the width of the transition is practically unchanged by the application of a dc field of about 400 Oe. The effect of the dc field is mainly to shift the diamagnetic transition to lower temperatures by about 3 K. For the zero-field data the resistive component \( \chi'' \) of the signal is very small and constant throughout most of the measured temperature range. Only above 8.5 K does \( \chi'' \) increase abruptly and it seems to reach a maximum at 9.2 K. For the field-on data the resistive component \( \chi'' \) exhibits a narrow peak at \( T = 6.3 \pm 0.2 \) K.

We determine the critical temperature of the samples as the midpoint of the temperatures corresponding to 10 % and 90 % of the full diamagnetic diamagnetic transition. Unfortunately, for the zero-field data of crystal 4 the temperature corresponding to 10 % of the diamagnetic transition is beyond the temperature range of our measurements. We thus estimate \( T_c \) as the point corresponding to 50 % of the full diamagnetic transition and obtain \( T_c = 9.1 \) K. From the temperature corresponding to a value of \( \chi'/\Delta \chi'_\text{max} \) of 90 % (\( T = 8.6 \) K), we obtain an upper limit of 1 K for the 10 to 90 % width \( \Delta T_c \) of the transition. The value of \( T_c \) obtained is in agreement with the position of the peak of \( \chi'' \). In the case of the field-on data the temperatures corresponding to 10 % and 90 % of the diamagnetic
The \( \chi' \)-data of crystal 5 (H\(_{ac} \parallel bc \)) exhibit (Fig. IV.28) a transition somewhat different from the one shown in Fig. IV.27. Starting from \( T > T_c \), the first half of the diamagnetic transition is rather steep, whereas the second part is much more rounded and the susceptibility value does not saturate down to the lowest temperature of our measurements (\( T \approx 1.4 \) K). The resistive part \( \chi'' \) of the complex susceptibility is extremely small throughout the range of the measurements, so that, due to the noise of the signal, no peak could be detected.

From the \( \chi' \)-data of crystal 5 it is possible to obtain the temperatures...
Fig. IV.28 Real part $\chi'$ of the complex susceptibility, normalized with the total diamagnetic signal $\Delta \chi'_{\text{max}}$ of crystal 5 (•). The measurements were performed in zero dc field. The zero-field data of crystal 4 (○) are also shown for comparison. Dashed lines are guides to the eye.

corresponding to 10 % and 90 % of the full diamagnetic transition. However, a qualitative analysis of the $\chi'(T)$-curve shows that the broadening of the curve takes place essentially in the second half of the transition, that is for $T < T_{50\%} = 8.5$ K. The same qualitative behaviour of the $\chi'(T)$-curve as function of the field orientation has been observed by Kanoda et al. [79] for a single crystal of the same organic compound. Their data show that, for the same geometry as for crystal 5 ($H_{\text{ac}} \parallel \text{bc}$), the penetration of flux lines lying parallel to the superconducting layers (bc-plane) in a direction also parallel to the layers is dominant. This is noteworthy, since in this configuration one would expect the penetration depth in the direction perpendicular to the layers to be mainly responsible for the decrease of $\chi'(T)$. As an explanation they invoke the layered structure of
this compound and the weak coupling existing between the superconducting layers.

In fact, it has been shown throughout this work that the coupling between the layers is very weak, so that this material behaves essentially as a stack of 2D superconducting layers. Assuming that the penetration mode described above (Kanoda et al.) is dominant, we have estimated the value of the corresponding penetration depth $\lambda^*_{bc}(0)$ to be about 50-60 Å.

Unfortunately, our ac susceptibility data were taken on two different single crystals in different field orientations and do not allow a systematic study of the field penetration into the samples. It is therefore not clear what penetration modes are responsible for the decrease of $\chi'(T)$ in each field orientation. However, all the fits that we have made indicate that the temperature dependence of $\lambda(T)$ is of the Ginzburg-Landau-type.
V. MAGNETIC RELAXATION EFFECTS IN A Y$_{1}$Ba$_{2}$Cu$_{4}$O$_{8+x}$ SINGLE CRYSTAL AT 4.2 K

In this chapter we present the results of the measurements of the remanent magnetization and of its time relaxation performed at 4.2 K on a single crystal of Y$_{1}$Ba$_{2}$Cu$_{4}$O$_{8+x}$ as function of the (maximum) applied field $H_i$. The measurements were performed with fields $H_i$ up to about 800 Oe and in three different field orientations. The three field orientations correspond to the three crystallographic axes of the crystal.

V.1 Specimens

Y$_{1}$Ba$_{2}$Cu$_{4}$O$_{8+x}$ (YBCO124) was discovered as a structure defect in decomposed powders of Y$_{1}$Ba$_{2}$Cu$_{3}$O$_{7.8}$ (YBCO123). Bulk synthesis of this compound was first achieved by Karpinski et al. [90] under high oxygen pressure. The critical temperature of this superconductor is $T_c \approx 80$ K for the bulk polycrystalline phase, whereas the best single crystals have a critical temperature $T_c$ of about 73 K. The crystal structure is orthorhombic with lattice constants $a = 3.871$ Å, $b = 3.840$ Å and $c = 27.240$ Å [91].

The crystal structures of YBCO123 and YBCO124 are very similar, but for an extra CuO chain between the CuO$_2$ planes in latter compound. This fact changes the distance between the superconducting layers significantly, namely from about 11.6 Å in YBCO123 to about 13.6 Å in YBCO124. Another consequence of the existence of extra CuO chains in the structure is to prevent the crystals from twinning. Moreover, in view of the larger layer spacing one would also expect a larger anisotropy of the superconducting parameters in YBCO124, as compared to YBCO123.

Measurements of the magnetic properties of YBCO124 on single crystal specimens are actually rather scarce up to now. Martinez et al. [92,93] have made a systematic study of the critical fields $H_{c1}$ and $H_{c2}$ as function of temperature and field orientation on a YBCO124 single crystal. They obtain values of $H_{c1}(0)$ [$H_{c2}(0)$] that are about three (three to five) times lower than those of YBCO123 independently of field orientation. This
implies that the anisotropy of both critical fields is not substantially different in the two compounds. It is thus clear that the values of the penetration depth $\lambda$ and of the coherence length $\xi$ will differ by less than a factor of five in these two materials. Magnetization measurements \[94,95\] have also shown that the critical currents of YBCO124 are approximately equal to the ones of YBCO123.

Our specimen is a single crystal with the shape of a parallelepiped and dimensions of about $1.2 \times 0.6 \times 0.3 \text{ mm}^3$. The surface and the corners are very irregular, so that strong local demagnetization effects are expected for any field orientation.

Measurements of the ac susceptibility as function of temperature yield a rather broad diamagnetic transition. The value of the critical temperature determined as the midpoint of the 10 % to 90 % transition width is 55 K. This value is also confirmed by the position of the peak of the dissipative component $\chi''$ as function of temperature.

Measurements of polarized light reflectance made by B. Bucher (ETH Zurich) have shown that the crystallographic axes of the crystal are approximately parallel to the edges of the parallelepiped. The a-axis is parallel to the longest dimension of the crystal, the b-axis to the middle dimension and the c-axis to the thickness.

The same crystal was measured in three different field orientations. Assuming an ellipsoidal shape, we have calculated the corresponding demagnetization factors. We obtain $D = 0.60 \pm 0.10$, $D = 0.28 \pm 0.08$ and $D = 0.11 \pm 0.03$ for the field orientations with $H \parallel c$, $H \parallel b$ and $H \parallel a$, respectively.

The single crystal used in our experiments has been provided by J. Karpinski and E. Kaldis, ETH Zurich.
V.2 Magnetic Relaxation Effects of the Remanent Magnetization at 4.2 K

V.2.1 Remanent Magnetization at 4.2 K as Function of the Field $H_i$

Values of the remanent magnetization were obtained for the three field orientations (see previous sections) from isothermal dc magnetization curves (at 4.2 K) up to a maximum applied field $H_i$ (§ III.3.1). The results are depicted in Fig. V.1 and show that the fully critical state is reached within the field range of our experiments only when the applied field is parallel to the a- or b-axis.

The data could be well fitted using the field dependences of $M_{rem}$ obtained (§ II.1) with Bean’s critical-state model. For $H_i \parallel b$ and $H_i \parallel a$ the fits to the $M_{rem}$ data yield values of $dJ_c$ of 800 and 650 Oe, respectively, whereas the corresponding values of $K$ are 51 and 27.5 $\Phi_0$/Oe. The

![Fig. V.1](image)

Fig. V.1 Remanent magnetization $M_{rem}$ at 4.2 K of a $Y_1Ba_2Cu_3O_{8+x}$ single crystal as function of the applied field $H_i$ for three different field orientations: (○) $H_i \parallel c$, (▲) $H_i \parallel b$, and (●) $H_i \parallel a$. The solid lines are fits to the data obtained on the basis of the critical-state model by Bean (see text).
calculated values of $K$ (§ III.5) are much higher, namely 500 $\Phi_0$/Oe for $H_i \parallel b$ and 370 $\Phi_0$/Oe for $H_i \parallel a$, showing that the screening of fields parallel to the superconducting layers is rather weak.

Qualitatively, a similar effect was observed in the organic superconductor (BEDT-TTF)$_2$Cu(SCN)$_2$ (previous chapter). In that case the weakness of the screening effects in the parallel field orientation was also confirmed by extremely low values of $H_{c1}$ (< 0.5 Oe) at all temperatures. In the case of $Y_1Ba_2Cu_{4}O_{8+x}$ the values of $H_{c1}$ determined from the magnetization curves (cf. § IV.2) confirm this effect only to some extent.

In fact, at 4.2 K and after correcting for demagnetization effects, we obtain $H_{c1}^b = 65 \pm 20$ Oe for $H \parallel b$ and $H_{c1}^a = 17 \pm 3$ Oe for $H \parallel a$. The value of $H_{c1}$ compares well with the ones obtained by Martinez et al. [92]. On the other hand, they do not observe any significant anisotropy of $H_{c1}$ between the a and b direction, whereas our value of $H_{c1}^a$ is about a factor of 4 smaller than the value of $H_{c1}^b$.

The fit to the remanent magnetization data for $H_i \parallel b$ (Fig. V.1) shows another interesting effect, already observed by other authors [96] at low temperatures. The fit follows the data points well in the field regime just below [Eq. (II-8)] the saturation of $M_{rem}$, but it clearly underestimates the values of $M_{rem}$ in the low-field regimes [(Eqs. (II-6) and (II-7)]. On the other hand, adjusting the fit parameters to the data points in the low-field regimes causes the fit to be about 50% too low in the saturation regime. It is likely that, at low temperatures, the strong increase of $M_{rem}$ at low fields is essentially due to flux penetrating the sample because of strong demagnetization effects at the corners. The fact that we observe this effect for applied fields parallel to the plane of the crystal (Fig. V.1, $H \parallel b$), confirms the idea that local demagnetization effects at the corners and at surface irregularities of the samples are involved in the process.

According to Ref. 96 the same problem occurs (for all field orientations) in the case of fits based on the Bean model with a field-dependent critical current. This would indicate that the field dependence of the critical current does not contribute substantially to this effect. It is noteworthy that at higher temperatures ($T \geq 35$ K) an almost inverse effect is observed [96], namely, that fitting the data in the low-field regime produces an overestimate of the saturation value of $M_{rem}$ of about a factor of two.

In the case of the remanent magnetization data measured with $H_i \parallel c$,
the values of the parameters $dJ_C$ and $K$ could not be obtained independently of each other from the fit (§ III.6.1), because of the limited field range of our experiments. We have made only a qualitative fit of the data in one field regime [Eq. (II-7)], because the first field regime [$H_{c1} < H_i < 2H_{c1}$, Eq. (II-6)] could not be determined. In this field range there is an essential broadening of the curve, due to strong demagnetization effects at the corners of the specimen. We calculate the value of $K$ and obtain $K = 560 \Phi_0/Oe$ (with $A_s = 0.80 \text{ mm}^2$ and $N_{\text{loop}} = 2$). In this field orientation ($H \perp$ layers) the calculated value of $K$ should be realistic (cf. § IV.2.1 and IV.3.1). This yields a value of $dJ_C$ of about 15 kOe.

Taking a value of $L_x = 0.6 \text{ mm}$, we estimate [Eq. (II-16)] the value of $J_{c}^{ab}$ to be of the order $2 \times 10^5 \text{ A/cm}^2$. It is clear that this value represents only a rough estimate of the order of magnitude of $J_{c}^{ab}$, because of the limited field range of our experiments. As discussed before, fitting $M_{\text{rem}}(H_i)$-data in the low-field regime (at low $T$) leads to essential underestimate of the value of $J_c$. Indeed, our value of $J_{c}^{ab}$ is about 5 times smaller than the one obtained by extrapolation to 4.2 K of the data (at $T \gtrsim 10 \text{ K}$) of Martinez et al. [94].

In the case of the remanent magnetization data measured with $H_i$ parallel to the $a$- and $b$-axis the fully critical state was reached within the field range of our experiments. The values of the aspect ratio $L_x/t$ of the crystal for $H_i \parallel b$ and $H_i \parallel a$ are 4 and 2, respectively. The (fully) critical state of the sample in these two field orientations is then mainly determined by the critical current component $J_c$ (§ II.1).

From the saturation values of $M_{\text{rem}}$ with $H_i \parallel b$ and $H_i \parallel a$, we obtain (using $M_{\text{rem}} = K L_x J_c^{c}/4$) values of $J_c$ of about $5 \times 10^3$ and $9 \times 10^3 \text{ A/cm}^2$, respectively. Taking into account anisotropy effects [Eq. (II-14)] does not bring any significant change to the values of $J_c$ previously calculated. An extrapolation of the values of $J_c$ in Ref. 94 to 4.2 K yields a value of $J_c(4.2 \text{ K})$ about an order of magnitude larger than ours.

IV.2.2 Time Relaxation of the Remanent Magnetization at 4.2 K as Function of the Field $H_i$

All the relaxation curves of the remanent magnetization measured at 4.2 K on the same sample in three different field orientations show
deviations from the logarithmic law. As in the previous chapter, we have fitted the relaxation data with the power law described by Eq. (IV-2) after setting \( t_0 \) equal to 1 s. According to the collective creep theory the power law approximation is justified (§ II.3) in the case of single-vortex creep.

From the saturation values of \( M_{\text{rem}} \) we calculate the average distance \( a_{\phi_o} \) between the flux lines inside the samples at the beginning of our relaxation experiments. We obtain values of \( a_{\phi_o} \) of about 1.5 \( \mu \text{m} \) for all samples. The values of the penetration depths \( \lambda_c \) and \( \lambda_{ab} \) obtained by Martinez et al. [93] for this compound are 1.2 \( \mu \text{m} \) and 0.18 \( \mu \text{m} \), respectively. This shows that the interactions between flux lines should still be weak in the range of our experiments, so that flux pinning (and creep) is likely to take place through motion of single vortices.

The collective pinning approach for high-\( T_c \) superconductors may be justified by the observation of very strong relaxation effects (cf. Refs. 52,53,81), even at very low temperatures. This indicates that in these materials pinning is essentially provided by weak pinning centres. Moreover, Dolan et al. [97] observed that at low temperatures vortices are pinned also in defect-free regions of \( \text{YBa}_2\text{Cu}_3\text{O}_7-\delta \) single crystals. They conclude that in this material there must be an extremely dense distribution of weak pinning centres. It was calculated by E.M. Chudnovsky [98] that in the case of Bi-Sr-Ca-Cu-O the source of pinning may be represented by oxygen vacancies in the CuO layers. These defects are believed to be the main pinning mechanism characterizing the high-\( T_c \) superconductors in general.

From the values of the fit parameters \( b \) and \( \beta \), we calculate the values of the initial logarithmic slope \( S = b - \beta \cdot \ln 10 \). The field dependences of \( S \) and \( M_{\text{rem}} \) are very similar for all field orientations. In order to normalize the values of \( S \) we calculate the part \( M_{\text{rem}}^{\text{out}} \) of the remanent magnetization, that decays outwards [Eqs. (II-10) to (II-13)].

The obtained values of \( R = S/M_{\text{rem}}^{\text{out}} \) show (Fig. V.2) that at 4.2 K relaxation effects in this \( \text{Y_1Ba}_2\text{Cu}_4\text{O}_{8+x} \) single crystal are almost as strong as in the organic compound (cf. Fig. IV.19, \( H_i \perp bc \)) analysed in the previous chapter. Quantitatively, these data confirm the high values of the relaxation rates previously obtained by our group [52,99] for a \( \text{Y_1Ba}_2\text{Cu}_3\text{O}_{7-\delta} \) single crystal at 4.2 K (\( H_i \parallel c \)). However, for \( H_i \parallel c \) the
fully critical state of the sample was not reached, so that these results are more difficult to interpret.

The parameters from the fit to the \( M_{\text{rem}} \) data for \( H_i \parallel c \) (\( dJ_c \sim 15 \text{ kOe} \)) show that the field value characterizing the full penetration of the sample (\( H_i \simeq dJ_c/2 \)) is much larger than the upper limit (\( H_i = 800 \text{ Oe} \)) of the field range of our experiments. Thus, the data points obtained for \( H_i \) smaller than 300 - 400 Oe (previous figure, \( H_i \parallel c \)) are very likely to be due to flux penetrating the corners of the sample because of strong demagnetization effects. This explains the extremely high values of the decay rate obtained in this field regime. Indeed, for \( H_i > 400 \text{ Oe} \) the values of \( R \) are smaller and almost independent of the field \( H_i \).

An estimation of the activation energy from these data points yields a value of \( U_c^{\perp} \) of about 8 meV. This value is about a factor of two smaller than the ones obtained by several groups \([100,101,102,103]\) for single crystals and oriented grains of \( \text{YBa}_2\text{Cu}_3\text{O}_7-\delta \) in the same field orientation.
This difference is partly due to the fact that our value of $U_0$ was calculated from the initial ($t \approx 1$ s) logarithmic slope $S$ of the relaxation curves. Another factor that is likely to contribute to this difference is the incomplete field penetration of our sample ($H_i \parallel c$). In fact, this field regime is often characterized by stronger relaxation effects than the fully critical state (Fig. V.2, $H_i \parallel b$). Moreover, most of the relaxation data previously quoted (except for Ref. 100) were obtained from measurements under constant applied field ($H \geq 1$ T) at low temperatures (5-6 K). It is well known that the determination of the irreversible part of the magnetization is not unproblematic, so that the correct normalization of the field-on relaxation rates is not trivial.

The data points for $H_i \parallel b$ and $H_i \parallel a$ in Fig. V.1 indicate that in these field orientations the sample reached the fully critical state for values of $H_i$ approximately equal to 800 Oe and 650 Oe, respectively. The corresponding data points on the right-hand side of Fig. V.2 yield values of $R$ between 0.06 and 0.08, so that the values of $U_c^I$ are of the order 10-12 meV. Again, due to the lack of relaxation data on YBCO124, we compare our results with relaxation data on YBCO123.

Yeshurun et al. [102] (and Refs. 13, 14 therein) have obtained values of $U_c^II$ ($H \parallel ab$) of the order of 0.15-0.5 eV for single crystals of YBCO123. On the other hand, Keller et al. [100] give a value of $U_c$ of about 25 meV for oriented grains of YBCO123, independent of field orientation. The interpretation of these results is not clear.

As a comparison, the values of $U_c^II$ and $U_c^I$ obtained by Biggs et al. [104] for the extremely anisotropic ($\Gamma \approx 3000$) compound Bi$_2$Sr$_2$CaCu$_2$O$_{8+8}$ are 48 and 12 meV at 10 K, respectively. One would expect that also in the Y-Ba-Cu-O compounds the layered structure of the material influences to some extent the pinning mechanisms for fields applied parallel to the layers. The relaxation data by Yeshurun et al. give evidence for the existence of this effect, whereas the results of our group and of Keller et al. do not. A systematic study of the relaxation phenomena in well-characterized samples is needed in order to clarify this issue.

Summarizing, the relaxation data on single crystals of YBCO124 at low temperatures ($T = 4.2$ K) give relaxation rates of about the same order of magnitude as in YBCO123. Unfortunately, due to the limited field range of our experiments, a more conclusive study of the anisotropy of the relaxation phenomena in YBa$_2$Cu$_4$O$_{8+x}$ was not possible.
The results of the analysis of the relaxation curves done with the power law time dependence yields some interesting features. From the values of the fit parameters $M_{\text{rem}}(1 \text{ s})$ and $M_{\text{rem}}(\infty)$, we calculate the decay fraction $b/M_{\text{rem}} = (M_{\text{rem}}(1 \text{ s}) - M_{\text{rem}}(\infty))/M_{\text{rem}}$ of the remanent magnetization. The obtained values range between 0.25 and 0.7 (Fig. V.3). The data at low fields show qualitatively different behaviours for $H_i \parallel c$, than for $H_i \parallel b$ and $H_i \parallel a$. For latter two field orientations $b/M_{\text{rem}}$ increases with the applied field $H_i$ and reaches a value of about 0.4 for $H_i \approx 780$ Oe (fully critical state). For $H_i \parallel c$ the values of $b/M_{\text{rem}}$ exhibit a maximum around $H_i \approx 150$ Oe, then fall down, and start to increase again reaching a value of 0.55 at the upper limit of our field range. The $H_{c1}$ data of Martinez et al. [92] and our $M_{\text{rem}}$ data (Fig. V.1) for this field orientation show that the peak of $b/M_{\text{rem}}$ occurs at fields approximately equal to $H_{c1}$. This is a further indication that flux trapped close to the surface of the specimen because of demagnetization effects is less effectively pinned than flux trapped inside the specimen.

![Graph showing the decay fraction $b/M_{\text{rem}}$ as a function of the field $H_i$ for three different field orientations.](image)

**Fig. V.3** Values of the decay fraction $b/M_{\text{rem}} = (M_{\text{rem}}(1 \text{ s}) - M_{\text{rem}}(\infty))/M_{\text{rem}}$ at 4.2 K as function of the field $H_i$ for three different field orientations: (○) $H_i \parallel c$, (▲) $H_i \parallel b$, and (●) $H_i \parallel a$. 
The determination of a typical value of $b/M_{\text{rem}}$ for $H_i \parallel c$ is rather difficult, because within the limited field range our experiments relaxation effects still depend on the number of flux lines trapped inside the specimen. A comparison with the relaxation data of (BEDT-TTF)$_2$Cu(SCN)$_2$ at 4.2 K (Fig. IV.10) shows that the corresponding values of $b/M_{\text{rem}}$ (in the fully critical state) are 10 to 20% higher than the ones obtained for the high-$T_c$ compound. However, the values of $b/M_{\text{rem}}$ obtained for YBCO124 confirm the idea that in high-$T_c$ superconductors induced currents can hardly be considered as "persistent".

We now present the values of the fit parameter $\beta$ as function of the applied field $H_i$. We find that the exponent $\beta$ and the relaxation rate $R$ have very similar field dependences. The next figure shows this effect for the relaxation data measured with $H_i \parallel c$. These data confirm the intuitive idea that a “strong” relaxation can subsist over a shorter period of time than a weaker relaxation. For this reason a weak relaxation may appear logarithmic in time within the observation range, whereas strong relaxations show deviations from the logarithmic time dependence at much shorter

![Graph](image_url)

**Fig. V.4** Values of $\beta$ (+) obtained from the power law fit to the relaxation curves at 4.2 K as function of the applied field $H_i$ ($H_i \parallel c$). The corresponding values of the normalized decay rate $R = S/M_{\text{out}}^{\text{rem}}$ (○) are also shown.
times. Moreover, it was also shown in section IV.3.2 that marked deviations from the logarithmic law are also typical of temperature regimes, where flux-creep is ruled by the competition of different kinds of pinning centres.

These results confirm the prediction of the theory of collective creep (§ II.3) for the case of single-vortex creep $[\alpha(k_B T/U_c)\ln(t/t_0) \ll 1]$. In fact, in this case the curvature of the relaxation curve is given by the power law exponent $\beta = k_B T/U_c$, i.e. by the value of the normalized relaxation rate.
VI. RELAXATION RATE OF THE REMANENT MAGNETIZATION OF CLASSICAL, CHEVREL PHASE, ORGANIC AND HIGH-\(T_C\) SUPERCONDUCTORS AT 4.2 K

In this chapter we will only give a brief overview of the magnitude of magnetic relaxation effects at 4.2 K in several families of type-II superconductors. To this purpose we will present the values of the relaxation rates of the remanent magnetization at 4.2 K as function of the applied field \(H_i\). All the measurements were taken in the 4K-Magnetometer (§ III.2 and III.3) as function of the applied field \(H_i\) up to about 500 Oe (800 Oe for \(Y_1Ba_2Cu_4O_{8+x}\)).

The measurements were performed on the following superconducting specimens: \(Pb_{0.4}In_{0.6}\) wire, polycrystalline Chevrel Phase \(PbMo_6S_8\), \(NbTi\) wires, sintered \(Sr_{0.2}La_{0.8}CuO_4\), \((BEDT-TTF)_2Cu(SCN)_2\) single crystals, \(Y_1Ba_2Cu_3O_{7-\delta}\) and \(Y_1Ba_2Cu_4O_{8+x}\) single crystals.

We will now briefly describe the specimens and some of the characteristic features obtained from the measurements of the isothermal dc magnetization curves. Thereafter we will present the values of the relaxation rate for the different specimens. The magnitude of the relaxation effects and the occurrence of deviations from the logarithmic time dependence in the different materials will be discussed.

The \(Pb-In\) sample was obtained from a wire with nominal composition \(Pb_{0.4}In_{0.6}\). It was 7.3 mm long and 1.0 mm thick and was measured with applied fields parallel to the longitudinal axis of the cylinder. This sample was actually used to study the influence of demagnetization effects at the corners and at the surface. The details of this analysis have been described by A. Pollini ([31], § 5.2). Following a suggestion by Prof. J.L. Olsen the edges of the sample were rounded without changing the length-to-diameter ratio of the sample. This way the shape of the sample becomes closer to an ellipsoid. The corresponding calculated demagnetization factor is \(D = 0.03\). The critical temperature \(T_C\) of this material is about 6.2 K.

Measurements of \(M_{rem}\) show that for \(H_i \geq 200\) Oe the fully critical state is achieved inside the specimen. A comparison with data taken on a piece of wire with the same dimensions, but with sharp edges, shows that the
saturation value of $M_{\text{rem}}$ of our sample is smaller by about 40%.

This can be explained by the existence of a surface energy barrier of the Bean-Livingston type [105]. This barrier prevents the flux from entering the sample with rounded edges more effectively at low (increasing) fields. On the other hand, in decreasing fields this kind of barrier is more effective in the sample with sharp edges and impedes to some extent the flux from escaping the specimen.

The analysis of the dc magnetization curves shows that the field of the first flux penetration is about 80 Oe (130 Oe) for the sample with sharp (rounded) edges. Here the fields of the first penetration were determined from the first deviation of the magnetization curve $M(H)$ from linearity. These results show that strong demagnetization effects at corners play an important role for flux penetration, even in the case of samples with a small demagnetization factor. In Ref. 31 it was shown that the determination of $H_{c1}$ from the first macroscopic deviation of the slope $-\partial M/\partial H$ of the magnetization curve $M(H)$ from the Meissner value is much less sensitive to local demagnetization effects. With this method, we obtain $H_{c1} \approx 138$ Oe (143 Oe) for the sample with sharp (rounded) edges. This method has been used throughout this work for the determination of the values of $H_{c1}$ of our samples.

The polycrystalline Chevrel Phase PbMo$_6$S$_8$ sample is a cylinder with dimensions of about 7.15 mm x 1.85 mm. The critical temperature is $T_c \approx 14$ K. The sample was measured in applied fields parallel to the axis of the cylinder. The corresponding demagnetization factor (ellipsoid approximation) is $D = 0.07$. From isothermal dc magnetization curves we obtain a value of $H_{c1}$ of 100 Oe ($\pm 5$). This sample was kindly provided by Prof. Ø. Fischer, University of Geneva.

A bundle made of about 120 pieces of insulated NbTi wire ($0.005"$ NbTi wire, $\varnothing = 0.13$ mm, S.H.E. Corporation, San Diego) with a critical temperature of 9.2 K was also used as specimen. This is the kind of wire that we have used to build the gradiometer (§ III.2.2) of the 4K-Magnetometer. The wires used as sample are about 4.0 mm long. The magnetic fields were applied perpendicularly to the axis of the wires. The corresponding demagnetization factor (for a single wire) is $D = 0.5$. With the aforementioned method, we obtain a value of the lower critical field $H_{c1}^*$ of $95 \pm 5$ Oe, i.e. $H_{c1} = 190 \pm 10$ Oe.

The Sr-La-Cu-O sample was first measured [106,107] in the dilution
refrigerator (§ III.1). It consists of a sintered Sr$_{0.2}$La$_{0.8}$CuO$_4$ parallelepiped with dimensions 7.8 x 1.8 x 1.8 mm$^3$. The value of the critical temperature is $T_c \approx 40$ K. In the 4K-Magnetometer it was measured in applied fields parallel to its longitudinal axis, so that the corresponding demagnetization factor is $D = 0.07$. From magnetization curves we determine a value of $H_{c1}$ of 100 ± 10 Oe.

The Y$_1$Ba$_2$Cu$_3$O$_{7-\delta}$ single crystal is a thin platelet with dimensions of 1.0 x 0.9 x 0.04 mm$^3$. It was measured in magnetic fields applied parallel to the c-axis, i.e. perpendicular to the basal plane of the crystal. The calculated demagnetization factor is $D \approx 0.92$. The relaxation data on this single crystal have been published elsewhere [52,99]. A recent analysis of the remanent magnetization data by D. Risold [41] yields a value of $H^*$ of about 3 kOe for this specimen. From this value we estimate (within a factor of two) the in-plane critical current $J_{cb}$ to be about $7 \times 10^4$ A/cm$^2$. It is clear that the determination of $J_{cb}$ in the limited field range of our experiments is extremely difficult due to demagnetization effects. Moreover, in the case of such a thin crystal self-field effects [21] are also expected to play an important role.

The Y$_1$Ba$_2$Cu$_4$O$_{8-x}$ single crystals have been described in the previous chapter (§ V.1). Note that, for convenience, the data taken with $H_i || b$ and $H_i || a$ will be plotted (Fig. VI.1) using the same symbol.

The (BEDT-TTF)$_2$Cu(SCN)$_2$ single crystals have been described in section IV.1. The relaxation data denoted by $H_i \perp bc$ ($H_i || bc$) correspond to crystal 1 and crystal 3 (crystal 2). For convenience, we have plotted only the data points obtained with the sample in the fully critical state.

In Fig. VI.1 we present the values of the relaxation rate of the remanent magnetization at 4.2 K as function of the applied field $H_i$ for all the specimens listed above. Due to the limited field range of our experiments it was not always possible to fit the $M_{rem}$ data with the field dependences calculated within the framework of the Bean model. This implies that the quantity $M_{rem}^{out}$ could not be determined for all the specimens. The values of the initial logarithmic slope $S = -\partial M_{rem}(t)/\partial \log t |_{t \rightarrow \infty}$ of the relaxation curves were then normalized by the values of $M_{rem}$. The remanent magnetization determined from the magnetization curve up to a maximal field $H_i$. This means that the values of $R$ calculated from data corresponding to samples in a subcritical state ($H_i < 2H^* = dJ_c$) may be
underestimated by a factor of two at the most. This is the case of the Chevrel Phase, NbTi, Sr-La-Cu-O, Y1Ba2Cu3O7-δ and Y1Ba2Cu4O8+x specimens. In order to make the comparison more relevant, all the data points shown in Fig. VI.1 were calculated using $M_{\text{rem}}$, instead of $M_{\text{rem}}^{\text{out}}$.

Fig. VI.1 Values of the normalized relaxation rate $R = -\partial \ln M_{\text{rem}} / \partial \log t = S/M_{\text{rem}}$ of the remanent magnetization at 4.2 K as function of the applied field $H_i$ for different specimens: (+) Pb0.4In0.6 "rounded" wire, (▼) polycrystalline Chevrel Phase PbMogS8, (△) NbTi wires, (●) sintered Sr0.2La0.8CuO4, (○) Y1Ba2Cu3O7-δ single crystal with $H_i \parallel c$, Y1Ba2Cu4O8+x single crystals with $H_i \parallel c$ (◇) and $H_i \parallel ab$ (◆), single crystals of (BEDT-TTF)2Cu(SCN)2 with $H_i \perp bc$ (●) and $H_i \parallel bc$ (★).
It is clear that the data points in Fig. VI.1 can be divided in two categories according to the values of $R$. In fact, all the data points measured on organic or high-$T_c$ samples lie in the upper part ($R \gtrsim 2 \times 10^{-2}$) of the figure. The data points in the lower part of the figure correspond to classical superconductors like PbIn and NbTi or to Chevrel Phase superconductors.

The relaxation data on the organic samples (for $H_i \perp bc$) yield the largest values of $R$: $R \sim 0.15, U_c \sim 5$ meV. These values truly represent the magnitude of the relaxation effects in this compound, since they correspond to samples in the fully critical state. Moreover, it was shown (Fig. IV.19) that at lower temperatures ($T \sim 3$ K) the value of $R$ can be almost twice as large in this compound. On the other hand, the relaxation data taken with $H_i \parallel bc$ on this compound give values of $R$ around 0.03, i.e. about a factor of 5 smaller than for $H_i \perp bc$. This difference can be ascribed to the layered structure of this material, that strongly impedes flux motion in the direction perpendicular to the layers. Our magnetization data (§ IV.2 and IV.3) indicate that at 4.2 K this material behaves essentially as a two-dimensional superconductor.

The relaxation data on the Y-Ba-Cu-O compounds for $H_i \geq 400$ Oe seem to cluster around a typical value of $R$ of about 0.06, almost independently of the field orientation. These data points yield typical values of the activation energy of the order of 10 meV. It should be kept in mind that these data correspond to samples in the fully critical state only for $H_i \parallel ab$-plane. As already discussed in section V.2.2, it is not clear to what extent the anisotropy of these compounds is relevant for the magnetic relaxation data.

The relaxation data on the sintered Sr-La-Cu-O specimen yield values of $R$ (0.03) slightly lower than the ones of Y-Ba-Cu-O. A comparison between these two superconducting systems in terms of anisotropy is not easy, due to the lack of measurements on single crystals of Sr-La-Cu-O. It is clear that in view of the different values of $T_c$ of these two compounds our data at 4.2 K point to much stronger relaxation effects in Y-Ba-Cu-O than in Sr-La-Cu-O.

The values of $R$ for the other three superconducting materials are clearly smaller. The NbTi wires exhibit relaxation rates $R$ of the order of 0.003-0.004. Considering that in this material the relaxation is essentially logarithmic, one can see that an induced current would decay by about
3% within 1 year. In such a situation induced currents can be considered persistent for any practical purpose.

In this sense the PbIn specimen shows an even weaker relaxation of the remanent magnetization. The values of R are typically of the order of $1.1 \times 10^{-3}$ in the fully critical state ($H_i > 200$ Oe). This yields a value of $U_0$ of about 0.7 eV [31], in good agreement with the result of Beasley, Labusch and Webb [23], who obtained $U_0 \approx 1$ eV for PbTl.

For the Chevrel Phase PbMo$_6$S$_8$ we obtain values of R of the order of $2-3 \times 10^{-3}$. These values point to rather weak relaxation effects in this material. However, Mitin [82] has obtained values of R of the order of $1.7 \times 10^{-2}$ for polycrystalline cylinders of Pb$_{1.2}$Mo$_{6.4}$S$_8$ at 4.2 K. This value refers to the relaxation of flux trapped inside a hollow cylinder after the external field has been switched off. A value of R of the order of $1 \times 10^{-2}$ was obtained at about 4 K by Rossel et al. [108] for a single crystal specimen of PbMo$_6$S$_8$. This value was determined from relaxation curves with a constant applied field ($H = 500$ Oe). Our data yield a value of R smaller by almost an order of magnitude. This difference cannot be due to the fact that our relaxation data correspond to a regime of incomplete field penetration, because a correct normalization of the slope S would shift the values of R upwards by a factor of two at the most. It is therefore not clear if such a difference can be ascribed only to the different experimental arrangements and to the different specimens.

We will now briefly comment on the occurrence of deviations from the logarithmic time dependence in the relaxation data measured at 4.2 K. In the cases, where an analysis of the relaxation curves was possible, we will give the typical values of the exponent $\beta$ obtained from the power law fit [Eq. (IV-2), with $t_0 = 1$ s].

The relaxation curves of the PbIn sample and the NbTi wires are logarithmic within the time window of the experiments ($t \leq 10^5$ s).

For the Chevrel Phase specimen the decay curves of $M_{rem}$ were measured up to values of $H_i$ of 2400 Oe. The relaxation curves could be well fitted by a power law. The obtained values of $\beta$ are of the order of 0.04 at low fields ($H_i \sim 300$ Oe), reach a maximum ($\beta \approx 0.06$) around 1000 Oe and decrease back to 0.04 at 2400 Oe.

For the Sr-La-Cu-O specimen the power law exponent $\beta$ is almost constant for all fields $H_i > 100$ Oe. The typical values of $\beta$ range between
0.02 and 0.025.

The results of the analysis of the time relaxation curves of the organic samples were presented in Chapter IV (see Figs. IV.11 and IV.26). For the field orientation $H_i \parallel bc$ deviations from the logarithmic law (at times $t \leq 10^5$ s) are observed only in the data at higher temperature ($T \geq 2$ K), but the magnitude of these effects is rather small. This fact, together with the weakness of the signals measured for this specimen, have hindered a more systematic study of the relaxation curves. For the other field orientation ($H_i \perp bc$) the relaxation curves measured above 1 K show clear deviations from the logarithmic time dependence (cf. Fig. IV.24). The obtained values of $\beta$ are higher than for all the other specimens. Indeed, in the temperature region of the peak of the rate $R$ (Fig. IV.26) they are of the order of 0.15-0.20.

The relaxation curves measured for the $Y_1Ba_2Cu_4O_{8+x}$ single crystal in three different field orientations show approximately the same features. The values of $\beta$ determined from the fits range between 0.05 and 0.12 for all field orientations.

In the case of the relaxation curves measured on the $Y_1Ba_2Cu_3O_{7.5}$ single crystal the situation is not clear. These curves are essentially logarithmic in the time window of the experiments, that in this case was unfortunately somewhat shorter ($t \leq 10^4$ s) than for the other samples. A more precise analysis of these data was therefore not possible. Nevertheless, small deviations from the logarithmic law seem to occur at higher fields.

Summarizing, the data confirm the general idea that deviations from the logarithmic time dependence and high values of the relaxation rate $R$ are strictly correlated. Indeed, the largest deviations occur for the data on the organic samples ($H_i \perp bc$), that also show the strongest relaxation effects. In general, we conclude from our relaxation data at 4.2 K and low fields ($H_i < 800$ Oe) that one may expect noticeable deviations from the logarithmic time dependence in the case of superconductors, whose relaxation curves are characterized by values of the initial relaxation rate $R$ of the order of one percent or more.
REFERENCES

33. Model Ionos 40, Messer Griesheim GmbH, 5342 Kirchen-Eueneuen.
35. round wire Vacryflux 5001, Type S1/0.07 mm, Vacuumshmelze GmbH, 6450 Hanau.
40. B. Hubler, Semester Work, ETH Zurich March 1990.


70. Y. Kopelevich, A. Gupta, P. Esquinazi, C.-P. Heidmann, and H. Müller, preprint.


89. V.N. Zavaritsky, to be published.
ACKNOWLEDGEMENTS

I wish to express my greatest thanks to Prof. Dr. A.C. Mota for her careful and continuous teaching during the completion of this research work. Her enthusiasm for science and her support were fundamental for the realization of this work.

I would like to thank Prof. Dr. J.L. Olsen for the possibility to do my PhD research work in his group. His scientific advice and his gentle humour contributed greatly towards the creation of a pleasant research environment during these four years.

Ai miei amici e colleghi di lavoro Piero Visani, Andrea Pollini e Tiziano Teruzzi un grazie di cuore. La loro competenza scientifica e la loro disponibilità per problemi di qualsiasi tipo hanno permesso di creare un ambiente di lavoro piacevole, anche nei momenti di stress. Il loro aiuto ha facilitato di molto la soluzione dei problemi che un lavoro di ricerca scientifica pone quotidianamente. Inoltre ricorderò sempre con piacere il tempo dedicato assieme a discussioni di ogni tipo e a attività sportive.

Ein herzlicher Dank geht an unseren neuen Doktoranden Klaus Aupke und Marco Nideröst für ihre Freundlichkeit und für ihre Bereitschaft bei Problemen aller Art.


J'aimerais remercier tout particulièrement mes deux diplomands Jacques Robadey et Daniel Risold pour le travail qu'ils ont fait avec moi. Ils ont beaucoup contribué à l'analyse des données expérimentales que j'ai utilisées dans ma thèse.

Ringrazio di cuore il mio amico e compagno di studi Ivan Biaggio per il suo competente aiuto nella soluzione di problemi informatici.

Un sentito grazie va all'amico e compagno di studio Angelo Bernasconi per le corse nei boschi, le discussioni, le cene e i momenti trascorsi assieme in questi anni a Zurigo.
Frau M. van der Mark, Frau I. Dutly und Frau Ch. Schütterle danke ich herzlich für ihre Hilfe bei administrativen Problemen.


I would like to thank Prof. E. Kaldis and Dr. J. Karpinski for providing the Y-Ba-Cu-O specimens investigated in this work.

Ich möchte mich bei Dr. B. Hilti für die Herstellung aller einkristallinen Proben aus organischen Supraleitern bedanken, die in dieser Arbeit untersucht worden sind.

CURRICULUM VITAE

1961
Born in Ambri (Ticino).

1967-1972
Primary School in Ambri.

1972-1975
Scuola Maggiore (secondary school) in Ambri.

1975-1976
Avviamento Agricolo Cantonale Mezzana (secondary school).

1976-1978
Ginnasio cantonale (secondary school) in Giornico.

1978-1981
Liceo cantonale (high school) in Bellinzona.

June 1981
Maturità tipo C.

1981-1987
Study of physics at the Swiss Federal Institute of Technology (ETH) in Zurich.

1987
Diploma work on “Thermometrie bei Millikelvin Temperaturen mit SQUID’s” in the Low Temperature group of Prof. Dr. J.L. Olsen under the supervision of Prof. Dr. A.C. Mota at the Solid State Physics Laboratory of ETH.
1987-1991 Research and teaching assistant in the group of Prof. Dr. J.L. Olsen under the supervision of Prof. Dr. A.C. Mota.


LIST OF PUBLICATIONS


2. A.C. Mota, A. Pollini, P. Visani, and G. Juri Physica C 162-164, 695 (1989) "Low-field flux motion in Y1Ba2Cu3O7-δ single crystal and in other superconductors".


7. A. Pollini, A.C. Mota, P. Visani, G. Juri, and J.J.M. Franse
Proceedings of ICTPS 90 (Ed. R. Nicolsky), Rio de Janeiro, Progress
in High Temperature Superconductivity 25, 37 (1990)
"Relaxation effects and critical currents in heavy-fermion
superconductors".

8. A. C. Mota, G. Juri, P. Visani, A. Pollini, T. Teruzzi, K. Aupke, and
B. Hilti
Physica C 185-189, 343 (1991)
"Flux motion by quantum tunneling".

9. A. Pollini, A. C. Mota, P. Visani, R. Pittini, G. Juri, T. Teruzzi, and
J.J.M. Franse
Physica C 185-189, 2625 (1991)
"Novel magnetic relaxation effects in superconducting UPt$_3$ single
crystals".