Magneto-optical Kerr effect in the plasma-edge region
evidence for interplay between Drude term and interband transitions

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Magneto-Optical Kerr Effect in the Plasma-Edge Region: Evidence for Interplay between Drude Term and Interband Transitions

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presented by

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CONTENTS

Abstract 1

Riassunto 3

1. Introduction 5

2. Physical Properties 9

2.1 LaX (X = S, Se, Te) 9

2.2 GdS 13

2.3 Eu(Gd, 1)O 16

2.4 CeSb 19

References 22

3. Theory 25

3.1 Optical Spectroscopy 25

3.1.1 Definitions 26

3.1.2 Kramers-Kronig Transformations 28

3.1.3 The Lorentz-Drude Model 29
3.2 Magneto-Optical Kerr Effect Spectroscopy 33
3.2.1 Definitions 33
3.2.2 Experimental Determination of the Conductivity Tensor 35
3.2.3 Classical Microscopic Model 37
3.2.4 The Semiclassical Approach 40
3.2.5 The Lorentz-Drude Model in Magneto-Optics 45

References 56

4. Experiment 59
4.1 Optical Experimental Set-up 59
4.2 Magneto-Optical Experimental Set-up 60
4.2.1 The $^3$He Cryostat 60
4.2.2 Optical Set-up 63
4.2.3 Magneto-Optical Experimental Technique 66
4.3 Samples 69

References 70

5. Results and Discussion 71
5.1 LaX (X = S, Se, Te) 71
5.1.1 Previous Magneto-Optical Measurement of LaSe 72
5.1.2 New Results and Discussion 73
5.1.3 Inverse Photoemission (IPE) Measurements 81
5.2 GdS 85
5.2.1 Results and Discussion 85
5.3 Eu(Gdx)O 92
5.3.1 Previous Optical and Magneto-Optical Investigations of Eu(Gdx)O 92
5.3.2 New Results and Discussion 94
5.4 CeSb 106
5.4.1 Previous Magneto-Optical Investigation of CeSb 107
5.4.2 New Results and Discussion 108
ABSTRACT

Optical and magneto-optical polar Kerr effect spectroscopy investigations of lanthanum monochalcogenides (LaS, LaSe and LaTe) were performed. Results show that magneto-optical spectra of these compounds, which are paramagnetic metals, are dominated by the presence of a sharp, narrow structure occurring between 2 and 3 eV, i.e. in the plasma-edge region, and a broader feature at ~5 eV. In the earliest, single work on LaSe, the sharp, narrow structure was directly related to an interband electronic transition involving empty f-states. In the present work instead, a model based on the Lorentz-Drude model normally used to explain optical spectra is developed. With the support of this phenomenological model, it is shown that the presence of both free electrons (Drude component) and one (or more) interband transitions (Lorentz oscillator) at higher energies involving polarised electronic states give rise to a sharp, narrow structure developing in the reflectivity minimum. The second, broader structure at higher energies is now directly related to the interband electronic transition involving empty f-states. This new interpretation is also indirectly confirmed by inverse photoemission measurements (IPE).
Because of the interesting results obtained on LaX, optical and magneto-
optical spectra of gadolinium sulphide (GdS) and pure and Gd-doped europium 
oxide (EuO) were also studied. Results for GdS, which is an antiferromagnetic 
metal with $T_N = 62$ K, show that the magneto-optical Kerr effect is dominated by 
the presence of a single structure developing in the reflectivity minimum region. 
Both magnitude and shape only weakly depend on magnetic ordering. Since these 
results are very similar to those of LaX, a similar explanation is given, with the 
only difference that in GdS the $f$-states are occupied and centred at higher energies 
with respect to the empty ones of LaX.

The magneto-optical spectra of pure and Gd-doped EuO (ferromagnets with 
$T_C = 69$ K) also confirm the new interpretation given to LaX and GdS. In Gd-
doped EuO, a new and differently shaped structure, centred in the reflectivity 
minimum, was measured, whereas the same structure was not found in pure EuO. 
The reason is simply that, with respect to Gd-doped EuO, the reflectivity minimum 
of pure EuO is below the low-energy limit of our magneto-optical spectrometer. 
The different line-shape is associated with the polarisation of free electrons (Drude 
component) and not with that of the interband transition at higher energies.

Lastly, with the aim of confirming and possibly better clarifying the origin 
of the giant $90^\circ$ Kerr rotation at 0.46 eV, discovered on cerium antimonide (CeSb) 
in 1996, a systematic study of its magneto-optical properties on brand-new 
samples was performed. Our results did not confirm the previous analysis, and 
showed a maximal Kerr rotation of $-17^\circ$ at 0.46 eV. These results were also 
confirmed by measurements performed on thin films of the same material grown 
by another research group.
RIASSUNTO

In questo lavoro è stata fatta un'analisi degli spettri ottici e magneto-ottici della serie dei Lantano-monocalcogenidi (LaS, LaSe e LaTe). I risultati mostrano che gli spettri magneto-ottici di questi materiali, che sono dei metalli paramagnetici, sono dominati dalla presenza di una struttura stretta e piccata tra 2 e 3 eV, centrata nella regione in cui si trova il minimo nelle curve di riflettività (frequenza di plasma schermata) e di una seconda struttura più larga a ~5 eV. Nella precedente e unica interpretazione, basata su una singola misura effettuata sul LaSe, la struttura stretta e piccata veniva associata con la manifestazione diretta di una transizione interbanda in uno stato f vuoto. In questo lavoro si è invece sviluppato un modello fenomenologico basato sul modello di Lorentz-Drude normalmente utilizzato per interpretare gli spettri ottici, che dimostra come la contemporanea presenza di elettroni liberi (transizioni intrabanda, contributo di Drude) e di una (o più) transizioni interbanda (contributo di Lorentz) ad alta energia che coinvolgano stati elettronici polarizzati, sia la causa dell’insorgere di una struttura stretta e piccata centrata nel minimo della riflettività. La seconda struttura a più alta energia è invece la manifestazione diretta della transizione interbanda che coinvolge gli stati...
Dall'analisi data ai LaX e al GdS, con la differenza che nel GdS gli stati f sono occupati e situati ad energia più alta rispetto agli stati f vuoti nei LaX.

Gli spettri magneto-ottici dell'EuO e dell'EuO drogato con Gadolinio (ferromagnetici per temperature minori di $T_C = 69$ K) hanno fornito una conferma della nuova interpretazione data ai LaX e al GdS. Infatti nell'EuO drogato si è misurata una nuova struttura centrata nel minimo della reflettività con una forma diversa dai casi dei LaX e del GdS, mentre la stessa struttura non è stata trovata nell'EuO puro. Il motivo è semplicemente che, rispetto al caso dell'EuO drogato, l'EuO puro ha il minimo di reflettività a energie più basse del limite inferiore dello spettrometro usato per le misure di magneto-ottica. La forma diversa della struttura è stata associata alla polarizzazione non della transizione interbanda ad alte energie, bensì degli elettroni liberi.

Con lo scopo di confermare la presenza e di eventualmente approfondire l'origine della rotazione Kerr record di 90° a 0.46 eV scoperta nell'antimoniuro di Cerio (CeSb) nel 1996, si è infine effettuato uno studio sistematico delle sue proprietà magneto-ottiche su nuovi campioni cresciuti appositamente. I risultati non confermano l'analisi precedente, mostrando a 0.46 eV una massima rotazione Kerr di $-17^\circ$, confermata anche da misure effettuate su film sottili dello stesso materiale cresciuti da un altro gruppo di ricerca.
1. INTRODUCTION

In the last few years, magneto-optical Kerr effect spectroscopy (MOKE) has revealed itself as a very powerful technique to study the electronic structure of several materials [1-2]. Since MOKE spectroscopy is 'selectively' sensitive to electronic transitions involving spin-polarised states (for instance, \( f \)-states), it is particularly suitable for studying the electronic structure of rare-earth compounds. The magneto-optical signal is proportional to the joint spin-polarisation between initial and final states of the optical transition. Therefore, very large observed magneto-optical Kerr angles arise from \( f \to d \) transitions. While transitions between \( f \)- and \( d \)-states have a strong magneto-optical signal, in conventional optical spectroscopy the signal of the \( f \)-states in metals nearly vanishes in the background of strong band-to-band transitions because, in diagonal conductivity, oscillator strengths of transitions involving quasi-localised \( f \)-states are very small.

It has recently been argued that even a non-magnetic or a weakly magnetic material, i.e. one with non-occupied spin-polarised states, may have a strong magneto-optical signal. This should be the case when the system has an empty spin-polarised state, which may be occupied in the optical absorption process. Evidence for a strong magneto-optical signal of a supposed empty spin-polarised
4f-state was found in the paramagnetic lanthanum selenide (LaSe) metal [3]. This pioneering work motivated us to investigate the whole lanthanum monochalcogenides series (LaX, with X = S, Se and Te). Results show that magneto-optical spectra are dominated by the presence of a sharp, narrow structure occurring in the plasma-edge region and arising from the presence of both free electrons (Drude component) and one interband transition (Lorentz oscillator) at higher energies involving polarised electronic states, i.e. f-states.

In order to clarify better the role played by empty f-states in LaX, especially with respect to their energy ‘position’, the gadolinium counterpart of LaS, gadolinium sulphide (GdS), was investigated. GdS also shows plasma-edge behaviour, orders antiferromagnetically below 62 K, and occupied electronic f-states are situated between 7 and 9 eV below the Fermi energy level.

Furthermore, since LaX and GdS are paramagnetic and antiferromagnetic metals respectively, we wished to analyse a ferromagnetic system exhibiting plasma-edge behaviour. Europium oxide (EuO) is ferromagnetic under 69 K and, if doped with ‘sufficiently high’ percents of gadolinium (EuGdₓO), seems to show plasma-edge behaviour. In this system the plasma-edge position may be shifted towards higher energies simply by increasing the amount of gadolinium as doping element.

Lastly, a systematic study of cerium antimonide (CeSb) magneto-optical properties was performed. The 14° Kerr rotation first discovered by Reim et al. [4] late in the 1980s and the even more astonishing 90° Kerr rotation found in the same compound by Pittini et al. [5] ten years later (over a broader spectral range) caused much excitement. We were motivated first of all to reproduce the giant 90° Kerr rotation on other specimens: besides the general need for reproducibility, we wanted to establish sample independence and the intrinsic nature of this effect. Second, since Pittini et al. found the 90° Kerr rotation at a magnetic field of 5 T, we wanted to investigate what happens to the ‘resonance’ upon increasing the magnetic field up to 10 T. The latter aspect is particularly intriguing if the 90° Kerr rotation is confirmed to be truly an intrinsic effect. The question was whether above 5 T the Kerr rotation stays constant and saturates to such a maximum value or eventually decreases again.

Before discussing the experimental results (Chapter 5), some of the physical properties of the various materials of interest for the present thesis and the theoretical fundaments of optical and magneto-optical spectroscopies are briefly summarised (Chapter 2 and Chapter 3). Chapter 3 shows also how, in this work,
the Drude-Lorentz model is developed to explain magneto-optical results. Chapter 4 reviews the optical and magneto-optical experimental set-up.

REFERENCES


2. PHYSICAL PROPERTIES

This chapter reviews some of the physical properties of the various materials of interest for the present thesis. The choice of physical properties was made in view of the discussion of the optical and magneto-optical investigation results (Chapter 5).

2.1 LaX (X = S, Se, Te)

The three lanthanum monochalcogenides, i.e. LaS, LaSe and LaTe, crystallise in the NaCl structure with the lattice parameters given in Tab. 2.1 and are characterised by a ionic-metallic type bonding [1-7]. In a first approximation, the chemical bonding scheme may be represented as follows: the metal atom gives up two 6s valence electrons (as in ionic bonding) to a chalcogen atom, while the 5d valence electron is delocalised. In the crystal, the 5d state forms an unfilled band, which gives rise to the metallic component of the bonding. Thus lanthanum monochalcogenides may be considered to be “monovalent” metals. Lanthanum itself is a typical trivalent metal [8].
2. Physical Properties

<table>
<thead>
<tr>
<th>Latt. Const. (Å)</th>
<th>$T_c$ (K)</th>
<th>$\gamma$ (mJ/K$^2$mol)</th>
<th>$\Theta_D$ (K)</th>
<th>$\chi_p$ (10$^{-5}$ emu/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LaS</td>
<td>5.856</td>
<td>0.84</td>
<td>3.28</td>
<td>276</td>
</tr>
<tr>
<td>LaSe</td>
<td>6.060</td>
<td>1.02</td>
<td>3.77</td>
<td>231</td>
</tr>
<tr>
<td>LaTe</td>
<td>6.421</td>
<td>1.48</td>
<td>4.65</td>
<td>175</td>
</tr>
</tbody>
</table>

Tab. 2.1: Summary of data on the lanthanum monochalcogenides.

![Graph of resistivity vs. temperature](image)

Fig. 2.1: Temperature variation of lanthanum monochalcogenides resistivity [4].

All these systems have a metallic character (Fig. 2.1, from Ref. [4]) with a carrier density in the range 2.0×10$^{22}$ cm$^{-3}$ [9], and are superconductors, having transition temperatures in the range 0.8 - 1.5 K (Tab. 2.1).

Magnetic susceptibility measurements indicate that conduction electron paramagnetism is dominant [10]. The Pauli paramagnetic susceptibility ($\chi_p$) of the
Physical Properties II

Conduction electrons has been found to increase from LaS to LaTe, suggesting that the density of states at Fermi energy \( n(E_F) \) should also increase from LaS to LaTe. The measured electronic specific heat coefficient (\( \gamma \)) also shows the same trend (Tab. 2.1) [3].

Lastly, the Debye temperatures (\( \theta_D \)) of these compounds decrease from LaS to LaTe (Tab. 2.1) [3] and this behaviour may be attributed to the following factors which are related to each other:

a) From S to Te, electronegativity decreases and hence the La-X bond strength should decrease from LaS to LaTe.

b) From S to Te, the atomic radius increases. Hence, interatomic spacing may be expected to increase from LaS to LaTe, causing the Debye temperature to decrease from LaS to LaTe.

**Fig. 2.2:** Band structure of LaS [1].
The band structure calculation shown in Figs. 2.2, 2.3 and 2.4 [1] was performed using the linear muffin-tin orbital method [11] within the atomic sphere approximation (LMTO-ASA). The main results are reported here. As the valence electron configurations of these systems are similar, their band structures show only small differences.

The LaS band structure has a metallic character with a large contribution to the \( n(E_F) \) coming from the La atom. The S-3p orbitals are separated from the conduction band by a gap of 1.09 eV (0.08 Ry), showing reasonably localised behaviour, and the S-3s orbitals lie below the S-3p orbitals. These features are clearly seen in Fig. 2.2 and Fig. 2.3, in which the calculated band structures and corresponding density of states (DOS) are shown. Fig. 2.2 shows that the band structure possesses three prominent sections, in which the lowest portion is due to the S-3s orbitals; the next three bands are due to hybridisation of La-6s and 5d with the S-3p orbitals. However, the primary contributions to these bands are due to the S-3p orbitals. The conduction band contains only one electron per primitive cell.

In the case of LaSe, the larger value of the lattice parameter compared with LaS and the smaller electronegativity of Se with respect to S reduces the strength of La-5d Se-4p hybridisation. The calculation shows that, in the case of LaSe, the conduction band (CB) is smaller than that of LaS, suggesting a weaker hybridisation in LaSe than in LaS. Further, the effect of this weaker hybridisation is also seen in the \( n(E_F) \): the La-5d bands are narrower than in LaS, causing a larger DOS in the case of LaSe (Fig. 2.4).

For LaTe, the lattice parameter is larger than that of LaSe, due to the larger size of the Te atom. But the electronegativity difference of this system is smaller

---

*Fig. 2.3: The partial and the total densities of states for LaS [1].*
than that of LaSe. Hence, considering these two factors, it may be concluded that La-Te hybridisation is much weaker than that of both LaS and LaSe. The band structure calculation, taken together with the above factors, demonstrates that it is in good agreement with the expected trends: the CB is smaller than that of LaSe and \( n(E_F) \) also increases (Fig. 2.4), following the experimentally observed trends with regard to \( \chi_p \) and \( \gamma \).

In conclusion, it has been shown that the widths of the \( p \)-band of \( X \) as well as the \( d \)-band of La decrease from LaS to LaTe, revealing weakening \( p-d \) hybridisation from LaS to LaTe. This in turn may be attributed to the increase of the DOS at \( E_F \). This increase in \( n(E_F) \) may very well explain the experimentally observed increase of \( \gamma \) and \( \chi_p \), as well as \( T_C \), from LaS to LaTe.

### 2.2 GdS

Gadolinium sulphide (GdS) crystallises in the fcc rocksalt structure and is an antiferromagnetic (AFM) metal with a carefully determined Néel temperature (\( T_N \)) of 62 K [12].

Several authors [13-15] have extensively studied its transport properties. Resistivity, measured as a function of temperature, shows typical metallic behaviour, with a progressive almost linear decrease with lowering temperature. At 50 - 60 K, a shoulder-like feature in the resistivity curves has been found. Some measurements on several samples with different stoichiometries were performed and the results are summarised in Fig. 2.5 [13]. The curves are ordered systematically with composition, i.e., carrier concentration increases in the lower
curves. However, one curve had to be shifted in order to obtain this sequence, the reason being that the sample with the largest carrier concentration (GdS$_{0.88}$) also had the greatest density of anion vacancies. Thus, the reduction in mobility due to defect scattering overcompensates the increase in carrier concentration.

![Graph of resistance vs. temperature for different GdS compositions](image)

**Fig. 2.5:** Temperature variation of Gd$_x$S and Gd$_y$S resistivity with different values of $x$ and $y$ [13].

For samples with $N_{el}/N_{Gd} \geq 1$, i.e., with vacancies in the anion lattice, there is a kink or peak near $T_N$, but for vacancies in the cation sublattice (Gd$_{0.94}$S) and $N_{el}/N_{Gd} \leq 1$, there is a maximum in the resistivity which has nothing to do with the Néel temperature. It thus becomes clear that, besides the scattering mechanisms typical of a magnetic material, such as defect, phonon and spin-disorder scattering, we have an additional scattering term, which only appears if there are vacancies in the Gd sublattice. This term is caused exclusively by deviations in the real antiferromagnet Gd$_{0.94}$S from the ideal antiferromagnet GdS. By constructing a
complementary lattice containing vacancies at the positions which are occupied in the real lattice and statistically distributed localised spins corresponding to the Gd vacancies in the real lattice. Nolting [16] performed experimental quantitative fits. The importance of this result was the demonstration that cation vacancies in a magnetic compound may completely dominate the scattering mechanisms of electrons in a metal.

The magnetic structure of GdS has been determined by inelastic neutron scattering in spite of the high absorption cross-section [17]. At 4.2 K an $AF_2$ spin structure is observed. Magnetic susceptibility ($\chi$) measurements strongly depend on stoichiometry [15,18-20] and the extracted values for $T_N$ differ from one sample to the other by sometimes more than 100 K. Curiously, in general not one but two anomalies in the $\chi(T)$ curves has been found. The first occurs between 40 and 70 K and is attributed to the transition of GdS into the AFM phase. The second occurs at 20 - 30 K and strongly depends on magnetic field and carrier concentration. Of course, it is possible that the $AF_2$ magnetic structure is preceded at $T_N$ by another structure and that the two peaks in $\chi$ are indicative of two magnetic phases of the $4f$ moments. Instead, if the $5d$ conduction electrons are only weakly coupled to the $4f$ system, the $5d$ matrix may exhibit magnetic order within itself, especially if the $5d$ electrons are considered more localised than the $s$ electrons. In any case, the question raised by the experiments cannot yet be answered.

Much work, both theoretical, by means of band structure calculations [21-23] and experimental, with optical reflectivity measurements [9,14,24], X-ray spectral investigation [25] and photoemission [26-28], has been devoted to the understanding of the electronic structure of gadolinium monochalcogenides. In the particular case of GdS, the schematic band picture is shown in Fig. 2.6 [15]. The conduction band is made up of the $5d$ Gd orbitals and lies below the band which originates from the

![Fig. 2.6: GdS electronic band structure scheme [15].](image-url)
6s orbitals. The conduction band is filled up to 1.2 - 2 eV with a maximum in the density of states near the Fermi level. The uppermost valence band is formed of the $p$ orbitals of the sulphide. Photoemission measurements [26-28], together with optical reflectivity data [9,14,24], clearly show that the localised $4f$ states lie between 7 and 9 eV, under the Fermi level.

2.3 Eu(Gdx)O

Europium chalcogenides, which all crystallise in the rocksalt structure, are an ideal system for the investigation of magnetic interactions. First, chalcogenides series contains all kinds of fundamental magnetic order. Second, these materials are nearly ideal Heisenberg magnets. Third, the undoped compounds are insulators or semiconductors. However, by suitable doping, one can introduce free carriers and thus study additional magnetic interactions. This session focuses on the main physical properties of the europium oxide compound (EuO) and on the effects produced by the introduction of a small amount of gadolinium (Gd) as dopant.

EuO is a ferromagnetic semiconductor ($E_g = 1.12$ eV at room temperature) with a Curie temperature ($T_C$) of 69 K [29]. The value of 69 K has been determined very accurately by both magnetic susceptibility ($\chi$) and specific heat ($C_p$) measurements [30,31]. However, many authors have pointed out that $T_C$ strongly depends on the stoichiometry of the various measured samples and particularly on: a) oxygen stoichiometry (EuO$_{1+x}$) [29,31]; and b) doping (i.e. EuGd$_x$O) [29,32]. The results may be summarised stating that higher values of $T_C$ are found in non-stoichiometric as well as in Gd-doped samples. The highest value is $T_C = 135$ K, found in EuGd$_{0.38}$O with $x = 3.4\%$ [32].

Shapira et al. [29] performed an accurate analysis of the transport properties of several EuO samples coming from different growth and prepared in different ways. The results of this research and of others in the literature [33-34] are summarised in Fig. 2.7. We observe that:

- Going from high to lower temperatures, the resistivity of all the investigated samples remains almost constant between 300 and 100 K. At the critical temperature $T_C$, there is a peak in $\rho(T)$ which then drops down by several orders of magnitude (as in case of sample Eu$_{1+y}$O), showing metallic behaviour at low temperatures.
- There is a strong sample-dependence: in particular, Gd-doped samples are characterised by much lower resistivity throughout the temperature range
and reduced drop at the critical temperature with respect to the nearly stoichiometric ones.

- Comparing the critical temperatures of several investigated samples, higher values are found for non-stoichiometric and doped samples with respect to pure EuO.

![Graph showing temperature variation of the zero-field resistivity of EuO samples](image)

**Fig. 2.7:** Temperature variation of the zero-field resistivity of some nearly stoichiometric and Gd-doped EuO samples [29,33-34].

The effects of applying an external magnetic field to the transport properties are again discussed in Ref. [29]. Here we merely observe that:

- In the high-temperature range, there is a progressive reduction in resistivity, up to some orders of magnitude, in some samples when the intensity of the externally applied magnetic field is increased.
- A relatively small increase in critical temperature is found.
In the low-temperature range, resistivity is almost independent of magnetic field.

Lastly, it should be noted that the application of external pressure produces drastic and fascinating changes in the physical properties of EuO. First, there is a general increase in $T_C$ with increasing pressure up to 90 kbar [35]. Second, resistivity as a function of pressure measured at room temperature shows a considerable drop at around 130 kbar and a semiconductor-metal transition at around 300 kbar [36-38]. Third, measurements of the pressure-volume relationship [39] have shown a valence transformation around 300 kbar, followed by a NaCl-to-CsCl-type transition around 400 kbar.

The common explanation for this great variety of phenomena is the following: by increasing pressure, we move from a region ($P < 130$ kbar) where $4f^7$-$5d$ gap closing is dominated by the crystal field downshift of the $5d$ band to one ($P > 150$ kbar), in which a hybridisation-gap mixed-valent semiconductor is stabilised. At higher pressures, around 300 kbar, volume drops abruptly but there is no change in structure. This volume decrease is ascribed to a change in the valence state of the Eu ion from divalent to trivalent, due to $4f$-$5d$ electron promotion. This $4f$-to-$5d$ electron promotion-delocalisation also explains the semiconductor-to-metal transition, which occurs at the same value of applied pressure.

![Fig. 2.8: EuO electronic band structure scheme [44].](image)

Without going into the details of a band-structure calculation of EuO, let us review the essential features of an energy-level scheme on which all investigators agree [40-43].

The highest occupied valence band is formed of $p^6$ states of the oxygen. Localised and partially occupied $4f^7(\Sigma S_{7/2})$ states of Eu$^{2+}$ are situated above this band and, at still higher energies, conduction bands with $5d$ and $6s$ symmetry are
found (Fig. 2.8, from Ref. [44]). \( n \)-type doping introduces donor levels somewhat below the bottom of the conduction band. If Gd is chosen as the donor and if \( \text{Gd}^{3+} \) substitutes for \( \text{Eu}^{2+} \), the donor is a shallow level (~0.017 eV) with \( 4f^7 5d^1 \) symmetry [45]. However, it is important to note that, with the incorporation of Gd, a usually uncontrolled change in the anion lattice, such as the formation of vacancies, may develop.

Thermal ionisation of shallow donors yields free conduction electrons and, at high dopant concentrations, impurity bands form and the Fermi level moves into the conduction band.

### 2.4 CeSb

Cerium antimonide (CeSb) crystallises in the rocksalt structure and is a paramagnetic metal with a Néel temperature of 16.2 K [46]. Ce-Sb bonding is mainly ionic, in which cerium appears as a formally trivalent cation. Resistivity as a function of temperature shows Kondo behaviour, with a drop of almost two orders of magnitude at ~25 K (Fig. 2.9) [46-48].

The magnetic order of CeSb is very complicated. The magnetic moments are tied to the [100] axis (Ising model), despite the symmetry of the \( \Gamma_7 \) ground state of the \( 4f \) level. Magnetisation measured along any other direction is the corresponding component of magnetisation along [100]. Moreover, only the [100] component of the external field induces magnetisation in CeSb. This very particular situation is shown in Fig. 2.10, in which the low-temperature magnetisation of CeSb is measured with external fields along [100], [110] and [111].

The calculated magnetisation curves shown in Fig. 2.10 are based on the Ising model [12,49]. For an increasing external field applied along [100], the Ising
model reproduces a fully saturated ionic moment above 3.8 Tesla (T), a ferrimagnetic $++-$ order corresponding to one-third of the saturated magnetisation between 0.7 T and 3.8 T and antiferromagnetism below 0.7 T. The step in the magnetisation at 2.2 T (Fig. 2.10) represents saturation of the sublattice moment. Following Busch et al. [49], between 0.7 T and 2.2 T the sublattice moment takes the value of 0.71 $\mu_B$ corresponding to crystal field ground state $\Gamma_7$.

**Fig. 2.10**: Magnetisation curves of CeSb measured at 1.5 K in external fields applied along [100], [110] and [111] directions [49].

Below $T_N$, CeSb loses its cubic symmetry and becomes slightly distorted tetragonally. Lattice parameter $c$ becomes 0.12 % shorter than $a$ [50]. Kanamori [51] has shown that the Ising model allows for a magnetic structure characterised by stacking of antiferromagnetically or ferromagnetically ordered [001] planes in tetragonal crystals. Neutron scattering confirmed this particular magnetic ordering in ferromagnetic layers in CeSb [52]. Anisotropic interactions are responsible for a very strong exchange within the planes, while exchange between the planes along [100] is weak. It was also shown that, above 8 K, the ferromagnetic planes are periodically stacked with non-magnetic layers [52]. Figure 2.11 shows the complex phase diagram of CeSb worked out by Rossat-Mignod et al. [52] through combined analysis of magnetisation measurements and neutron scattering data.
The CeSb electronic structure may be described as follows: the nominally anionic $p$ bands mainly lie below the Fermi level, and the cationic $d$ bands are mainly situated just above $E_F$. The top of the valence $p$ band at the $\Gamma$ point and the bottom of the conduction $d$ band (at the $X$ point of the Brillouin zone) overlap, making CeSb semimetallic. By electroneutrality, the number of conduction $d$ electrons is the same as the number of holes in the $p$ band. Of course, this delicate charge balance strongly depends on the relative energy positions of the top of the valence $p$ band and the bottom of the conduction $d$ band.

The $f$ states are energetically situated in the anionic valence $p$ band. Their spatial extent is strictly limited to the cation site, and the direct $f$-$f$ overlap of the atomic wavefunctions may be neglected. However, the $f$ states are moderately delocalised through $f$-band hybridisation processes [53]. It is this weak delocalisation of $f$ states which causes the fascinating and unusual properties of CeSb.
REFERENCES

2. Physical Properties


3. THEORY

This chapter briefly summarises the theoretical fundamentals of optical and magneto-optical polar Kerr effect spectroscopy. A detailed derivation of all the quantities of interest goes beyond the scope of this work, but may be found in Refs. [1-7].

3.1 Optical Spectroscopy

Optical spectroscopy is a powerful experimental technique for investigating the complete excitation spectrum of a medium over a very broad spectral range. Light reflected at normal incidence from an ideal surface can be derived quite simply from the electric and magnetic field boundary conditions at the interface of the solid surface with vacuum [1,2].
3. Theory

3.1.1 Definitions

Under the action of an electromagnetic wave, the dielectric response of a medium is described by the Maxwell equations (neglecting the non-linear effects of the material response) \([3,7]\):

\[
\begin{align*}
\nabla \cdot \mathbf{D} & = 0 \\
\nabla \cdot \mathbf{B} & = 0 \\
\n\nabla \times \mathbf{E} & = -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t} \\
\n\nabla \times \mathbf{H} & = \frac{1}{c} \left( \mathbf{D} + 4\pi \mathbf{J} \right) = \frac{1}{c} \mathbf{D}
\end{align*}
\]

where:

\[
\begin{align*}
\mathbf{D} & = \mathbf{E} + 4\pi \mathbf{P} \\
\mathbf{B} & = \mathbf{H} + 4\pi \mathbf{M}
\end{align*}
\]

and

\[
\begin{align*}
\mathbf{P} & = \alpha \mathbf{E} \\
\mathbf{M} & = \chi \mathbf{H} \\
\mathbf{J} & = \sigma \mathbf{E}
\end{align*}
\]

The dielectric constant and magnetic permeability are, in isotropic or cubic materials, defined by:

\[
\begin{align*}
\varepsilon_1 & = 1 + 4\pi \alpha_1 \\
\mu_1 & = 1 + 4\pi \chi_1
\end{align*}
\]

where index '1' indicates the real part of the corresponding function. For the time and space dependence of the electric and magnetic field vector, the plane-wave form (\(\mathbf{g}\) is the complex wave vector)

\[
\begin{align*}
\mathbf{E}(t, r) & = E_0 e^{i(\omega t - \mathbf{g} \cdot \mathbf{r})} \\
\mathbf{B}(t, r) & = B_0 e^{i(\omega t - \mathbf{g} \cdot \mathbf{r})}
\end{align*}
\]

will be used in this work. So far, the sign of the phase in eq. (3.4) is arbitrary, but its choice will determine the signs in many following definitions of optical and magneto-optical quantities. Wave vector \(\mathbf{g}\) is proportional to the complex index of refraction. With the sign of the wave vector as in eq. (3.4), the complex index of refraction is written as:

\[\tilde{n} = n - ik\]
where \( k \) represents optical absorption and \( n \) governs the spatial dependence of the phase and is the dispersive part of the complex index of refraction.

At optical frequencies, the magnetic permeability may be set at 1 (\( \mu = 1 \)) even for ferromagnets (i.e. magnetic dipole moments cannot be oriented in the \( H \) field of light because of its quick oscillations [8,9]). With the complex dielectric function and complex optical conductivity defined as:

\[
\tilde{\varepsilon} = \varepsilon_1 - i\varepsilon_2 \\
\tilde{\sigma} = \sigma_1 + i\sigma_2
\]  

(3.6)

one obtains the relations:

\[
\begin{align*}
\varepsilon_1 &= n^2 - k^2 \\
\varepsilon_2 &= 2nk \\
n &= \sqrt{\frac{\varepsilon_1^2 + \varepsilon_2^2 + \varepsilon_1}{2}} \\
k &= \sqrt{\frac{\varepsilon_1^2 + \varepsilon_2^2 - \varepsilon_1}{2}}
\end{align*}
\]  

(3.7)

It can also be shown that, between the real and imaginary parts of \( \tilde{\varepsilon} \) and \( \tilde{\sigma} \), the following equations hold:

\[
\begin{align*}
4\pi\sigma_1 &= \omega\varepsilon_2 \\
4\pi\sigma_2 &= \omega(\varepsilon_1 - 1)
\end{align*}
\]  

(3.8)

The complex optical reflectance is now defined by:

\[
\tilde{r} = \rho \cdot e^{i\delta} = \frac{n - 1}{n + 1}
\]  

(3.9)

(Fresnel equations) where \( \delta \) is the phase and \( \rho \) the reflectance amplitude. Consequently reflectivity \( R = \tilde{r} \cdot \tilde{r}^* \), which is the measured quantity, is related to \( n \) and \( k \) as:

\[
R = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2}.
\]  

(3.10)
3. Theory

3.1.2 Kramers-Kronig Transformations

From the preceding equations it is evident that knowledge of reflectivity alone is not sufficient to compute the optical functions (i.e. $n$ and $k$), which are the important quantities in order to discuss the optical spectra. However, the real and imaginary parts of the reflectance, or equivalently reflectivity $R$ and phase $\delta$, are related to each other through some dispersion relations known as Kramers-Kronig transformations.

Kramers-Kronig transformations are based on the principle of causality. Causality means that there can be no effect before a cause. The cause in optical phenomena is the incident electromagnetic wave, and the effect is the motion of electrons and nuclei. Thus, light cannot be reflected or absorbed by a system before the arrival of the primary light wave. Following Ref. [1], the relation between phase $\delta$ and reflectivity $R$ is:

$$\delta(\omega) = \frac{\omega}{\pi} \int_{0}^{\infty} \frac{\ln \left[ R(\omega')/R(\omega) \right]}{\omega - \omega'} d\omega'. \quad \text{(3.11)}$$

Equation (3.11) implies that we know the reflectivity from zero to infinity, which is far beyond experimental limits. We overcome this problem by extrapolating our data at both higher and lower energies. For energies larger than the highest measurable energy, reflectivity is assumed to drop off as $\omega^{-s}$ where $s$ varies from 2 (to simulate interband transitions) to 4 (describing the free electron asymptotic limit). From the lowest measurable point down to zero, the extrapolation depends on the physical properties of the investigated system. In the case of an insulator, reflectivity is extrapolated to a constant value. For a conducting material, with the condition $\omega \ll \Gamma$ ($\Gamma$ being the scattering rate of the free electrons, see § 3.1.3), we use the Hagen-Rubens extrapolation:

$$R(\omega) = 1 - \sqrt{\frac{8\varepsilon_{0}\omega}{\sigma_{dc}}} \quad \text{(3.12)}$$

where $\sigma_{dc}$ is dc conductivity.

When phase and reflectivity are both known, we can calculate the dielectric function:
\[\varepsilon_1(\omega) = \frac{(1 - R(\omega))^2 - 4R(\omega)\sin^2 \delta(\omega)}{(1 + R(\omega) - 2\sqrt{R(\omega) \cos \delta(\omega)})^2}\]  
\[\varepsilon_2(\omega) = \frac{4(1 - R(\omega))\sqrt{R(\omega) \sin \delta(\omega)}}{(1 + R(\omega) - 2\sqrt{R(\omega) \cos \delta(\omega)})^2}\]  

(3.13)

and then, with eqs. (3.7) and (3.8), the other optical functions.

### 3.1.3 The Lorentz-Drude Model

The Lorentz-Drude model is a simple phenomenological approach based on the classical dispersion theory, describing the dielectric response of a medium under the action of an electromagnetic wave in terms of harmonic oscillators (h.o.) [1]. The basic concept is that an electron bound to the nucleus may be described in the same way as a small mass bound to a large one by a spring (Lorentz model). In this way, the motion of an electron bound to the nucleus is described by:

\[
m_j^* \frac{d^2 r_j}{dt^2} + m_j^* \gamma_j \frac{dr_j}{dt} + m_j^* \omega_j r_j = eE
\]  

(3.14)

where \(m_j^*\) is the electronic effective mass, \(e\) the electronic charge and \(E\) the local electric field. The term \(m_j^* \gamma_j (dr_j/dt)\) represents viscous damping and provides for an energy loss mechanism; \(m_j^* \omega_j r_j\) is the restoring force of the Hook’s law. The small force \(-ev_j \times B/c\) arising from the interaction of the electron with the magnetic field of the light wave is negligible, because the velocity of the electron is small compared with \(c\).

With a periodic time-dependent electric field as in eq. (3.4) (plane-wave form), one can obtain for the optical conductivity:

\[
\tilde{\sigma}(\omega) = \frac{\omega_{ps}^2}{4\pi} \frac{i\omega}{\omega_j^2 - \omega^2 + i\gamma_j \omega}
\]  

(3.15)

where:

\[
\omega_{ps} = \sqrt{\frac{4\pi N_e e^2}{m_j^*}}.
\]  

(3.16)
In eqs. (3.15) and (3.16), \( \omega_p \) is the resonance frequency, \( \gamma \) the damping parameter, \( \omega_{pj} \) the mode strength of the harmonic oscillator and \( N_j \) the density of electrons bound with resonance frequency \( \omega_p \). Since in general there are many possible excitations, eq. (3.15) may be generalised in the following way:

\[
\tilde{\sigma}(\omega) = \frac{1}{4\pi} \sum_j \omega_{pj}^2 \frac{i\omega}{\omega_j^2 - \omega^2 + i\gamma_j \omega} \tag{3.17}
\]

with the condition \( \sum_j N_j = N \), where \( N \) is the number of electrons per unit volume.

Equation (3.17) also has a corresponding quantum mechanical expression [3]:

\[
\tilde{\sigma}(\omega) = \frac{1}{4\pi} \sum_j \omega_{pj}^2 f_j \frac{i\omega}{\omega_j^2 - \omega^2 + i\gamma_j \omega} \tag{3.18}
\]

with the sum rule:

\[
\sum_j f_j = 1. \tag{3.19}
\]

There is a formal similarity between eq. (3.17) and eq. (3.18), but the meaning of some corresponding terms is quite different. For example, in eq. (3.17), \( \omega_p \) is the resonance frequency of the bound electron, whereas in eq. (3.18) it is the transition frequency of the electron between two atomic states, separated in energy by \( \hbar \omega_j \) (interband transition). The parameter \( f_j \), called oscillator strength, is the relative probability of a quantum mechanical transition (§ 3.2.3 and § 3.2.5).

The Drude model describes the behaviour of the free electrons. The approach is exactly the same as in the Lorentz model and, since the electrons are free and not bound to the nucleus, the restoring force in eq. (3.14) is set at zero (\( \omega_p = 0 \)). Equation (3.15) gives:

\[
\tilde{\sigma} = \frac{\omega_p^2}{4\pi} \frac{1}{\Gamma + i\omega} \tag{3.20}
\]

where \( \omega_p \) is the plasma frequency and \( \Gamma \) the damping of the free electrons. The damping is the ordinary scattering rate of electrons associated with electrical
resistivity. We can now write the real part of the optical conductivity as the result of the sum of the free electron contribution (the Drude term representing the \textit{intraband transitions}) and a set of finite frequency excitations (the Lorentz oscillators representing the different \textit{interband transitions}): 

\[
\sigma_1(\omega) = \frac{1}{4\pi} \left( \frac{\omega_p^2 \Gamma}{\omega^2 + \Gamma^2} + \sum \omega_j^2 \frac{\omega_j^2 \gamma_j}{(\omega_j^2 - \omega^2)^2 + \omega_j^2 \gamma_j^2} \right). \tag{3.21}
\]

\textbf{Fig. 3.1:} Typical reflectivity curves. The parameters of the metal (plain line) are: \( \hbar \omega_p = 2 \text{ eV} \) and \( \hbar \Gamma = 0.04 \text{ eV} \). For the insulator (dotted line), the absorption at resonance frequency \( \hbar \omega_i = 0.8 \text{ eV} \) has strength \( \hbar \omega_{pi} = 8 \text{ eV} \) and half-width \( \hbar \gamma_i = 1 \text{ eV} \) [10].

Figure 3.1 shows how the typical reflectivity curves of a metal (plain line) and an insulator (dotted line) look alike [10]. For a metal, reflectivity increases to 100\% as energy tends to zero and the steep decrease at 2 eV indicates plasma energy \( \hbar \omega_p \). For an insulator, reflectivity at low energies reaches a constant value, which depends on higher energy absorptions.
Optical conductivities obtained with the same parameters used to calculate $R$ are shown in Fig. 3.2. In a metal (plain line) at low energy, $\sigma_1$ reaches its $dc$ value, which corresponds to the value obtained with resistivity measurements. The area under the curve is proportional to the plasma energy $\hbar \omega_p$ and the width is the scattering rate $\hbar \Gamma$. In an insulator, the position of the maximum at 0.8 eV indicates the transition resonance frequency, the area under the curve is proportional to the mode strength $\hbar \omega_{\text{qj}}$ and the full width at half maximum is the damping $\hbar \gamma$. Since no free electrons are present in the insulator, there is no Drude term and the optical conductivity tends to zero as energy also approaches zero.

**Fig. 3.2:** Typical conductivity curves. Optical conductivities are obtained with the same parameter set of Fig. 3.1 [10].
3.2 Magneto-Optical Kerr Effect Spectroscopy

Magneto-optical Kerr effect spectroscopy is an ellipsometric type of spectroscopy which measures the characteristics of the axial anisotropy induced by sample magnetisation.

3.2.1 Definitions

An externally applied magnetic field acts on a sample as a source of an axial anisotropy with the axis along the field direction. These premises are the same, both in a magneto-optical experiment and in the measurement of the Hall effect. The latter is considered as the 'static' limit of magneto-optics, in the same way as dc conductivity is for optical experiments (eq. (3.12)).

In the geometry of the polar Kerr effect (Fig. 3.3), i.e. for a light beam travelling parallel to the external field (\( \mathbf{g} \parallel \mathbf{H} \)), the polarisation eigenstates corresponding to the axial anisotropy of the magnetised sample are the circular polarisations. A different optical absorption for the two circular polarisations leads to a change in light polarisation, which may be expressed by two quantities, the azimuthal polarisation rotation \( \theta_k \) and the polarisation ellipticity \( \varepsilon_k \) (Fig. 3.4).
A circularly polarised wave reflected by a medium with index of refraction \( \tilde{n} \) and propagating along the negative direction of the \( z \)-axis is described by the electrical field vector:

\[
E_x(t, z) = E_0 e^{i(\omega t - 2\pi \tilde{n} z / \lambda_0)} (e_x \pm e_y)
\]

(3.22)

where upper + and lower \(-\) refer to right (RCP) and left (LCP) hand circular polarisations, \( \lambda_0 \) is the wavelength in vacuum, \( \tilde{n}_z \) is the complex refraction index for the two states of polarisation and \( e_x \) and \( e_y \) are unit vectors in \( x \)- and \( y \)-directions, respectively.

**Fig. 3.4:** Polarisation plane for the polar magneto-optical Kerr effect with a rotation of 30 degrees and an ellipticity of 10 degrees [7].

With these definitions, it may be shown that, without any approximation [4]:

\[
\theta_K = -\frac{1}{2} (\delta_+ - \delta_-)
\]

\[
\varepsilon_K = \arctan\left(-\frac{\rho_+ - \rho_-}{\rho_+ + \rho_-}\right)
\]

(3.23)
where $\rho_+, \rho_-, \delta_+$ and $\delta_-$ are defined as in eq. (3.9) and are related to the complex index of refraction through the following equations:

$$
\delta_+ = \arctan \left( -\frac{2k_+}{n_+^2 + k_+^2} \right) \quad (3.24)
$$
$$
\rho_+ = \frac{\sqrt{(n_+^2 + k_+^2 - 1)^2 + (2k_+)^2}}{(n_+ + 1)^2 + k_+^2}. \quad (3.25)
$$

### 3.2.2 Experimental Determination of the Conductivity Tensor

In isotropic systems, complex optical functions $\tilde{\varepsilon}$ and $\tilde{\sigma}$, defined in § 3.1.1, are scalars, but in anisotropic materials they become tensors of the second rank. After the introduction of an external magnetic field $H$ oriented, for instance, along the preferential $z$ direction ($H = H_z$), the tensors are no longer diagonal, even in cubic or isotropic media. In fact, off-diagonal elements appear in the plane perpendicular to the magnetisation, i.e. the $xy$-plane. From general symmetry considerations and for a cubic system, it follows that conductivity tensor $\tilde{\sigma}$ may be defined as [4]:

$$
\tilde{\sigma} = \begin{pmatrix}
\tilde{\sigma}_{xx} & \tilde{\sigma}_{xy} & 0 \\
-\tilde{\sigma}_{xy} & \tilde{\sigma}_{xx} & 0 \\
0 & 0 & \tilde{\sigma}_{zz}
\end{pmatrix} \quad (3.26)
$$

where $\tilde{\sigma}_{ij} = \sigma_{ij} + i\sigma_{ij}$ are complex functions. Decomposing eq. (3.26) into a Hermitian and an anti-Hermitian contribution [11], it may be shown that the absorptive parts of the diagonal and off-diagonal elements are $\sigma_{1xx}$ and $\sigma_{2xy}$, respectively. Because $\tilde{\sigma}_{ij}(H) = \tilde{\sigma}_{ij}(-H)$, the diagonal and off-diagonal tensor elements are respectively even and odd functions of magnetisation [12]. Furthermore, it may be shown that the linear components of the conductivity tensor are related to the circular ones through:

$$
\tilde{\sigma}_z = \tilde{\sigma}_{xx} \pm i\tilde{\sigma}_{xy}
$$

and thus:

$$
\tilde{\sigma}_{xx} = \frac{(\tilde{\sigma}_+ + \tilde{\sigma}_-)}{2} \quad \text{and} \quad \tilde{\sigma}_{xy} = \frac{(\tilde{\sigma}_+ - \tilde{\sigma}_-)}{2i}. \quad (3.27)
$$

Even for Kerr rotations and ellipticities of several degrees, the following approximations hold:
\[
\begin{align*}
\sin(\delta_+ - \delta_-) & \approx \delta_+ - \delta_- \\
\cos(\delta_+ - \delta_-) & \approx 1 \\
(\rho_+ - \rho_-)^2 & \ll 2\rho_+\rho_-
\end{align*}
\] (3.28)

With (3.28), substituting eq. (3.9) into eq. (3.23), we find the following relations between the complex indices of refraction and the Kerr rotation and ellipticity:

\[
\theta_K = -\text{Im} \left( \frac{\tilde{n}_+ - \tilde{n}_-}{\tilde{n}_+\tilde{n}_- - 1} \right) \] (3.29)

\[
\varepsilon_K = -\text{Re} \left( \frac{\tilde{n}_+ - \tilde{n}_-}{\tilde{n}_+\tilde{n}_- - 1} \right) \] (3.30)

It is straightforward to relate \( \tilde{n}_\pm \) and the conductivity tensor by inserting eqs. (3.27) and (3.8) into eqs. (3.29) and (3.30):

\[
\tilde{n}_+^2 - \tilde{n}_-^2 = -\frac{8\pi}{\omega} \tilde{\sigma}_{xy} \] (3.31)

\[
\frac{\tilde{n}_+ - \tilde{n}_-}{\tilde{n}_+\tilde{n}_- - 1} = i\tilde{\sigma}_{xx}\tilde{n} = \frac{\tilde{\varepsilon}_{xy}}{i(\tilde{\varepsilon}_{xx} - 1)\sqrt{\tilde{\varepsilon}_{xx}}} \]

where:

\[
\tilde{n} = \frac{1}{2}(\tilde{n}_+ + \tilde{n}_-) \] (3.32)

Taking the real and imaginary parts according to eqs. (3.29) and (3.30) yields:

\[
\theta_K = \frac{4\pi}{\omega} \left[ \frac{B\sigma_{1xy} + A\sigma_{2xy}}{A^2 + B^2} \right] \] (3.33)

\[
\varepsilon_K = \frac{4\pi}{\omega} \left[ \frac{A\sigma_{1xy} - B\sigma_{2xy}}{A^2 + B^2} \right] \] (3.34)

with coefficients:

\[
A = (\sigma_{2xy} n - \sigma_{1xy} k) \frac{4\pi}{\omega} = n^3 - 3n k^2 - n \] (3.35)

\[
B = (\sigma_{1xy} n + \sigma_{2xx} k) \frac{4\pi}{\omega} = -k^3 + 3n^2 k - k \] (3.36)
Inversely, we have:

\[
\sigma_{\text{yy}} = -\frac{\omega}{4\pi} \left( B\theta_k + A\epsilon_k \right) \tag{3.37}
\]
\[
\sigma_{\text{xy}} = -\frac{\omega}{4\pi} \left( A\theta_k - B\epsilon_k \right). \tag{3.38}
\]

The last equations make clear that, in the Kerr configuration, it is absolutely necessary to measure all four quantities \( \theta_k, \epsilon_k, n \) and \( k \), in order to obtain reliable information on \( \sigma_{\text{yy}} \) and hence to be able to relate magneto-optics to band structure features which is certainly the ultimate goal.

### 3.2.3 Classical Microscopic Model

In the phenomenological Lorentz model, the motion equation of the electrons in the presence of an externally applied magnetic field \( H \) is given by [6]:

\[
m_j \frac{d^2 r_{\pm}}{dt^2} + m_j \gamma_j \frac{dr_{\pm}}{dt} + m_j \epsilon_j r_{\pm} = eE + \frac{e}{c} \frac{dr_{\pm}}{dt} \times H \tag{3.39}
\]

where, with respect to eq. (3.14), we have the additional term on the right side of the equation and \( \pm \) stands for RCP and LCP. In the same way as in § 3.1.3, for the optical conductivity one can obtain:

\[
\tilde{\sigma}_z = \frac{\omega_{\pm}^2}{4\pi} \frac{i \omega}{\omega_j^2 \pm \omega \omega_{\epsilon_j} - \omega^2 + i \gamma_j \omega}
\]

with

\[
\begin{align*}
\omega_{\pm}^2 &= \frac{4\pi N_j e^2}{m_j} \\
\omega_{\epsilon_j} &= \frac{eH}{m_j c}
\end{align*}
\]

(3.40)

Note that the only difference between eq. (3.15) and eq. (3.40) is the presence of the term \( \pm \omega \omega_{\epsilon_j} \), proportional to \( H \), where \( \omega_{\epsilon_j} \) is the cyclotron frequency.

Of course, in this classical model, the effects of selection rules, occupation of states and spin polarisation cannot be taken into account. Only a quantum mechanical description of the solid can introduce these three fundamental features into a description of the optical absorption process. In order to overcome these shortcomings at least partially, we therefore introduce oscillator strengths \( f^\pm \), which multiply \( \omega_{\pm}^2 \) in eq. (3.40). The total circular optical conductivity,
taking into account all optical excitations in the experimental photon energy range, may be written as:

\[
\tilde{\sigma}_x = \sum_j f_j^2 \frac{\omega_j^2}{4\pi} \frac{i\omega}{\omega_j^2 \pm \omega \omega_{\epsilon_j} - \omega^2 + i\gamma_j \omega}.
\] (3.41)

For diagonal and off-diagonal optical conductivities in the magnetic field, from eq. (3.27) it follows that:

\[
\tilde{\sigma}_{xx} = \sum_j \frac{i\omega \omega_j^2}{8\pi} \frac{\gamma_j^2 (f_j^+ + f_j^-) (\omega_j^2 - \omega^2 + i\gamma_j \omega) - (f_j^+ - f_j^-) \omega \omega_{\epsilon_j}}{(\omega_j^2 - \omega^2 + i\gamma_j \omega)^2 - (\omega \omega_{\epsilon_j})^2}.
\] (3.42)

\[
\tilde{\sigma}_{xy} = \sum_j \frac{\omega \omega_j^2}{8\pi} \frac{(f_j^+ - f_j^-) (\omega_j^2 - \omega^2 + i\gamma_j \omega) - (f_j^+ + f_j^-) \omega \omega_{\epsilon_j}}{(\omega_j^2 - \omega^2 + i\gamma_j \omega)^2 - (\omega \omega_{\epsilon_j})^2}.
\] (3.43)

**Fig. 3.5:** Energy level scheme for the simplest magneto-optically active transition between the atomic $^2S_{1/2}$ ($S = \frac{1}{2}, L = 0, J = \frac{1}{2}$) and $^2P_{1/2}$ ($S = \frac{1}{2}, L = 1, J = \frac{1}{2}$) states [4,13].

The Drude contribution to $\tilde{\sigma}_{xy}$ is obtained, as in § 3.1.3, by setting $\omega_j$ at 0. Furthermore, we notice that, in the case of equal oscillator strengths for absorption of right and left circular polarised light (i.e. $f_j^+ = f_j^-$ = 1, as we will see in § 3.2.5) and neglecting the field-dependent term in the denominator of eqs. (3.42) and (3.43), the Lorentz model for optics already discussed in § 3.1.3 is recovered for
both diagonal and off-diagonal conductivities. In eqs. (3.42) and (3.43), the contributions proportional to the sum and difference of oscillator strengths \( f_i^+ \) for the two circular light polarisations are separated to differentiate the cases in which the oscillator strengths for absorption of the circular polarisations in the external magnetic field are either equal or different. In the first case \( (f_i^+ = f_i^-) \), we speak of 'diamagnetic' absorption, whereas in the second case \( (f_i^+ \neq f_i^-) \) we speak of 'paramagnetic' absorption. In order to clarify this important distinction, we make use of the following two level simplified model shown in Fig. 3.5 [4,13].

The applied magnetic field splits the atomic levels of the initial and final states (Zeeman splitting) characterised by the momentum quantum numbers \( S-L-J \) into sublevels characterised by the now non-degenerate magnetic quantum numbers \( m_i \). The selection rules for right and left circular polarised light require \( m_i = \pm 1 \), respectively. Thus, the resonance frequency will generally be different for the two circular polarisations, leading to the so-called diamagnetic line-shape, which is shown in Fig. 3.6a for the simplest magneto-optically active transition between the atomic \(^2\text{S}_{1/2} \) (\( S = \frac{1}{2}, L = 0, J = \frac{1}{2} \)) and \(^2\text{P}_{1/2} \) (\( S = \frac{1}{2}, L = 1, J = \frac{1}{2} \)) states. This contribution is independent of magnetisation, in a first approximation. A very different, so-called paramagnetic, line-shape occurs if the occupation of the

---

**Fig. 3.6:** Off-diagonal conductivity diamagnetic a), and paramagnetic b) line-shapes.
Zeeman-split ground states is different (Fig. 3.6b). This leads to a term proportional to magnetisation.

As a final remark, it is important to point out that the real and imaginary parts of $\tilde{\bar{\sigma}}_{xy}$ are Kramers-Kronig correlated [4,14]:

$$\sigma_{1xy}(\omega) = -\frac{2}{\pi} \sum_{\omega'} \sigma_{2xy}(\omega') \frac{\omega}{\omega^2 - \omega'^2} \omega' \Delta \omega'$$

(3.44)

$$\sigma_{2xy}(\omega) = \frac{2\omega}{\pi} \sum_{\omega'} \frac{1}{\omega^2 - \omega'^2} \sigma_{1xy}(\omega') \Delta \omega'.$$

(3.45)

These equations may be used for a rough extrapolation of $\tilde{\bar{\sigma}}_{xy}$ below the lowest energy limit of the measurement. For this purpose, an extrapolation of $\sigma_{1xy}$ is chosen in such a way that eq. (3.45) reproduces $\sigma_{2xy}(\omega)$ in the measured spectral range, and vice versa [15]. Kerr rotation $\theta_K$ and Kerr ellipticity $\varepsilon_K$ are not Kramers-Kronig correlated.

### 3.2.4 The Semiclassical Approach

The semiclassical approach is based on the quantum-mechanical treatment of matter and a classical treatment of electromagnetic radiation.

a) **interband transitions**

If we look at interband transitions as a result of the semiclassical approach, the transition rate for the absorption of circular polarised light is found, from a time-dependent perturbation theory, to be [7]:

$$W_{rr}^\pm = \frac{\pi}{4} \left( \frac{\hbar |E|^2}{\epsilon_m} \right)^2 \omega_{s s}^\pm |M_{s r}^\pm|^2 \left( \delta(\omega_{s s}^\pm - \omega) + \delta(\omega_{s s}^\pm + \omega) \right)$$

(3.46)

where the absorption frequencies for right and left circular polarised light are generally different, because different final or initial states are involved in the two transitions. The first $\delta$ function in eq. (3.46) refers to light absorption, whereas the second $\delta$ function in eq. (3.46) refers to light emission processes. Limiting treatment to light absorption processes (the emission contribution is analogous), it may be shown that [16]:

$$\text{Reduction of } W_{rr}^\pm$$

$$\text{Reduction of } W_{rr}^\pm$$

The reduction of absorption rate is given by:

$$R_{rr}^\pm = \frac{W_{rr}^\pm}{W_{rr}^0}$$

(3.46a)

where $W_{rr}^0$ is the absorption rate for circular polarised light without magnetic fields. The reduction of absorption rate is due to magnetic field effects and can be expressed as:

$$R_{rr}^\pm = \frac{1}{2} \left( 1 - \frac{\hbar \omega}{\omega_{s s}^\pm} \left( \frac{\hbar |E|^2}{\epsilon_m} \right)^2 |M_{s r}^\pm|^2 \right)$$

(3.46b)

The reduction of absorption rate is a function of the magnetic field strength $H$, the frequency of the radiation $\omega$, and the material parameters $\epsilon_m$ and $M_{s r}^\pm$. The reduction of absorption rate is related to the magnetization process and can be expressed in terms of the magnetization $M_s$ and the magnetic moment $\mu$:

$$R_{rr}^\pm = \frac{1}{2} \left( 1 - \frac{\hbar \omega}{\omega_{s s}^\pm} \left( \frac{\hbar |E|^2}{\epsilon_m} \right)^2 |M_{s r}^\pm|^2 \right)$$

(3.46c)

The reduction of absorption rate is a measure of the magnetization process and can be expressed in terms of the magnetic field strength $H$, the frequency of the radiation $\omega$, and the material parameters $\epsilon_m$ and $M_{s r}^\pm$. The reduction of absorption rate is related to the magnetization process and can be expressed in terms of the magnetization $M_s$ and the magnetic moment $\mu$:

$$R_{rr}^\pm = \frac{1}{2} \left( 1 - \frac{\hbar \omega}{\omega_{s s}^\pm} \left( \frac{\hbar |E|^2}{\epsilon_m} \right)^2 |M_{s r}^\pm|^2 \right)$$

(3.46d)

The reduction of absorption rate is a function of the magnetic field strength $H$, the frequency of the radiation $\omega$, and the material parameters $\epsilon_m$ and $M_{s r}^\pm$. The reduction of absorption rate is related to the magnetization process and can be expressed in terms of the magnetic field strength $H$, the frequency of the radiation $\omega$, and the material parameters $\epsilon_m$ and $M_{s r}^\pm$. The reduction of absorption rate is related to the magnetization process and can be expressed in terms of the magnetic field strength $H$, the frequency of the radiation $\omega$, and the material parameters $\epsilon_m$ and $M_{s r}^\pm$.
where:

\[ M^±_f = \langle f | (x \pm iy) | i \rangle \]  

are the quantum mechanical electrical dipole matrix elements of the transition between the initial \( |i \rangle \) and final \( |f \rangle \) states, and \( V \) is the volume of the solid.

In the above equations, the only difference between optical and magneto-optical absorption is reduced to the different relative sign of the contributions for the two circular polarisations. In an optical experiment, the sum of the absorption of the two circular polarisations enters into the conductivity. Each of the two contributions ‘mix’ in the absorptive part of the diagonal optical conductivity, whereas the difference between the circular matrix elements enters into off-diagonal conductivity. Magneto-optics is selectively sensitive only to transitions in which the circular matrix elements are not equal or the absorption frequencies for the circular polarisations are different, due to exchange, spin-orbit, crystal-field and Zeeman splitting of the involved states. In eq. (3.47), it is clear that, for paramagnetic light absorption in which one of the two circular polarisations is not absorbed (due, for instance, to the selection rules), the absorptive parts of the diagonal and off-diagonal conductivities have the same absolute value.

However, it has been shown by several authors [12,14,16-20] that the very large magneto-optical effects observed for magnetically ordered materials cannot be explained simply by the Zeeman effect, but need the introduction of spin-orbit coupling. The influence of spin-orbit coupling has been considered to be a perturbation of either the initial [18] or final [17] state energies of an interband transition, or as a modification of the initial state wavefunctions [21], which is of importance if the ground-state orbital momentum is quenched. It has been pointed out that, in general, magneto-optical effects are proportional to:

\[ \sum_i (s_i \times \nabla V) \cdot p_i \]  

which is the spin-orbit energy of an electron with spin \( s \) and momentum \( p \) moving through electrical field \( -\nabla V \) inside the medium, and where the sum is over all the
electrons of the medium. This vector sum indicates that the magneto-optical effects are proportional to the product of magnetisation $\Sigma s_i$ and spin-orbit energy. In non-magnetic systems or at temperatures above the magnetic ordering temperature and if no external field is applied, there are as many electrons with up-spin as there are with down-spin. $\Sigma s_i$ is then equal to zero. Instead, with an applied field, the electrons in the vicinity of the Fermi surface have a non-zero average value of spin due to Pauli paramagnetism, and $\Sigma s_i$ is then proportional to the applied field. Based on the concept of proportionality to spin-polarisation and spin-orbit coupling, expressions for $\sigma_{xy}$, which allow quantitative calculations for interband and intraband transitions, have been developed [14,16].

The expression for the absorptive part of off-diagonal conductivity $\sigma_{xy}$ may be written as the sum of separate contributions from spin-up and spin-down electron states:

$$\sigma_{xy}(\omega) = \sigma_{xy}^{\uparrow}(\omega) + \sigma_{xy}^{\downarrow}(\omega).$$  \hspace{1cm} (3.50)

For a given interband transition $a \rightarrow b$, the concept of the total weight is introduced [14]:

$$\langle \sigma_{xy} \rangle_{a \rightarrow b} = \int_{a \rightarrow b} \sigma_{xy} \, d\omega$$  \hspace{1cm} (3.51)

as the total magneto-optic absorption due to transitions between all sublevels $\alpha$ and $\beta$ of states $a$ and $b$ at all energies. With the approximation that the only effect of spin-orbit coupling is the splitting of degenerate energy levels, the total weight may be written as:

$$\langle \sigma_{xy} \rangle_{a \rightarrow b} = \frac{\pi e^2}{4\hbar \omega V} \sum_{\alpha \beta} \{ \alpha_{\alpha \beta}^+ \left[ \beta(x+iy)\alpha \right] \|^2 - \alpha_{\alpha \beta}^- \left[ \beta(x-iy)\alpha \right] \|^2 \} \sigma, n_{\alpha} n_{\beta}$$  \hspace{1cm} (3.52)

where $\alpha$ and $\beta$ may now be viewed as the spin-orbit split sublevels of $a$ and $b$, respectively. Joint spin-polarisation $\sigma_j$ is defined as:

$$\sigma_j = \frac{n_{\alpha} n_{\beta} - n_{\beta} n_{\alpha}}{n_{\alpha} n_{\beta} + n_{\alpha} n_{\beta}}$$  \hspace{1cm} (3.53)

where $n_{\alpha}$ and $n_{\beta}$ are the occupied initial and unoccupied final states, respectively.
It is helpful to compare the off-diagonal weight $\langle \sigma_{2,\gamma} \rangle_{a\to b}$ with the corresponding diagonal weight $\langle \sigma_{1,\gamma} \rangle_{a\to b}$ [4], which is defined in an analogous way by:

$$\langle \sigma_{1,\gamma} \rangle_{a\to b} = \frac{\pi e^2}{4\hbar V_{ab}} |\langle b | \mathbf{r} | a \rangle|^2 n_a n_b \quad (3.54)$$

where $\mathbf{r}$ is the dipole operator. While all optical transitions contribute to eq. (3.54), only those states with a net spin-polarisation add to eq. (3.52), which is an elegant way of separating, e.g., magnetic orbital $f$-states from non-magnetic orbital $p$-states (as, for instance, $f$- and $p$-states in Kondo-like materials).

b) intraband transitions (free electron contribution)

Since the skew-scattering mechanism determines the Hall effect of magnetic metals, as it has been proposed in Ref. [22], it is to be expected that it should also govern the intraband contribution of off-diagonal optical conductivity.

In the skew-scattering model, frequency $\Omega$ corresponds to the spin-orbit splitting of the conduction carriers. Sample magnetisation couples to the orbital moments of the electronic states through spin-orbit interaction. As a consequence, an electrical dipole moment of the form shown below results:

$$\mathbf{p}(k) = A \mathbf{k} \times \mathbf{M} . \quad (3.55)$$

Following Refs. [4,7,23], it may be shown that the free electron contribution to off-diagonal conductivity for a spherical Fermi surface and for $\mathbf{E}$ along the $x$-axis may be written as:

$$\tilde{\sigma}_{xy}(\omega) = \frac{\omega^2}{4\pi} \left( \sigma_{2,\gamma} \right) \left[ -\frac{\Omega}{\Omega^2 + (\gamma + i\omega)^2} + \frac{P_0}{e V_F} \left( 1 - \frac{i\omega(\gamma + i\omega)}{\Omega^2 + (\gamma + i\omega)^2} \right) \right] \quad (3.56)$$

where $\Omega = s/\tau_s$, $\gamma = 1/\tau$, $V_F$ is the Fermi velocity, $P_0$ is the macroscopic dipole moment, and:

$$\langle \sigma_{2,\gamma} \rangle = \frac{n_+ - n_-}{n_+ + n_-} \quad (3.57)$$

is the spin polarisation (with respect to the direction of the external field) of conduction charge carriers.
From eq. (3.56), we obtain for the absorptive part of the off-diagonal conductivity:

\[
\sigma_{xy}(\omega) = \frac{\omega_p^2 \langle \sigma_z \rangle}{4\pi} \left[ \frac{2\gamma\Omega\omega}{(\Omega^2 + \gamma^2 - \omega^2)^2 + 4\gamma^2 \omega^2} \right] - \frac{P_0}{eV_F} \left[ \frac{\omega\gamma(\Omega^2 + \gamma^2 - \omega^2)}{(\Omega^2 + \gamma^2 - \omega^2)^2 + 4\gamma^2 \omega^2} \right].
\] (3.58)

In the limit \(\omega \gg \Omega \) and \(\omega \gg \gamma\), the first and second terms of eq. (3.58) are proportional to \(\omega^3\) and \(\omega^1\), respectively. If the skew-scattering frequency is identified with \(\omega_c\) and \(\langle \sigma_z \rangle = 1\), we note that the first term of eqs. (3.56) and (3.58) corresponds to the classically derived Drude term for \(\sigma_{xy}\) (compare with eq. (3.43) with \(f^+ = f^- = 1\)). In this case (\(\Omega = \omega_c\) and \(\langle \sigma_z \rangle = 1\)) and if the damping is small enough to allow a screened plasma frequency \(\omega_{\text{min}}\) well below the lowest energy interband transition, this term, together with the presence of interband transitions at higher energies, can dominate the magneto-optical response of metals. It may be viewed as a splitting of the plasma-edge for the two circular polarisations (different \(\omega_{\text{min}}\) for RCP and LCP, Fig. 3.7 from Ref. [24]) and may lead to relatively large magneto-optical effects when the plasma-edge in the optical reflectivity is steep (§ 3.2.5). In condition \(4\omega_{\text{min}}^2 \gg \omega_c^2\), the position of the minimum in the reflectivity for RCP and LCP components in the presence of an externally applied magnetic field is shifted according to [6,24]:

**Fig. 3.7:** Plasma-edge splitting for RCP and LCP in InSb [24].
which means that the shift may be considered, as a first approximation, proportional to the externally applied magnetic field.

Figure 3.8 shows the experimental Kerr effect of paramagnetic silver for an applied field of 1 T [25], which is a typical example of a Kerr signal due to plasma-edge splitting. Obviously, larger Kerr effects may be generated if \( \omega_k \) is enhanced relative to \( \omega_{\text{min}} \) either by reducing \( \omega_{\text{min}} \) or by increasing \( \omega_k \). In the same way, a larger Kerr effect may occur if a skew-scattering mechanism, in which \( \Omega > \omega_k \), dominates. Enhancement of plasma-edge splitting due to a skew-scattering mechanism successfully reproduces \( \theta_k \) and \( \epsilon_k \) measured in TmS and TmSe [23].

3.2.5 The Lorentz-Drude Model in Magneto-Optics

In the last section it was pointed out that the plasma-edge splitting phenomenon observed in metals may give rise to relatively large Kerr rotations and ellipticities in the region where the reflectivity minimum occurs (Fig. 3.8, Ref. [25]). In this section, this aspect is deeply analysed and discussed in detail, with a simple numerical case representing a system characterised by a Drude component (free electrons, intraband transitions) and a single harmonic oscillator at higher energies (bound electrons, a single interband transition).

Starting from eq. (3.43) and separating the real and imaginary parts, we can calculate the off-diagonal components of the conductivity tensor and then obtain
θ_k and ε_k from eqs. (3.33) and (3.34). As we will see in Chapter 5, the calculation procedure for a real system starts with a set of parameters that "fits" the optical reflectivity and conductivity, at least qualitatively. Since the measured reflectivity cannot distinguish between right and left circularly polarised light, coefficients A and B are calculated from eqs. (3.35) and (3.36), where n and k are obtained directly from eqs. (3.21), (3.8) and (3.7). Note that eqs. (3.37) and (3.38), as well as eqs. (3.33) and (3.34), hold under the approximations (3.28). Consequently, the calculated θ_k and ε_k hold with good approximation only if:

\[ \theta_k, \epsilon_k \leq 10^\circ - 12^\circ. \]  

(3.60)

References [7,26] report relations between θ_k, ε_k and \( \tilde{\sigma}_{xy} \) which hold without any restrictions.

We observe that, for eqs. (3.42), (3.43), (3.33) and (3.34), the sum rule (3.19) becomes [27]:

\[ f_j^+ + f_j^- = 2 \]  

(3.61)

for both the Drude component and each harmonic oscillator j. This last observation is the consequence of the two following important facts:

i) The density of j electrons with plasma frequency \( \omega_{pj} \) (the density of free charge carriers with spectral weight \( \omega_p \) for the Drude component) is already included in the plasma frequency itself (eq. (3.16)), to which we give a numerical evaluation (Tab. 3.1).

ii) From eqs. (3.42) and (3.43) and for \( H = 0 \) (\( \omega_{kj} = 0 \) for each j) and \( f_j^+ = f_j^- = 1 \) (again for each j), the optical situation is recovered for both diagonal (eq. (3.21)) and off-diagonal components of the conductivity tensor (i.e. \( \tilde{\sigma}_{xy} \equiv 0 \), as expected).

<table>
<thead>
<tr>
<th>Drude</th>
<th>( \omega_p = 3.0 \text{ eV} )</th>
<th>( \Gamma = 0.2 \text{ eV} )</th>
<th>( f_j^+ = 1, f_j^- = 1 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lorentz</td>
<td>( \omega_{p1} = 3.5 \text{ eV} )</td>
<td>( \gamma_1 = 1.2 \text{ eV} )</td>
<td>( \omega_1 = 4 \text{ eV} )</td>
</tr>
<tr>
<td>( \epsilon_{\infty} = 1 )</td>
<td></td>
<td></td>
<td>( B = 1 \text{ T} )</td>
</tr>
</tbody>
</table>

Tab. 3.1: Parameter set used to calculate the curves shown in Figs. 3.9 and 3.10.
With the set of parameters in Tab. 3.1 representing a realistic situation that we will often face in Chapter 5, we obtain the reflectivity and the real part of optical conductivity (Fig. 3.9, plain lines), the real and imaginary parts of $\sigma_{xy}$ and

![Graph of reflectivity and optical conductivity](image)

**Fig. 3.9:** Reflectivity and optical conductivity curves calculated with the parameter set of Tab. 3.1 and for different values of the Drude plasma frequency.

the Kerr rotation and ellipticity (Fig. 3.10, plain lines). Oscillator strengths $f^z$ are equal to one for both the Drude component and the harmonic oscillator (Drude component and h.o. unpolarised case). Reflectivity shows typical metallic behaviour and reaches 100% at low energies. At ~2.5 eV we notice the steep
plasma-edge with the deep reflectivity minimum and, at higher energies, the onset of the interband transition. $\sigma_1$ shows metallic behaviour at lower energies and the interband transition centred at 4 eV. Off-diagonal conductivity shows a structure

![Graph showing off-diagonal conductivity and polar Kerr rotation $\theta_K$ and ellipticity $\varepsilon_K$ curves calculated with the parameter set of Tab. 3.1 and with different values of the Drude plasma frequency ($\omega_p$ only for $\omega_p = 3.0$ eV).]

**Fig. 3.10:** Off-diagonal conductivity and polar Kerr rotation $\theta_K$ and ellipticity $\varepsilon_K$ curves calculated with the parameter set of Tab. 3.1 and with different values of the Drude plasma frequency ($\omega_p$ only for $\omega_p = 3.0$ eV).

with a diamagnetic line-shape (i.e. $f_1^+ = f_1^- = 1$) associated with the interband transition at 4 eV. The shape and magnitude of $\theta_K$ and $\varepsilon_K$ are comparable with those already seen in Fig. 3.8 for silver. The important fact is that the structure in $\theta_K$ and $\varepsilon_K$ arises where the reflectivity minimum occurs and is the result of the
3. Theory

The presence of both the Drude component and the interband transition at higher energies and of their mutual interplay. In order to prove the last statement, we calculated the same quantities with two larger values for the spectral weight of the Drude component ($\omega_p = 3.6$ eV for dashed and $\omega_p = 4.2$ eV for dotted lines, respectively). We note that:

- the reflectivity minimum is shifted towards higher energies;
- the structure in the optical conductivity does not move, being the direct optical manifestation of the interband transition at 4 eV;
- the structure in the off-diagonal conductivity does not move, being the direct magneto-optical manifestation of the diamagnetic interband transition at 4 eV;
- the peak in Kerr rotation $\theta_K$ ($\varepsilon_K$ behaves in the same way) 'follows' the reflectivity minimum.

The same results may be obtained by decreasing rather than increasing $\omega_p$ (in this case, the reflectivity minimum shift would be towards lower energies). Moreover, the structure width may also be changed by changing the scattering rate of the free electrons and/or the damping of the harmonic oscillator. In addition, if instead of $\omega_p$ we change the resonance frequency of the harmonic oscillator $\omega_1$, we change not only the position of the structure in $\theta_K$ and $\varepsilon_K$ (it always coincides with the reflectivity minimum) but also the position of the structure in $\sigma_1$ and $\sigma_{xy}$, because this structure is always centred in $\omega_1$.

An explanation for this variety of phenomena is found in Ref. [28]. Feil and Haas [28] started rewriting the polar Kerr effect in the following way:

$$\theta_K + i\varepsilon_K = \frac{\tilde{\varepsilon}_{xy}}{\sqrt{\varepsilon_{xx}(1 - \tilde{\varepsilon}_{xy})}}$$  (3.62)

where $\tilde{\varepsilon}_{xx}$ and $\tilde{\varepsilon}_{xy}$ are the diagonal and off-diagonal parts of the complex dielectric tensor, respectively. With a suitable choice of parameters and using a constant value for $\tilde{\sigma}_{xy}$, Feil and Haas found a large Kerr effect centred in the reflectivity minimum. They argue that this large Kerr effect is associated with the denominator of eq. (3.62), which has a singularity in the reflectivity minimum because $\tilde{\varepsilon}_{xy} = 1$. Separating the real and imaginary parts of eq. (3.62), we obtain, in agreement with eqs. (3.33) and (3.34) [29]:

$$\theta_K + i\varepsilon_K = \frac{4\pi}{\omega} \frac{1}{A^2 + B^2} \left[ B\sigma_{1xy} + A\sigma_{2xy} \right] + i \left[ A\sigma_{1xy} - B\sigma_{2xy} \right].$$  (3.63)
It is now interesting to look at the pre-factor \((A^2+B^2)^{-1}\) calculated with the same set of parameters as in Tab. 3.1 and for \(\omega_p = 3\) eV (plain line), 3.6 eV (dashed line) and 4.2 eV (dotted line). The results are represented in Fig. 3.11 where there is a resonance-like feature centred in the reflectivity minimum. This pre-factor may be one of the reasons for the observed enhanced Kerr effect in the reflectivity minimum region.

Brändle [30] introduced the new concept of 'C and D factors' defined as follows, from eqs. (3.33) and (3.34):

\[
\Theta_K = -\frac{4\pi}{\omega} \left[ \frac{B}{A^2 + B^2} \sigma_{xy} + \frac{A}{A^2 + B^2} \sigma_{yz} \right] = -4\pi \hbar (D\sigma_{1xy} + C\sigma_{2xy}) \quad (3.64)
\]

\[
\varepsilon_K = -\frac{4\pi}{\omega} \left[ \frac{A}{A^2 + B^2} \sigma_{xy} - \frac{B}{A^2 + B^2} \sigma_{yz} \right] = -4\pi \hbar (C\sigma_{1xy} - D\sigma_{2xy}) \quad (3.65)
\]

with \(A\) and \(B\) as in eqs. (3.35) and (3.36) and:

\[
C = \frac{1}{\hbar \omega} \left( \frac{A}{A^2 + B^2} \right) \quad \text{and} \quad D = \frac{1}{\hbar \omega} \left( \frac{B}{A^2 + B^2} \right). \quad (3.66)
\]

In Fig. 3.12, these two factors are calculated with the same set of parameters as in Tab. 3.1. The trend of \(C\) and \(D\) is almost identical to that of \(\varepsilon_K\) and \(\Theta_K\) in Fig. 3.10 (plain lines) respectively. This suggests that, since \(C\) and \(D\) depend only on optical functions \(n\) and \(k\) (eqs. (3.66), (3.35) and (3.36)), the Kerr structure in the reflectivity minimum has mainly an 'optical origin', whereas \(\tilde{\sigma}_{xy}\), representing the magneto-optical response of the system, only seems to play a marginal role.
We will see below that these observations are only partially true, since the role played by \( \sigma_{xy} \) becomes absolutely dominant if Drude component polarisation and/or oscillator polarisation are taken into account. In fact, the model proposed by Feil and Haas has a considerable limitation, in that \( \sigma_{xy} \) is assumed to be energy-independent and is set to a constant value (\( \sigma_{xy} = 1 + i \) in Ref. [28]). As pointed out in Ref. [29], this assumption is incompatible with the Kramers-Kronig relation. A constant \( \sigma_{xy} \) leads to \( \sigma_{2xy} = 0 \). In addition, because \( \sigma_{xy} \) and \( \sigma_{xy} \) rely on the same optical intraband and interband transitions, these functions of \( \omega \) are interdependent. The assumption of Drude and Lorentz terms for \( \sigma_{xx} \) but a constant \( \sigma_{xy} \) violates the self-consistency requirement.

Furthermore, the \( C \) and \( D \) factors proposed by Brändle [30] are two quantities which must be multiplied by \( \sigma_{xx} \) and \( \sigma_{2xy} \) in order to obtain \( \theta_K \) and \( \varepsilon_K \) (eqs. (3.64) and (3.65)). This means that direct comparison between \( C \) and \( D \) and \( \theta_K \) and \( \varepsilon_K \) makes sense only if \( \sigma_{xy} \) is assumed to be almost energy-independent.

With respect to the Feil and Haas model [28] and to Brändle's observations about the \( C \) and \( D \) coefficients [30], our way of calculating \( \theta_K \) and \( \varepsilon_K \) from eq. (3.43) and eqs. (3.33) and (3.34) presents the two following advantages:

i) It does not require any assumption on \( \sigma_{xy} \) and holds for all the parameter sets that give a result in accord with the condition (3.60);

ii) It gives us the possibility of 'weighting' the transitions associated with RCP and LCP through the \( f^+ \) and \( f^- \) coefficients, respectively, in accordance with the sum rule (3.61).
We want now to give an idea of the dramatic changes that the introduction of a polarisation produces on the magneto-optical Kerr effect shown in Fig. 3.10 (plain lines). We will distinguish two different cases: a) polarisation of bound electrons; b) polarisation of free electrons.

a) polarisation of bound electrons (Lorentz oscillator)

Figure 3.13 shows the calculated Kerr rotation and ellipticity with the same

![Graph showing Kerr rotation and ellipticity](image)

**Fig. 3.13:** Polar Kerr rotation $\theta_K$ and ellipticity $\varepsilon_K$ and off-diagonal conductivity curves (inset) calculated with the parameter set of Tab. 3.1 and with a small polarisation of the h.o., i.e. $f_1^* = 1.01$ and $f_1 = 0.99$. 
parameters as in Tab. 3.1, but with a very small polarisation of the harmonic oscillator at 4 eV, i.e. \( f_i^+ = 1.01 \) and \( f_i^- = 0.99 \). With respect to Fig. 3.10, note that the Kerr effect is more than a factor of 100 larger and that the shape has completely changed. In the inset of Fig. 3.13, the off-diagonal components of the conductivity tensor are also plotted and they show a paramagnetic line-shape (as expected, because \( f_i^+ \neq f_i^- \)) and an increase of more than a factor of 100 with respect to the corresponding quantities in Fig. 3.10. Figure 3.14 shows Kerr rotation as function of energy for \( f_i^+ \) ranging from 1 to 1.1. It is important to note that the structure in \( \theta_k \) and \( \varepsilon_k \) remains centred where the reflectivity minimum occurs and that the optical quantities \( R \) and \( \sigma_i \), representing the optical response of the medium, are polarisation-independent.

\[\text{Fig. 3.14: Polar Kerr rotation } \theta_k \text{ curve calculated with the parameter set of Tab. 3.1 and with polarisation of the h.o. ranging from 1 to 1.1, i.e. } 1 \leq f_i^+ \leq 1.1.\]

\[b) \text{ polarisation of free electrons (Drude component)}\]

Figure 3.15 shows what happens when we polarise the Drude component, i.e. \( f^+ = 1.01 \) and \( f^- = 0.99 \). With respect to Fig. 3.10, enhancement of more than a
factor of 100 is again obtained for $\theta_K$ and $\varepsilon_K$, but the shape remains almost unchanged. Figure 3.16 shows Kerr rotation as function of energy and for $f^+$

![Graph showing Kerr rotation and ellipticity curves](image)

**Fig. 3.15**: Polar Kerr rotation $\theta_K$ and ellipticity $\varepsilon_K$ curves calculated with the parameter set of Tab. 3.1 and with a small polarisation of the Drude component, i.e. $f^+ = 1.01$ and $f^- = 0.99$.

ranging from 1 to 1.1. Again we note that the structure in $\theta_K$ and $\varepsilon_K$ remains centred where the reflectivity minimum occurs and that the optical quantities are polarisation-independent.

In both the previous cases, i.e. when a polarisation is introduced, the dependence of $\theta_K$ and $\varepsilon_K$ on externally applied magnetic field intensity is almost negligible, since it produces changes smaller than $0.1^\circ$ even for magnetic fields up
to 10 T. Without polarisation, $\theta_K$ and $\varepsilon_K$ are, in a first approximation, proportional to the magnetic field (eqs. (3.59), (3.43), (3.33) and (3.34)).

**Fig. 3.16:** Polar Kerr rotation $\theta_K$ curve calculated with the parameter set of Tab. 3.1 and with polarisation of the Drude component ranging from 1 to 1.1, i.e. $1 \leq f^+ \leq 1.1$.

Furthermore, we observe that, in $a)$ and $b)$, the polarisation was always chosen with $f^+ > f^-$. If we reverse the Drude component or the Lorentz oscillator polarisation ($f^+ < f^-$), the effect produced is a change of sign in both $\sigma_{xy}$ and in $\theta_K$ and $\varepsilon_K$, in agreement with eq. (3.43). Note that, whereas for the Drude component both ways of polarisation are possible in principle, for the Lorentz oscillator we argue that only the case $f^+ > f^-$ is physically meaningful. This follows from the fact that $f^+$ is associated with the RCP through the selection rule $\Delta m_j = +1$ and $f^-$ is associated to the LCP through the selection rule $\Delta m_j = -1$ (see Fig. 3.5). In this way, RCP has a higher resonance frequency than LCP and the starting energy level is at lower energy than that associated with LCP. In a system in the fundamental state, the most populated level is that at low energy. Since $f^\pm$ are the oscillator strengths and represent the ‘weight’ of the transition, we expect $f^+ > f^-$. 
REFERENCES


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4. EXPERIMENT

We review here optical and magneto-optical experimental facilities. While optical ones will only be briefly described, the magneto-optical experimental set-up will be presented in detail.

4.1 Optical Experimental Set-up

In order to cover the wide spectral range extending from the far-infrared (FIR) to the ultra-violet (UV), i.e. from 0.025 to 12 eV, three different spectrometers with overlapping energy ranges are used.

From 0.025 eV (FIR) up to the mid-infrared (MIR), a Fourier transform fast scanning Brucker IFS 48 interferometer is used with a globar lamp light source. To obtain the absolute value of sample reflectivity, it must always be measured against that of an already known reference material. A gold mirror is generally used, reflecting almost 100% over the whole spectral range. A home-made $^4$He exchange gas cryostat allows measurements in the temperature range 2 - 300 K.
From the infrared (IR) up to the visible and UV range, a **home-made spectrometer** with a halogen lamp as light source and a photomultiplier as detector is used. In this spectrometer, an optical system equipped with a turning light guide allows direct measurement of the focused incident light beam and alternatively the intensity of the light beam reflected by the sample. In this way no reference mirror is needed. With this equipment, the crystals can be cleaved in situ in a vacuum of \(10^{-10}\) mbar.

In UV up to 12 eV, a **McPherson 225** spectrometer is used, employing a double beam technique, which also avoids the need for a reference mirror. The light source is a hydrogen lamp and the detector is a photomultiplier. Also in this case, cleaving in situ in a vacuum of \(10^{-9}\) is possible.

The experimental data from the various spectrometers are then assembled to perform Kramers-Kronig transformations (§ 3.1.2).

### 4.2 Magneto-Optical Experimental Set-up

This set-up consists of two main parts: a \(^3\)He cryostat and a home-made optical spectrometer. A data acquisition system based on an EG&G 5210 Lock-in Amplifier and a set of stepping motors driving the various optical elements allow for fully automatic polar Kerr effect measurement.

#### 4.2.1 The \(^3\)He Cryostat

Figure 4.1 shows a schematic section of the \(^3\)He cryostat used in this work [1,2]. The stairlike shape of the top flansh indicates the modular construction of this cryostat. There are three modules. The outermost one includes the liquid nitrogen (N\(_2\)) vessel and the outer wall of the main \(^4\)He bath (2). The intermediate module holds the magnet with its wiring (6). The innermost module consists of the \(^3\)He insert (sample space (10) with sorption pump (4)), the 1 K pot (3) and the inner wall of the main \(^4\)He bath (2). The liquid nitrogen vessel acts as a cold trap and enhances the isolation vacuum (V). With a vacuum of \(10^{-7}\) mbar, a holding time of two days is reached for liquid helium. The 1 K pot is in thermal contact with the exchange gas collected in the sorption pump (10 and 4). Liquid \(^4\)He is pumped from the He bath through a needle valve into the 1 K pot and evaporated. By pumping the evaporated \(^4\)He from the 1 K pot, temperatures down to about 1.5 K in the sample chamber can be reached. Using \(^3\)He as exchange gas (10), further
cooling down to 0.3 K can be achieved by pumping the condensed $^3$He in the sample chamber with the sorption pump.

**Fig. 4.1: The $^3$He Cryostat.**

Optical access is at the bottom of the cryostat, through strainfree optical windows (11, 12, 13 and 14). The windows (12) and (13) do not significantly
enhance the holding time of the liquid He, although they do cause significant reflection losses (about 22%). Therefore, the cryostat operates without them. The warm outermost window (14) has a diameter of 35 mm and the cold innermost one (11) of 13 mm. The distance between the two windows is 230 mm, giving an opening angle of 5.7°.

The superconducting magnet consists of several concentric NbTi and Nb$_3$Sn solenoids ($T_c = 9.2$ K and 9.5 K, respectively) and allows for fields up to 13.2 Tesla. The sample is placed at the centre of the magnet. The light beam is focussed on to the sample or reference gold mirror and both are mounted on a disk (9) at the bottom of the driving rod (8). Half-turns of the driving rod easily perform switches between sample and reference.

Three sensors are used to monitor sample temperature during the experiment. A thermocouple (AuFe / Chrome) is used in the range between 4 K and room temperature, and a Ge sensor covers the accessible temperature range below 4.2 K, but its calibration curve is strongly field-dependent. A carbon resistor aids for low-temperature measurements in the field, owing to its relatively low magnetoresistance.

The cryostat windows are made of BaF$_2$. Among the transparent materials between 0.1 and 5.5 eV, BaF$_2$ has the advantages of being not light-sensitive and only weakly hygroscopic, and of having a cubic crystal structure and a relatively small refraction index. However, as it is sensitive to sudden temperature variations, care must be taken in cooling down the cryostat and inserting the sample holder (7 in Fig. 4.1). Figure 4.2a shows the sealing arrangement for

![Fig. 4.2: a) Old and b) new sealing arrangement for the cold window.](image-url)
the cold window (11 in Fig. 4.1). The BaF\(_2\) window is stuck to top of the holder and is tilted by about 5° in order to avoid multiple reflections with the sample. A PVC holder is used to keep the window in the right position during fixing and is then removed. Stycast 2850 FT, used as glue, is diluted by 7% Catalyst 24 LV. The resulting fluid mass is held in place by capillarity because the space between window and Cu holder is kept as small as possible (~0.1 mm).

Sealing the cold BaF\(_2\) window is a very delicate procedure. To overcome problems related to the different expansion coefficients of BaF\(_2\), the glue and the Cu holder, the holder wall is cone-shaped, with thickness ranging between 0.3 and 0.1 mm, in order to make it flexible. It is also annealed to reduce strains.

Even with all these technical tricks, the BaF\(_2\) cold window sometimes broke and we risked to lose large amounts of \(^3\)He through the vacuum chamber. For the window holder, this problem was overcome by making the small but efficient modification shown in Fig. 4.2b. In comparison with the old ones, the new holder walls are tilted by 5°, so that the window can be stuck on with its borders parallel to the holder walls. In this way, the cause of window breaking, probably due to the anisotropic pressure on the window corners (Fig. 4.2a), is avoided. Besides, as may easily be shown by means of geometrical calculations, the amount of light passing through the window in the new configuration was not reduced but increased since the cold window diameter was increased from 12 to 13 mm.

### 4.2.2 Optical Set-up

Figure 4.3 shows the optical set-up [1,2]. The spectral range 0.23 - 5.6 eV is covered, making use of the following optical elements:

- Three different light sources, i.e. a globar lamp (0.23 - 1.2 eV), a halogen filament in a quartz bulb (0.23 - 2.8 eV), and a xenon high-pressure lamp with a water-cooled IR filter (2.7 - 5.6 eV).
- A dispersive double Zeiss MM3 monochromator with three sets of prisms: CaF\(_2\) (0.23 - 1.5 eV), Glass (0.5 - 3.4 eV) and Quartz (1.3 - 5.6 eV). The monochromator slit opening value, which may be varied between 0.01 and 2 mm, is set at the beginning of each measurement and is the result of the best compromise between the two following factors:
  a) spectral resolution;
  b) signal intensity at the detector.
- Spherical Al mirrors, yielding a 1 to 1 image of the monochromator slit on the sample and of the sample on the detector. Plane Al mirrors (M\(_2\) to M\(_5\) in...
Fig. 4.3) turn the light beam from horizontal on the optical table to vertical in the cryostat and again horizontal on the optical table. The mirrors are made of MgF$_2$-coated aluminium.

![Diagram of optical set-up](image)

**Fig. 4.3:** The optical set-up: MM3 = Monochromator; P, A = Polarisers (Glan, BaF$_2$); C = Soleil-Babinet Compensator ($\epsilon_K$ measurement); $F_{\Omega}$ = Faraday Cell (Signal Modulation).

- Al wire on a BaF$_2$ substrate (0.23 - 1.2 eV) and Glan-Thompson (0.5 - 5.6 eV) polarisers (P = polariser and A = analyser in Fig. 4.3).
- Two phase-shifters, i.e. a CdS (0.23 - 2 eV) and quartz Soleil-Babinet (0.5 - 5.6 eV) compensator (C in Fig. 4.3). The phase-shifters are based on the Sénarmont principle [3] and are inserted in the light beam to measure its ellipticity.
- An AC-driven solenoid with a longitudinal bore in the center (Faraday cell, F_0 in Fig. 4.3). An optical glass placed in the bore (Faraday modulator) is subject to the oscillating magnetic field of the solenoid and causes an oscillating circular phase shift on transmitted light. The effect on the light beam is an oscillating Faraday rotation:

\[
\theta_F(t) = \gamma_0(t) = \gamma_0 \sin \Omega t
\]

of the azimuth of the polarisation, where \(\gamma_0\) is the modulator Faraday rotation amplitude. Frequency \(\Omega\) is the frequency of the current in the solenoid. The Faraday cell operates with an amplified sinusoidal signal oscillating at \(\Omega = 80\) Hz. Typically, the Faraday ellipticity of a material is negligible in its transparency window. Three different Faraday modulators are used: yttrium iron garnet \(Y_3Fe_5O_{12}\) (called YIG), SF59 glass and Suprasil 1 in the spectral intervals 0.23 - 1, 0.5 - 2.9 and 2.7 - 5.6 eV, respectively.

<table>
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<td>BaF_2</td>
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<td>InSb</td>
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<td>BaF_2</td>
<td>CdS</td>
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<td>InSb</td>
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<td>Glan</td>
<td>Quartz</td>
<td>SF59</td>
<td>InSb</td>
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<tr>
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<td>Glass</td>
<td>Glan</td>
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<td>SF59</td>
<td>Si</td>
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<td>Glan</td>
<td>Quartz</td>
<td>SF59</td>
<td>Photom.</td>
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<tr>
<td>2.7 - 5.6</td>
<td>Xe</td>
<td>Quartz</td>
<td>Glan</td>
<td>Quartz</td>
<td>Suprasil</td>
<td>Photom.</td>
</tr>
</tbody>
</table>

Tab. 4.1: Spectrometer optical arrangements used to cover the whole spectral range 0.23 - 5.6 eV.

- Three different detectors, i.e. a photovoltaic N_2-cooled InSb detector (0.23 - 1.5 eV), a Si photocell (1.2 - 2.5 eV) and a photomultiplier (1.6 - 5.6 eV). In order to obtain the best signal-to-noise ratio over the whole spectral range 0.23 - 5.6 eV, the various optical elements are arranged as shown in Tab. 4.1.
4.2.3 Magneto-Optical Experimental Technique

In magneto-optical Kerr effect measurements, the basic idea is to find the analyser position $\beta_{\text{min}}$, corresponding to the lowest signal at the detector when the polarised light is reflected by both the sample (Sam) and the gold mirror (Ref), without and with the phase-shifter compensator. The light intensity at the detector is proportional to the squared absolute value of the polarisation amplitude at the detector.

No phase-shifter is needed to measure polar Kerr rotation $\theta_k$. With the low incidence angle of less than $3^\circ$ on Sam and Ref, the difference between the reflection coefficients parallel or perpendicular to the plane of incidence can be neglected [1]. If light is reflected by the sample, by making use of matrix formalism and the Bessel series, it can be shown that, in first approximation, the modulated light intensity at the detector is given by [2,4]:

$$I_\Omega = I_0 J_1(2\gamma_0) R_{\text{sam}} \sin^2(\beta - \alpha - \theta_k) \sin \Omega \theta$$  \hspace{1cm} (4.2)

where $J_1(2\gamma_0)$ is the first term of the Bessel expansion in $\gamma_0$, $R_{\text{sam}}$ is sample reflectivity, $\beta$ is analyser position, $\alpha$ is polariser position and $\theta_k$ the Kerr rotation of the sample. Analyser angle $\beta_{\text{min}} = \alpha + \theta_k$ is determined from the zeroes of $I_\Omega$. If light is reflected by the gold reference, $\beta_{\text{ref}} = \alpha$ is determined by means of the same procedure (gold mirror Kerr rotation and ellipticity are $\equiv 0$ over the whole spectral range [5]). The sample Kerr rotation is then obtained from:

$$\theta_k = \beta_{\text{min}} - \beta_{\text{ref}}.$$  \hspace{1cm} (4.3)

Modulated intensity $I_\Omega$ as a function of analyser orientation has a sinusoidal shape and a period of $90^\circ$. It becomes linear near the zeroes. Analyser orientation corresponding to a minimum of $I_\Omega$ is determined as the crossing ($x_0$) of the regression lines that fit the modulated intensity in the linear region, according to the fitting function:

$$y = A \cdot |x - x_0| + B.$$  \hspace{1cm} (4.4)

Figure 4.4 shows the measurement of a minimum of $I_\Omega$, the so-called “V-curve”. A mean standard deviation of $0.0017^\circ$ is determined for $\beta_{\text{min}} = x_0$ for the linear-regression fit of Fig. 4.4. Repeated measurements of the same V-curve reproduce minimum position $\beta_{\text{min}}$ within an error of a few thousandths of a degree. The
typical experimental uncertainty of $\beta_{\text{min}}$ lies between $0.001^\circ$ and $0.01^\circ$ over the whole photon energy range available.

Fig. 4.4: A typical "V-curve" measurement.

According to eq. (4.2), note that sample reflectivity $R_{\text{sam}}$ may simultaneously be determined from the V-curve fitting procedure. In fact, $R_{\text{sam}}$ is given by

$$R_{\text{sam}} = R_{\text{Gold}} \frac{A_{\text{sam}}}{A_{\text{ref}}}$$

(4.5)

where $R_{\text{Gold}}$ is reference gold mirror reflectivity and $A_{\text{sam}}$ and $A_{\text{ref}}$ are the slopes of the V-curves for Sam and Ref, respectively.

To measure polar Kerr ellipticity $\varepsilon_K$, the phase-shifter is driven in the light beam just before the Faraday cell (Fig. 4.3). By setting the phase shift to $\lambda/4$ and
orienting the axes of the phase-shifter parallel to the main axes of the polarisation ellipse, i.e. \( \varphi = \alpha + \theta_k \), ellipticity may be measured within the Sénarmont principle [3]. In this case, the modulated intensity is given by [2,4]:

\[
I_\Omega = I_0 J_1(2\gamma_0) R_{\text{sam}} \left| \frac{\sin 2(\beta - \varphi + \varepsilon_K)}{\cos 2\varepsilon_K} \right| \sin \Omega t.
\] (4.6)

The Kerr ellipticity of the sample is then obtained in the same way as eq. (4.3) from:

\[
\varepsilon_K = \beta_{\text{sam}}^2 - \beta_{\text{PhSh}}^2 - (\beta_{\text{sam}}^\text{ref} - \beta_{\text{PhSh}}^\text{ref}) = \beta_{\text{sam}}^2 - \beta_{\text{PhSh}}^2 - \theta_k
\] (4.7)

where superscripts ‘sam2’ and ‘PhSh’ refer respectively to sample and reference measurement with the phase-shifter in the light beam.

Modulated intensity \( I_\Omega \) in eq. (4.6) as a function of analyser orientation (\( \beta \)) has again a period of 90° but, as a function of phase shifter orientation (\( \varphi \)), it has a period of 180°. In fact, for \( \varphi = \alpha + \theta_k \pm 90° \), \( I_\Omega \) becomes:

\[
I_\Omega = I_0 J_1(2\gamma_0) R_{\text{sam}} \left| \frac{\sin 2(\beta - \varphi - \varepsilon_K)}{\cos 2\varepsilon_K} \right| \sin \Omega t.
\] (4.8)

Observe that the only difference between eq. (4.6) and eq. (4.8) is in the sign of \( \varepsilon_K \). This means that, if measurement of \( \beta_{\text{min}} \) is performed with the phase-shifter orientation shifted by ± 90°, \( \varepsilon_K \) is obtained with the opposite sign.

There are a few conclusive remarks:

- the first order depolarisation effects of mirrors M2 and M3 (forming a \( \sim 45° \) angle with the light beam) are compensated by mirrors M4 and M5 [1];
- possible contributions to light polarisation from both small misalignments and any anisotropic effect of the mirrors may be neglected when measurements are performed with the magnetic field in both directions (magneto-optical effects are odd in the applied field, whereas both small misalignments and anisotropic mirror effects are in first approximation field-independent);
- the Faraday effect of the cryostat windows contributes to the modulated signal through an additional polarisation rotation, independent of the
measured sample. With measurements against the reference, this effect is eliminated.

4.3 Samples

All the samples used in this work were high-quality single crystals grown by F. Hulliger, E. Kaldis, K. Mattenberger or O. Vogt by mineralization. With this method, the starting material, in the form of pellets, is encapsulated in tungsten crucibles and heated for several weeks to just below its melting point. Crystals are formed by recrystallisation. The pellets are then converted into an assembly of a few crystals only. Details can be found in Ref. [6].

The structure and lattice constants of the single crystals were checked by X-ray analysis. In some cases, magnetisation and resistivity measurements were performed, and confirmed that the single crystals were of the desired quality. However, as some compounds (in particular, CeSb and LaTe) are subject to oxidation, all the samples were either cleaved in situ and measured, or cleaved and then directly mounted on the sample holders of the various spectrometers in an Ar glove box, to prevent air contamination.

The typical dimensions of cleaved samples were 2x3x0.5 mm³. Kerr effect measurements performed on various cleaved crystals of the same material did not reveal any differences over the whole spectral range. A different light intensity at the detector due to light losses caused by different scattering mechanisms at the cleaved surface of the several samples was found, but it did not influence the Kerr effect results, as shown in § 4.2.3 (eqs. (4.2) and (4.6)).

Reference [4] mentions the fact that a sample with a cleaved or polished surface may give a different magneto-optical response. Uranium Arsenide (UAs) for example [4,7], reveals dramatic differences in θK when samples with polished or cleaved surfaces are examined. According to Ref. [4], polishing alters the first 100 nm and creates instability, especially in materials in which equilibrium among magnetic phases is critical. Although some of the compounds analysed in the present work do not present this problem (e.g. LaX are paramagnetic metals), all the measured sample surfaces are cleaved.
REFERENCES

5. RESULTS AND DISCUSSION

This chapter presents the results and discussion of optical and magneto-optical measurements on the various materials presented in this thesis (Chapter 2). Some of them, i.e. LaSe, EuO and CeSb, have already been optically and magneto-optically analysed by other authors [1-13]. Therefore, in order to discuss, inter alia, analogies and differences between the previous investigations and ours, we will also briefly summarise the available results from the literature.

5.1 LaX (X = S, Se, Te)

In § 3.2.4 (eqs. (3.52) and (3.53)) it was shown that the magneto-optical signal is proportional to the coupled spin-polarisation of the electronic states involved in the transition. Because of this fact, it has recently been argued [1] that even a non-magnetic or a weakly magnetic material, i.e. one with non-occupied spin-polarised states, may have a strong magneto-optical signal. This should be the case when the system has an empty spin-polarised state which may be occupied in the optical absorption process. Evidence for a strong magneto-optical signal of a supposed
empty spin-polarised $4f$-state was found in the paramagnetic LaSe metal [1-2]. This pioneering work motivated us to investigate the whole LaX series with X = S, Se and Te.

5.1.1 Previous Magneto-Optical Measurement of LaSe

Figure 5.1a shows the results of polar magneto-optical Kerr effect measurements on LaSe at 1.5 K and in a magnetic field of 10 T, performed by Pittini et al. [1]. A high, narrow Kerr rotation peak is observed at 2.77 eV, followed by a broader structure around 3 eV. The Kerr rotation reaches the remarkable height of $+2.1^\circ$. Figure 5.1b shows the real and imaginary parts of the complex off-diagonal components of the conductivity tensor, computed from Kerr effect spectra and optical functions, obtained after Kramers-Kronig transformations of reflectivity measurements (§ 3.1.2). Two structures with diamagnetic line-shape occur at 2.77 and 3.01 eV. Pittini et al. [1] stated that the sharp feature in $\theta_K$ and $\varepsilon_K$ was the direct magneto-optical manifestation of an electronic transition involving empty $f$-states. This interpretation was based on the fact that the sign of $\tilde{\sigma}_{xy}$ was opposite to that encountered when measuring the Ce-counterpart series [1,3]. Moreover, the additional line found at about 3 eV (i.e., at the high-frequency side of the sharp
resonance in $\theta_K$ and $\varepsilon_K$ was considered proof that both $4f^{\uparrow}$ spin-orbit states may be optically populated, in contrast to the CeX series, in which only the $4f^{\uparrow}$ ground state contributes to the transition [1-3].

5.1.2 New Results and Discussion

Figure 5.2a shows our reflectivity $R(\omega)$ as a function of energy for the LaX series. One can clearly identify the deep minimum at the sharp onset of the metallic contribution, which leads to the so-called plasma-edge feature, in which course

![Graph showing reflectivity and optical conductivity for LaS, LaSe, and LaTe at 300 K.]

Fig. 5.2: Reflectivity a) and optical conductivity b) spectra for lanthanum monochalcogenides at room temperature.
$R(\omega) \rightarrow 100\%$ for $\omega \rightarrow 0$. This plasma-edge, which also defines the screened plasma frequency, occurs at 3.0, 2.7 and 2.2 eV for LaS, LaSe and LaTe, respectively. Reflectivity minimum values decrease from LaTe to LaSe and LaS reaching the very low value of 0.5% in LaS.

In order to perform Kramers-Kronig transformations and calculate the optical functions, reflectivity data were extrapolated towards low energies, according to the Hagen-Rubens extrapolation (eq. (3.12)), with $dc$ conductivity values at 300 K extracted from Fig. 2.1 [14]. Figure 5.2b shows the real part of optical conductivity $\sigma(\omega)$ as a function of energy. This quantity is characterised by a Drude-like component below 3 eV and by several absorptions ascribed to electronic interband transitions (i.e. $p \rightarrow d$ interband transitions) between 3 and 12 eV. Both the onset of interband transitions (OIT) and the maximum in $\sigma(\omega)$ (MS1) shift towards lower energies from LaS to LaTe. The following values, for OIT and MS1 respectively, are extracted from $\sigma(\omega)$ curves: 3.3 and 5.3 eV for LaS, 2.8 and 4.8 eV for LaSe, and 2.0 and 4.1 eV for LaTe, with a total shift from LaS to LaTe of 1.3 for OIT and 1.2 eV for MS1. The explanation of these shifts must be sought both in the decrease of electronegativity from S to Te (§ 2.1, Ref. [15]), which reduces the distance between the valence $p$-band of the chalcogen atom and in the conduction $d$-band of lanthanum, and in the increased narrowness of the bands themselves (§ 2.1, Ref. [15]).

Figure 5.3 shows $\theta_k$ and $\varepsilon_k$ as functions of energy, measured at 2 K and at a magnetic field of 10 T. In all three compounds, we observe a very sharp, narrow peak in $\theta_k$, which occurs at 3.05, 2.63 and 2.2 eV for LaS, LaSe and LaTe, respectively. The narrowness of these peaks increases from LaS to LaSe and LaTe; the absolute values of the Kerr rotations are 0.99, 2.54 and 1.52 degrees, respectively. At higher energies, we observe the presence of a second, broader structure. This feature is well recognised only in LaTe between 3 and 5 eV. For LaS and LaSe, this absorption could not completely be resolved, since it fully develops at energies beyond the high-energy limit of our spectrometer (5.6 eV). Moreover, reflectivity values may also be extracted from the magneto-optical experiment (§ 4.2.3, eq. (4.5)). In the spectral range of the magneto-optical investigation, it was verified that $R(\omega)$ is temperature, and magnetic-field independent, in accordance with our direct measurement of $R(\omega)$ at 300 K and $H = 0$ (Fig. 5.2a).

Therefore, with $\theta_k$ and $\varepsilon_k$, and the optical functions $n$ and $k$ obtained at 300 K (for more details, see § A5.1), we can calculate (eqs. (3.37) and (3.38)) the off-diagonal components ($\tilde{\sigma}_{xy} = \sigma_{12y} + i\sigma_{21y}$) of the complex conductivity tensor.
Fig. 5.3: Magneto-optical Kerr effect of lanthanum monochalcogenides at 2 K and 10 T.

(§ 3.2.2). The latter quantities separate absorptive and dispersive parts (§ 3.2.2) and are shown in Fig. 5.4. The off-diagonal components are characterised on one
Fig. 5.4: Off-diagonal conductivity of lanthanum monochalcogenides at 2 K and 10 T.

hand by the strong absorption between 3 and 5 eV for LaTe and, on the other, by a sharp onset around 4 eV for LaS and LaSe. At least for LaTe, this absorption has a
diamagnetic line-shape. Moreover, there is a weak feature between 2 and 3 eV in all spectra.

Our magneto-optical LaSe spectra show some important and essential changes with respect to the previous investigation (§ 5.1.1, Ref. [1]). Although the peak at 2.63 eV is almost equally sharp in both measurements, the sign is opposite, being negative in our new spectra and positive in Ref. [1]. This is due to the incorrect calibration of the spectrometer and, in particular to the incorrect definition of the magnetic field direction in the previous investigation [1]. Moreover, for the whole series of compounds, we do not observe the second peak, found at approximately 3 eV in LaSe, as reported in Ref. [1]. Furthermore, as far as the conductivity tensor is concerned, there are now important differences with respect to the previous data [1]. Indeed, the feature between 2 and 3 eV is very sensitive to the values of $n$ and $k$ (§ 5.1) and is not so well pronounced in our new data of $\sigma_{33}$ and $\sigma_{22}$. In the first report, it was claimed that a weak absorption exactly coincides with the minimum of $R(\omega)$ at the onset of the plasma-edge [1], for which, however, we now do not find any clearcut evidence (§ 5.1). It was also suggested that this absorption, combined with the low values of the optical constants near the minimum in $R(\omega)$, amplifies the off-diagonal conductivity, extracted from the magneto-optical Kerr spectra [1,3]. We clearly establish here, with our new and comprehensive investigation of the whole LaX series, that this is not the case.

Instead, it is quite interesting to observe that the sharp structure in $\theta_K$ and $\varepsilon_K$ between 2 and 3 eV always and systematically occurs at the energy at which the onset of plasma-edge behaviour develops. We argue that the large, sharp Kerr effect in both $\theta_K$ and $\varepsilon_K$ follows directly from the interplay between an optical (interband) transition and the plasma resonance. This situation may be modelled by applying the phenomenological Lorentz-Drude fit (§ 3.2.5). Our goal here is mainly to discuss the essential features of the experimental data ($\theta_K$ and $\varepsilon_K$) with a minimum set of components and a realistic set of parameters in the fitting procedure.

For the whole LaX series, we considered a three-component picture, consisting of a Drude term for the free charge carriers and two harmonic oscillators (h.o.) for the optical transitions. We also assumed that the first harmonic oscillator, i.e., the one close to the plasma-edge, has an intrinsic polarisation. Figures 5.5, 5.6 and 5.7 present $\theta_K$ and $\varepsilon_K$ calculated within this phenomenological approach, for parameters reproducing the experimental conditions (magnetic field), and qualitatively estimating the intrinsic properties
(Drude component and Lorentz oscillators), compared with the corresponding experimental results of LaS, LaSe and LaTe, respectively. The parameter sets used in the calculation are reported in Tabs. 5.1, 5.2 and 5.3.

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$\varepsilon_{oo} = 3.84, B = 10$ T

**Tab. 5.1:** Fitting parameters for LaS.

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<tbody>
<tr>
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<td>$\gamma_1 = 0.67$ eV</td>
<td>$\omega_1 = 4.85$ eV</td>
</tr>
<tr>
<td>2nd h.o.</td>
<td>$\omega_{p2} = 5.31$ eV</td>
<td>$\gamma_2 = 3.38$ eV</td>
<td>$\omega_2 = 5.62$ eV</td>
</tr>
</tbody>
</table>

$\varepsilon_{oo} = 3.59, B = 10$ T

**Tab. 5.2:** Fitting parameters for LaSe.

<table>
<thead>
<tr>
<th>Drude</th>
<th>$\omega_p = 5.0$ eV</th>
<th>$\Gamma = 0.08$ eV</th>
<th>$f^+ = 1, f^- = 1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1st h.o.</td>
<td>$\omega_{p1} = 3.5$ eV</td>
<td>$\gamma_1 = 0.40$ eV</td>
<td>$\omega_1 = 3.5$ eV</td>
</tr>
<tr>
<td>2nd h.o.</td>
<td>$\omega_{p2} = 2.5$ eV</td>
<td>$\gamma_2 = 1.80$ eV</td>
<td>$\omega_2 = 4.0$ eV</td>
</tr>
</tbody>
</table>

$\varepsilon_{oo} = 4, B = 10$ T

**Tab. 5.3:** Fitting parameters for LaTe.

In spite of the simplicity of this model calculation, the main experimental features are well reproduced. First, the optical transitions (the first one is polarised) lead to the broad feature with onset at about 5 eV in LaS and LaSe, and centred at 3.5 eV in LaTe. Second, the calculation shows the appearance of the sharp feature in $\theta_K$ and $\varepsilon_K$ at the frequency of the plasma-edge onset. Moreover, the intensity of this feature is very sensitive to the degree of polarisation which, however, is extremely small (see Tabs. 5.1, 5.2 and 5.3). It is quite astonishing and
remarkable that such a minuscule but essential polarisation is enough to induce this very sharp and narrow Kerr effect. Probably, this polarisation may be reconciled with covalent-like mixing between the localised (empty) $f$ and delocalised $p$-$d$ band states. It is also important to stress that the usual plasma-edge

![Graphs for LaS and LaSe](image)

**Fig. 5.5:** Calculated Kerr rotation and ellipticity for LaS (Tab. 5.1) compared with experimental results.

**Fig. 5.6:** Calculated Kerr rotation and ellipticity for LaSe (Tab. 5.2) compared with experimental results.
splitting by the cyclotron frequency cannot alone account for the quite large magneto-optical signal [16]. The latter mechanism usually gives a magneto-optical response about 100 times smaller (Fig. 3.8). Nevertheless, enhancement of plasma-edge splitting and therefore of the magneto-optical signal may be achieved by assuming that a skew-scattering mechanism dominates [16]. Such a model, combined with the steep plasma-edge, successfully reproduces $\theta_k$ and $\varepsilon_k$ measured in TmS and TmSe [16]. In any case, the magneto-optical signal in the LaX series (Fig. 5.3) has a quite different shape from the one which would be expected in the case of simple plasma-edge splitting (Fig. 3.8).

Concerning the assignment of the original optical transition between 3.5 and 5 eV interplaying with the plasma resonance, we tend to speculate that $f$-states are involved, because of the necessary polarisation. Such a transition should be the one into the empty $4f^1$ state and its resonance frequency should coincide with $\omega_1$ in Tabs. 5.1, 5.2 and 5.3 for LaS, LaSe and LaTe, respectively. From $\omega_1$ numerical values, we have the indication of a first shift of 0.15 eV towards low energies from LaS to LaSe and a further shift of 1.35 eV, again towards low energies from LaSe to LaTe.

Our optical and magneto-optical investigation demonstrates the importance of the localised (empty) $f$-states in the electronic structure of the LaX series. Whereas in the previous analysis (§ 5.1.1, Ref. [1]), Pittini et al. believed that the

![Fig. 5.7: Calculated Kerr rotation and ellipticity for LaTe (Tab. 5.3) compared with experimental results.](image-url)
sharp, narrow structure of the Kerr effect response of LaSe was the direct magneto-optical manifestation of an interband transition into the $4f^{-1}$ empty state centred at $\sim 2.7$ eV [1], we believe that the presence of the optical interband transition into the $4f^{-1}$ empty state leads to a double magneto-optical signal, in view of:

a) the sharp feature at the frequency coinciding with the onset of the reflectivity plasma-edge and arising from the interplay between the interband (polarised) transition itself and the free charge carrier response;
b) the broad absorption developing at higher frequencies ($\sim 5$ eV) as a direct "fingerprint" of the optical (interband) transition.

The relevant fact evinced from the discussion within the phenomenological classical dispersion theory is the very small yet essential polarisation associated with the interband transition.

Lastly, as regards the numerical results of the model calculations for LaX, we would like to point out that:

- The parameter values in Tabs. 5.1, 5.2 and 5.3 were obtained with simultaneous calculation of the four quantities $R(\omega)$, $\sigma(\omega)$, $\theta_k$ and $\varepsilon_k$ and are a compromise between various sets of values best fitting a single quantity ($R(\omega)$ or $\sigma(\omega)$ or $\theta_k$ or $\varepsilon_k$). In particular, in order to obtain the sharp, narrow peaks in $\theta_k$ and $\varepsilon_k$ at the onset of the plasma-edge, in all the materials we set a damping value smaller than expected from the $R(\omega)$ and $\sigma(\omega)$ experimental curves, for both the Drude component and the polarised harmonic oscillator.
- Adding new harmonic oscillators (eventually polarised) in the calculation procedure might have produced better agreement over the whole spectral range with measured curves (especially for LaTe). However, in this way, the goal of the present discussion, i.e., to give a modelled picture of a simple system representing a realistic situation, would not have been reached.

5.1.3 Inverse Photoemission (IPE) Measurements

In collaboration with a research group from the Freie University of Berlin (Germany), an inverse photoemission (IPE) investigation of lanthanum monochalcogenides was undertaken [17].

Photoemission (PE) is a well-known technique to study $f$-electrons in rare-earth and actinide compounds. In photoemission experiments, a photon with
energy $h\omega$ is absorbed by the material under investigation. If the photon energy is 'high enough', an electron with kinetic energy $E_{kin}$:

$$E_{kin} = h\omega - E_B - \Phi_{cry} \quad (5.1)$$

is emitted. In eq. (5.1), $E_B$ is the electron bonding energy in the crystal and $\Phi_{cry}$ is the work required to remove the electron from the crystal. PE yields information about the status of the emitted electrons in the crystal.

Inverse Photoemission (IPE) is based on the inverse process, i.e. an electron is absorbed and a photon is emitted. Analogously to eq. (5.1), the energy of the emitted photon is:

$$h\omega = E_{kin} + \Phi_{cath} - E_B \quad (5.2)$$

where $\Phi_{cath}$ is the work of the electron gun. Indicating by $U_{acc}$ the difference of potential between the analysed sample and the cathode of the gun, it is $E_{kin} = e \cdot U_{acc}$. There are two different ways of performing IPE measurements:

1) The detection of photon intensity is performed at a fixed wavelength $\lambda_{ph}$ and the electron kinetic energy is varied.

2) The electron kinetic energy is fixed and photon intensity is measured over the whole spectral range.

With respect to the first, the second method, which is the one used to perform IPE measurements on LaX, has the advantage of better resolution (~0.3 eV, instead of ~0.6 eV of the first method) on the energy scale; it is also more appropriate for the study of unoccupied electronic band states. The disadvantages are the ultra-high vacuum environment, and the long measurement time, which creates problems related to surface effects [18-20].

Figure 5.8 shows IPE results on lanthanum monochalcogenides compared with lanthanum alone. In all samples, we observe that the measured points (empty circles in Fig. 5.8) are the result of two different contributions. The first one (dashed lines) comes from core levels of surface atoms and the second one (plain lines) from core levels of bulk atoms. These two contributions are shifted with respect to each other by the amounts of energy reported in Tab. 5.4 under the column SCS (Surface Core-level Shift). This shift is due to the different binding conditions to surface atoms with respect to bulk atoms, i.e., surface atoms have fewer neighbour atoms, which means fewer binding partners and therefore less binding energy. The position of the plain-line maximum indicates the position of
Fig. 5.8: IPE measurements for lanthanum monochalcogenides compared with pure lanthanum [17].

the empty bulk f-states (when the IPE experiment is in progress they are occupied by the electrons of the incident beam) with respect to the Fermi level (Tab. 5.4).
First of all, we notice that, in all materials, the position of the empty $f$-states is between 5 and 6 eV above the Fermi level. Second, from LaS to LaTe, a shift towards lower energies is observed. These two aspects are in good agreement with results found in magneto-optics (Figs. 5.3 and 5.4). In fact, in contrast with the

<table>
<thead>
<tr>
<th>Material</th>
<th>Energy Bulk 4f (eV)</th>
<th>SCS (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>La metal film</td>
<td>5.59 (±0.05)</td>
<td>0.69 (±0.03)</td>
</tr>
<tr>
<td>LaS</td>
<td>5.43 (±0.08)</td>
<td>0.53 (±0.03)</td>
</tr>
<tr>
<td>LaSe</td>
<td>5.26 (−0.15 and +0.10)</td>
<td>0.5 (−0.05 and +0.03)</td>
</tr>
<tr>
<td>LaTe</td>
<td>5.16 (−0.20 and +0.15)</td>
<td>0.52 (±0.1)</td>
</tr>
</tbody>
</table>

*Tab. 5.4: IPE measurements results for pure lanthanum and lanthanum monochalcogenides: energy of the bulk 4f level and Surface Core-level Shift [17].*

previous results of Pittini et al. [1], our new magneto-optical investigation shows that polarised $f$-states are found at about 5 eV, with a shift towards low energies going from LaS to LaTe. Comparing the values of $\omega_t$ in Tabs. 5.1, 5.2 and 5.3 and the position of the empty $f$-states in Tab. 5.4, we observe that the agreement is very good for LaS and LaSe and that for LaTe there is a difference of ~1.5 eV between the resonance frequency of the polarised oscillator in the magneto-optical calculations and the empty $f$-states position given by IPE measurements. On one hand, this difference may partially be explained by recalling that, in magneto-optics, we are measuring an interband transition with a covalent-like mixing character between localised (empty) $f$ and delocalised $p$-$d$ band states. The behaviour of the latter transitions is extracted from optical conductivity $\sigma_1(\omega)$ (Fig. 5.2b): the maximum of $\sigma_1(\omega)$ (already indicated with abbreviation MSI) is found at 5.3, 4.8 and 4.1 eV for LaS, LaSe and LaTe, respectively, showing a shift from LaS to LaTe of 1.2 eV. This shift is ~0.9 eV larger than that of the position of empty $f$-states from LaS to LaTe, as derived from IPE results. On the other hand, in the IPE technique one electron is added to the crystal, which is then negatively charged. The new electronic configuration may produce effects on the relative energy position of the energy bands with respect to the Fermi level, particularly of the empty $f$-states.
5.2 GdS

The role played by $f$-states in the magneto-optical spectra of lanthanum monochalcogenides has been extensively discussed in the last paragraph. In order to clarify better this role, especially with respect to the energy ‘position’ of the $f$-states, the gadolinium counterpart of LaS, GdS, was investigated. As already seen in § 2.2, GdS orders antiferromagnetically below 62 K, and electronic $f$-states are situated between 7 and 9 eV below the Fermi energy level [21]. Therefore, energies clearly higher than the high-energy limit of 5.6 eV of our magneto-optical spectrometer are required in order to excite $f$-electrons into the free states of the conduction $d$-band.

5.2.1 Results and Discussion

Figure 5.9 presents reflectivity and the real part of the optical conductivity as functions of energy for GdS compared with the analogous quantities for LaS (see also Fig. 5.2). Both materials show metallic behaviour, since reflectivity presents a clear plasma-edge and goes to 100\% at low energies. GdS has a sensible lower $R(\omega)$ in the mid-infrared and infrared spectral range. This trend is in agreement with the $dc$ value: the resistivity of GdS (Fig. 2.5) is almost one order of magnitude larger than that of LaS (Fig. 2.1). Like LaS, also GdS shows plasma-edge behaviour with a deep reflectivity minimum at 3.1 eV. At higher energies, the onset of interband transitions is evident.

In order to perform Kramers-Kronig transformations, the experimental data were extrapolated towards low energies using the Hagen-Rubens extrapolation with $dc$ conductivity values at 300 K taken
5. Results and Discussion

from Fig. 2.5 [22-24]. \( \sigma(\omega) \) clearly shows Drude behaviour at low energies and some interband transitions at \( \hbar \omega > 4 \) eV, i.e. \( p \rightarrow d \) interband transitions (Fig. 2.6). Magneto-optical Kerr rotation and ellipticity measured at 2 K and 80 K and at a magnetic field of 10 T (Figs. 5.10a and 5.10b) show a single structure centred at 3.15 and 3.10 eV for 2 and 80 K, respectively. There is also a very small onset in \( \theta_K \) at 5 eV. As already pointed out for the LaX series, the position of this structure coincides with the reflectivity minimum and, in spite of the fact that the two measurements were performed at lower (2 K) and higher (80 K) temperatures than the Néel temperature \( (T_N = 62 \) K), temperature dependence is very small. Comparing the \( \theta_K \) and \( \varepsilon_K \) of GdS with the corresponding measurements for LaS (Fig. 5.3), we observe that the structure in GdS is less sharp and narrow and that the magnitude of the highest Kerr rotation is lower, being \(-0.35^\circ \) in GdS and \(-0.99^\circ \) in LaS. Another difference is the presence of the relatively high, sharp positive peak of \(+0.2^\circ \) in the Kerr rotation \( \theta_K \) of GdS at 3.75 eV. In the spectral range of the magneto-optical investigation, it was verified that GdS reflectivity is temperature and magnetic-field independent, in accordance with our direct measurements of \( R(\omega) \) at 300 K and \( H = 0 \) (Fig. 5.9).

Therefore, with \( \theta_K \) and \( \varepsilon_K \), and the optical functions \( n \) and \( k \) obtained at 300 K (for more details, see § A5.1.) the off-diagonal components of the complex
conductivity tensor were calculated (eqs. (3.37) and (3.38), § 3.2.2). Figure 5.11 shows the real and imaginary parts of the off-diagonal conductivity at 2 K and 10 T. They are one order of magnitude smaller with respect to the corresponding quantities of LaS (Fig. 5.4) and indicate the presence of a weak interband transition with a paramagnetic line-shape centred at 3.7 eV.

We believe that the structure in $\theta_k$ and $\epsilon_k$ arises, as in LaX, from the interplay between the Drude component and an interband transition involving polarised states at higher energies. In general, peak sharpness and narrowness are related to the scattering rate of both the Drude component and the harmonic oscillator (§ 3.2.5). Comparing the reflectivity curves of GdS and LaS (Fig. 5.9), we notice that the minimum in LaS is deeper and the decrease in reflectivity steeper than in GdS. This may explain why the magneto-optical structure in GdS is broader than in LaS. Furthermore, peak magnitude is mainly related to the polarisation of the harmonic oscillator and to the relative position of the oscillator (the resonance frequency) with respect to the plasma-edge.

In order to give a more quantitative picture, $\theta_k$ and $\epsilon_k$ with eqs. (3.43), (3.33) and (3.34) were calculated for GdS in the same spirit as in § 5.1.2. Figure 5.12 shows the results of calculations with parameters in Tab. 5.5 compared with the experimental results. A three-component picture was considered, consisting of a Drude term for the free charge carriers and two harmonic oscillators for the optical transitions. It was also assumed that the first harmonic oscillator, i.e., the one close to the plasma-edge, had an intrinsic polarisation.

Precisely as the case of LaX, in order to obtain a narrow peak in $\theta_k$ at the onset of the plasma-edge in the calculation procedure, a smaller damping value had to be set which was smaller than the one expected from the experimental

**Fig. 5.11:** Off-diagonal conductivity of GdS at 2 K and 10 T.
curves for $R(\omega)$ and $\sigma_1(\omega)$ both for the Drude component and the polarised harmonic oscillator. In this way, only qualitative agreement was obtained with these experimental curves. Furthermore, in the case of GdS, in order to fit the positive peak in $\theta_K$ at 3.75 eV, a very small spectral weight had to be set in the polarised oscillator and consequently that of the second oscillator (unpolarised) for screening the plasma frequency was increased. With this set of parameters, $\theta_K$ was very well fitted, whereas for $\varepsilon_K$ only the main trend is reproduced (Fig. 5.12). Interestingly enough, a new calculation with another set of parameters was carried out. The difference with respect to the first set (Tab. 5.5) is simply that the first oscillator was eliminated and the polarisation (which is always necessary in order to obtain the right line-shape with the right intensity) was ‘transferred’ to the second oscillator. With this new set of parameters, the changes in the calculated
\( R(\omega) \) and \( \sigma_1(\omega) \) are very small (the spectral weight of the first oscillator is small). Figure 5.13 shows the results with this new set of parameters for \( \theta_K \) and \( \varepsilon_K \). Because of the larger spectral weight of the second oscillator, the values for \( f^+ \) and \( f^- \) are: \( f^+ = 1.0024 \) and \( f^- = 0.9986 \), i.e., smaller than with the first set of parameters. As expected, in this second case, the positive peak in \( \theta_K \) at 3.75 eV was not reproduced, whereas \( \varepsilon_K \) was very well reproduced.

Since we are now left with two different parameter sets, Figs. 5.14a and 5.14b compare the measured off-diagonal components of the conductivity tensor with the corresponding quantities calculated with the first and second parameter sets, respectively. In the first case (Fig. 5.14a), quite a good agreement is obtained and, in particular, the paramagnetic structure at 3.75 eV is very well reproduced. In the second case (Fig. 5.14b), the agreement is much worse and the paramagnetic structure is shifted to about 5 eV. These results may easily be explained as follows: in Fig. 5.14a, the paramagnetic structure at 3.7 eV is the direct manifestation of the ‘light’ (i.e. small oscillator strength) harmonic oscillator (polarised) centred at 3.73 eV; while in Fig. 5.14b this oscillator is absent and is ‘substituted’ by the ‘heavy’ (i.e. large oscillator strength) harmonic oscillator (polarised) centred at 4.75 eV.

Concerning the assignment of the optical transition interplaying with the plasma resonance, we speculate that the gadolinium \( f \)-states are probably involved.

**Fig. 5.13:** Calculated Kerr rotation and ellipticity for GdS (Tab. 5.5, with a small modification, see text) compared with experimental results.
5. Results and Discussion

Fig. 5.14: a) Calculated off-diagonal conductivity for GdS with parameters of Tab. 5.5, and b) with parameters of Tab. 5.5 with a small modification (as Fig. 5.13, see text), compared with experimental results.

because of the necessary polarisation, although the bare $f$-states are situated between 7 and 9 eV under the Fermi level (§ 2.2, Ref. [21]). This polarisation may be reconciled with a covalent-like mixing between localised $f$ and delocalised $p$-$d$ band states. It is not so clear how this covalent-like mixing manifests itself. With our model calculations, we have given two different pictures: in the first we have an additional polarised oscillator centred at 3.7 eV and in the second we have given an 'f-character' (through a very weak polarisation) to $p$-$d$ transitions. The
second picture seems to give a more ‘realistic’ scenario and is similar to the case of LaS, but the first one shows better agreement with the experimental results (especially with respect to off-diagonal conductivity). As a final comment, it should be noted that differences between the LaS system and GdS are generally to be expected at least because LaS is an empty f-state system whereas in GdS the f-states are occupied. In any case, since the Kerr rotation peaks of LaS and GdS in the reflectivity minimum region are both negative, these differences do not manifest themselves in a change of sign in the magneto-optical Kerr effect when empty and occupied f-state systems (i.e. LaS and GdS, respectively) are measured, as believed by Pittini et al. (§ 5.1.1, Ref. [1]).

Lastly, because of the very small difference in $\theta_K$ and $\varepsilon_K$ between the measurements at 2 and 80 K, the magnetic ordering of GdS does not appear to play an important role in its magneto-optical response. Indeed, magnetisation (Fig. 5.15) as a function of temperature measured at 10 T shows a small difference between 2 K and 80 K (i.e. $M = 0.89\mu_B$ and $M = 0.73\mu_B$ at 2 and 80 K respectively). We believe that this small difference, combined with temperature broadening effects, may explain the very small decrease of the absolute values of $\theta_K$ and $\varepsilon_K$ over the whole spectral range at 80 K with respect to the same values at 2 K.

Fig. 5.15: Magnetisation of GdS as a function of temperature at 10 T.
5.3 Eu(Gdx)O

The optical and magneto-optical results of LaX and GdS showed that a structure in \( \theta_k \) and \( \varepsilon_k \) in the region where the \( R(\omega) \) minimum occurs is in principle always to be expected in materials exhibiting plasma-edge behaviour and interband transitions involving different oscillator strengths for RCP and LCP. Since LaX and GdS are paramagnetic and antiferromagnetic metals respectively, we wished to analyse a ferromagnetic system exhibiting plasma-edge behaviour. Eu(Gdx)O is ferromagnetic under 69 K (§ 2.3) and seems to show plasma-edge behaviour at least for 'sufficiently highly' doped samples (§ 5.3.1 and 5.3.2). Furthermore, as will be shown in § 5.3.1, in this system the position of the minimum in \( R(\omega) \) may be shifted towards higher energies simply by increasing the amount of gadolinium as doping element.

5.3.1 Previous Optical and Magneto-Optical Investigations of Eu(Gdx)O

The literature contains extensive analyses both of the optical properties of EuO and Gd-doped EuO [4-8] and of the magneto-optical properties of EuO [8,10-11]. We briefly summarise the results.

Reflectivity measurements at room temperature from \( 5\times10^{-3} \) to 12.5 eV have been performed on pure EuO and Gd-doped EuO samples with different degrees of doping. Figure 5.16 shows the results [5-6]. Measurements of optical absorption and Faraday rotation at different temperatures have also been performed, and a magnetic red shift of the absorption edge with decreasing temperature has been found [9].

The first thing that the authors of Refs. [5-7] note is that the semiconductor-to-metal transition occurring almost at the Curie temperature in both EuO and Gd-doped samples is not found in the optically analysed samples. This observation is in sharp contrast with the contents of § 2.3 [25-27]. The authors of Refs. [5-7] believe that the samples used for transport measurements, in addition to Gd, also incorporated a sizeable amount of oxygen vacancies, which are the cause of semiconductor-metal transition near \( T_C \). They claim that, with oxygen vacancies as the only defect, a semiconductor-metal transition is found, but with Gd-doping alone a semiconductor-metal transition is absent. From analysis of the optical data, the authors also derived carrier concentrations at various temperatures (\( n(T) \)).
behaviour of $n(T)$ in the different samples does not show the predicted semiconductor-metal transition [25-27].

![Reflectivity curves](image)

**Fig. 5.16:** Reflectivity measurements of $Eu_{1-x}Gd_xO$ at 300 K for various values of $x$ [5-6].

The reflectivity curves in Fig. 5.16 show that:

a) At least for the pure EuO samples and for the 0.66% Gd-doped ones, reflectivity does not go to 100% for low energy values, which is a clear indication of the non-metallic character of these samples at room temperature. At higher concentrations of Gd, plasma-edge behaviour develops.

b) On increasing the Gd concentration, the deep reflectivity minimum of EuO, which occurs at 0.06 eV, moves to higher energies and seems to saturate at $\sim$0.5 eV.

c) At energies higher than 2 eV, there are no evident differences between the pure and Gd-doped EuO samples.
Figures 5.17a and 5.17b show measurements of the polar magneto-optical Kerr effect on pure EuO in the spectral range 0.5 - 5.3 eV and the corresponding calculated off-diagonal components of the conductivity tensor, respectively [8,10]. Besides the remarkably large magnitude of both Kerr rotation and ellipticity (−7°

for $\theta_K$ at 1.4 eV and −6° for $\varepsilon_K$ at 1.6 eV), the authors assign the two main structures in $\tilde{\sigma}_{xy}$ occurring at 1.3 (A) and 5.0 eV (B) to transitions from the localised 4f$^7$ state to the coupled 5d$^1$4f$^6$ final state [11]. The 3.7 eV energy separation between the two transitions is assigned to the 3.7 eV crystal field splitting of the 5d states [28] (see Fig. 5.18 from Ref. [8]).

5.3.2 New Results and Discussion

Our optical and magneto-optical analysis was performed both on a doped EuGd$_x$O sample with a nominal value of $x = 1.6\%$ and on a pure EuO sample. Figures 5.19a and 5.19b show reflectivity $R(\omega)$ and real part of the optical conductivity $\sigma(\omega)$ as a function of energy at various temperatures for the doped sample. We observe:

- A metallic behaviour, since reflectivity tends to 100% at low energies.
- A general increase in the spectral weight of the Drude component (free electrons) on lowering temperature.
- The presence of the plasma-edge between 0.4 and 0.5 eV, the position of which is temperature-independent.
- Various temperature-independent interband transitions between 1 and 12 eV.

Fig. 5.18: EuO atomic energy-level scheme [8].

Hagen-Rubens (H-R) extrapolation of the reflectivity spectra was carried out choosing the $\sigma_{dc}$ values in best agreement with the reflectivity points measured in the lowest part of the spectral range. Figure 5.20 shows $\sigma_{dc}$ values at various temperatures used for H-R extrapolation. For comparison, the resistivity of EuGd$_x$O with $x = 2\%$, already presented in Fig. 2.7 [26], is also shown. The agreement between the two data sets is quite good, indicating the metallic character of our sample, at least from the optical point of view.
Fig. 5.19: Reflectivity a) and optical conductivity b) of EuO + 1.6 % Gd at various temperatures.

Figures 5.21a and 5.21b show $R(\omega)$ and $\sigma_1(\omega)$ as functions of energy at various temperatures for the pure EuO sample. With respect to the doped sample, we observe that:
The pure EuO is clearly non-metallic, since reflectivity does not tend to 100% at low energies (for \( \hbar \omega < 0.025 \) eV, the measured data were completed according to Fig. 5.16 [5-6]).

There is no relevant temperature dependence; i.e., only the phonon centred at \( \sim 0.028 \) eV shows temperature dependence and becomes sharper at low temperatures.

In the absence of the Drude component (there are no free electrons), the minimum in \( R(\omega) \) is shifted down to \( \sim 0.06 \) eV.

For \( \hbar \omega > 1 \) eV, \( R(\omega) \) and \( \sigma(\omega) \) for pure EuO show the same behaviour as the same quantities for the Gd-doped sample (Figs. 5.19a and 5.19b).

Figure 5.22 shows \( \theta_K \) and \( \varepsilon_K \) for Gd-doped EuO in the spectral range 0.23 - 5.5 eV, at 2 K and at a magnetic field of 5 T. Comparing these results with the previous measurements on pure EuO performed in the spectral range 0.5 - 5.3 eV at 10 K and 4 T (Fig. 5.17a) we note that the general trend is similar, but there are the three following important differences:

1) In the doped sample, in the low energy part of the spectral range, a new, sharp, narrow feature between 0.4 and 0.7 eV is measured, in which \( \theta_K \) increases to +3.5° at 0.55 eV and then decreases to -1.34° at 0.62 eV, while \( \varepsilon_K \) reaches the very large value of +7.54° at 0.58 eV. This feature develops close to the low-energy limit of the spectrometer used for the previous measurements [8,10-11].

2) The measurement of the doped sample is slightly ‘shifted’ towards high energies with respect to measurements [8,10-11] on pure EuO.

3) The structure at \( \sim 5 \) eV in the doped sample is not as well pronounced as in the undoped sample.

The origin of these differences cannot be due to the different experimental conditions of the two measurements, as the Gd-doped sample was measured in the same experimental conditions as the undoped sample (10 K and 4 T, Fig. 5.17a) and, for energies higher than 0.8 eV, no difference was found with respect to the
Fig. 5.21: Reflectivity a) and optical conductivity b) of pure EuO at various temperatures. The inset shows optical conductivity between 0.023-0.034 eV.

measurements on the same Gd-doped sample at 2 K and 5 T (Fig. 5.22). It is argued here that the new structure at low energies arises because of the presence of the plasma-edge in the same spectral region (Fig. 5.19a). Actually, in optical
measurements, i.e. without any externally applied magnetic field, the reflectivity minimum occurs between 0.4 and 0.5 eV. Representing on the same graph magneto-optical Kerr rotation $\theta_K$ measurements in the low-energy range together with the reflectivity extracted directly from the magneto-optical spectrometer measured in the same experimental conditions (Fig. 5.23), we can directly appreciate how the new structure, centred between 0.5 and 0.6 eV, ‘follows’ the position of the reflectivity minimum. In comparison, Fig. 5.23 also shows the
reflectivity at 10 K and without any magnetic field (Fig. 5.19a). The shape of this new structure is different from that found in LaX and GdS. A possible explanation is given below.

![Graph](image)

**Fig. 5.23**: Kerr rotation $\theta_K$ and reflectivity of EuO + 1.6 % Gd measured at the same experimental condition, compared with reflectivity data at 10 K and without magnetic field.

Off-diagonal conductivity components were calculated, as usual, from $\theta_K$ and $\varepsilon_K$ and from $n$ and $k$ obtained after Kramers-Kronig transformations of reflectivity data. With respect to LaX and GdS, in this case, for Kramers-Kronig transformations we used the reflectivity extracted from the magneto-optical
spectrometer, since, in the presence of the magnetic field, the position of the reflectivity minimum is shifted by \( \sim 0.1 \) eV towards higher energies with respect to the reflectivity minimum position with no magnetic field (Fig. 5.23). This point is discussed in detail in § A5.1.

Figure 5.24 shows \( \sigma_{1xy} \) and \( \sigma_{2xy} \) over the spectral range 0.23 - 5.0 eV. Comparing these quantities with those already presented for pure EuO (Fig. 5.17b), there are only a few small differences: structure A is shifted from 1.3 to 1.5 eV, and structure B is also shifted towards high energies, since it is not completely resolved for \( \hbar \omega \leq 5 \) eV. These two structures are the magneto-optical evidence of transitions from the localised \( 4f^7 \) state to the coupled \( 5d^14f^6 \) final state (§ 5.3.1, Ref. [11], Fig. 5.18 from Ref. [8]). In the low part of the spectral range, i.e. where the new, sharp, narrow structure in \( \theta_K \) and \( \varepsilon_K \) arises, we have no indication of any interband transition occurring (see § A5.1).

Interestingly enough, we also measured the magneto-optical response of pure EuO down to 0.23 eV. Figure 5.25 shows \( \theta_K \) and \( \varepsilon_K \) of EuO measured at \( T = 10 \) K and \( B = 4 \) T, i.e. in the same experimental conditions as the old measurement on pure EuO (Fig. 5.17a), in the low-energy spectral range compared with Gd-doped EuO \( \theta_K \) measurements at 2 K and 5 T (Fig. 5.22) and with the old measurements on pure EuO (Fig. 5.17a). Besides the very good agreement with the old measurement over the whole spectral range (in Fig. 5.25 the high-energy spectral range is not shown because the two measurements perfectly overlap), we observe that pure EuO does not show any structure in \( \theta_K \) and \( \varepsilon_K \) between 0.23 and 0.8 eV. Neither do the corresponding off-diagonal components \( \sigma_{1xy} \) and \( \sigma_{2xy} \) show any structure between 0.23 and 0.8 eV. This result is not surprising. It confirms once again that the structure at 0.5 eV in Gd-doped EuO occurs at the onset of
plasma-edge behaviour. In fact, between pure and doped EuO, there are two fundamental differences:

1) EuO is a semiconductor and not a metal, i.e., there is neither plasma-edge behaviour nor a Drude component and consequently no interplay with the interband electronic transition at higher energies.

2) The reflectivity minimum of pure EuO corresponding to the plasma-edge in doped EuO is found below the low-energy limit of our magneto-optical spectrometer, i.e. 0.06 eV (Fig. 5.21a).

\[ \text{EuO} \]
\[ T = 10 \, \text{K} \]
\[ B = 4 \, \text{T} \]

\[ \theta_K \]
\[ \varepsilon_K \]

Fig. 5.25: Magneto-optical Kerr effect of EuO at 10 K and 4 T compared with old measurements of Reim et al. [8,10] on the same material and with \( \theta_K \) of EuO + 1.6% Gd at 2 K and 5 T already shown in Figs. 5.22 and 5.23.

The situation can be modelled using the phenomenological model described in § 3.2.5 and already used for LaX and GdS. In this case, we considered a four-
component picture, consisting of a Drude term for the free charge carriers and three harmonic oscillators for the optical transitions. We concentrated our calculations on the lower part of the spectral range, without taking into account the structure B at 5 eV (Fig. 5.22). Figure 5.26 shows $R(\omega)$, $\sigma(\omega)$, $\theta_K$ and $\varepsilon_K$ calculated with the parameters of Tab. 5.6 compared with the experimental results of doped EuO. The main features of all the experimental curves are qualitatively and quantitatively very well reproduced. However, as already noted, the feature at low energies in $\theta_K$ and $\varepsilon_K$ shows a different line-shape with respect to LaX and GdS. In order to reproduce it with model calculations, the first oscillator interplaying with the Drude component must be unpolarised (in LaX and GdS it was polarised), while the Drude component must be polarised (in LaX and GdS it was unpolarised). This reveals one difference in the physical mechanism originating the large Kerr effect in the reflectivity minimum region in LaX and GdS with respect to Gd-doped EuO. In LaX and GdS, the presence of an interband transition involving polarised electronic states ($f$-states) is fundamental, whereas in doped EuO the polarisation of the Drude component is more relevant in order to obtain both the magnitude and the line-shape of the experimental results. The polarisation of the free electrons is induced by the external magnetic field, which produces an energy shift of the bottom of the conduction band, proportional to the cyclotron frequency. The situation is represented in Fig. 5.27. The two sides of the conduction band show free electrons with spin-up and spin-down and their volume density $n^\uparrow$ and $n^\downarrow$, respectively. Consequently, with no external magnetic field ($B = 0$), we have $n^\uparrow = n^\downarrow$, which in our model means $f^+ = f^-$ for the Drude component. With $B \neq 0$, we have $n^\uparrow \neq n^\downarrow$, which in our model means $f^+ \neq f^-$ for the Drude component.

We may conclude that the physical mechanism which generates Kerr effects in the reflectivity minimum region is revealed by the shape of the structure itself,
which is directly related either to the interband polarised electronic transitions interplaying with the Drude component or to the polarisation of the free electrons.

We would like now to discuss briefly the case of pure EuO. The difference with respect to Gd-doped EuO is in the presence of a phonon centred at 0.028 eV and the lack of the Drude component. If we substitute the Drude component in Tab. 5.6 (Gd-doped EuO case) with a Lorentz oscillator centred at 0.028 eV, leaving the other parameters unchanged, the calculated $\theta_K$ and $\varepsilon_K$ show no change.

![Graphs showing measured and calculated data for reflectivity, optical conductivity, Kerr rotation, and ellipticity](image1.png)

**Fig. 5.26:** Calculated reflectivity, optical conductivity, Kerr rotation $\theta_K$ and ellipticity $\varepsilon_K$ for EuO + 1.6 % Gd (Tab. 5.6), compared with experimental results.
with respect to the curves of Fig. 5.26. This result is actually a generalisation of the concept of \textit{interplay} between the Drude component and a Lorentz oscillator at 1st h.o.\hspace{1cm} \omega_{p1} = 0.115 \text{ eV} \hspace{1cm} \gamma_1 = 0.001 \text{ eV} \hspace{1cm} \omega_\infty = 0.0284 \text{ eV} \hspace{1cm} f_{1+} = 1, f_{1-} = 1

2nd h.o.\hspace{1cm} \omega_{p2} = 1.700 \text{ eV} \hspace{1cm} \gamma_2 = 1.200 \text{ eV} \hspace{1cm} \omega_\infty = 0.9000 \text{ eV} \hspace{1cm} f_{2+} = 1, f_{2-} = 1

3rd h.o.\hspace{1cm} \omega_{p3} = 0.800 \text{ eV} \hspace{1cm} \gamma_3 = 0.600 \text{ eV} \hspace{1cm} \omega_\infty = 1.9000 \text{ eV} \hspace{1cm} f_{3+} = 2, f_{3-} = 0

4th h.o.\hspace{1cm} \omega_{p4} = 3.500 \text{ eV} \hspace{1cm} \gamma_4 = 2.000 \text{ eV} \hspace{1cm} \omega_\infty = 3.1000 \text{ eV} \hspace{1cm} f_{4+} = 1, f_{4-} = 1

\begin{align*}
\epsilon_\infty = 1, B = 5 \text{ T}
\end{align*}

\textbf{Tab. 5.7:} Fitting parameters for pure \textit{EuO}.

higher energies, as already discussed. In fact, we can now say that the occurrence of a steep, deep minimum in the reflectivity curves is, in principle, enough to give rise to large Kerr effects in the spectral region where the reflectivity minimum occurs. Indeed, calculations performed with a set of parameters suitable for pure \textit{EuO} (Tab. 5.7) show that a large Kerr effect may be found around 0.06 eV, i.e. where the reflectivity minimum occurs due to the ‘plasma-edge-like’ shape of $R(\omega)$ on the high-frequency side of the phonon absorption (Fig. 5.28). Its magnitude and line-shape depend on the polarisation of the Lorentz oscillators, which \textit{interplay} together and lead to the formation of the minimum in $R(\omega)$. Unfortunately, the hypothetical beha-
viour of $\theta_K$ and $\varepsilon_K$ at 0.06 eV is below the low-energy limit of our magneto-optical spectrometer (0.23 eV).

![Graph](image)

**Fig. 5.28:** Left figure: calculated reflectivity and Kerr rotation $\theta_K$ (inset) for EuO (Tab. 5.7) compared with experimental results; right figure: low-energy range calculated Kerr rotation and ellipticity.

### 5.4 CeSb

The search for materials with very large magneto-optical Kerr rotations has attracted always much interest. The 14° Kerr rotation first discovered in CeSb by Reim et al. [8,12] late in the 1980s and the even more astonishing 90° Kerr rotation found in the same compound by Pittini et al. ten years later (over a broader spectral range) [13] caused much excitement. The occurrence of such a very large magneto-optical Kerr rotation in materials at the borderline between well-localised and itinerant $f$ electron states also attracted much interest from the theoretical viewpoint [29-33].

We were motivated first of all to reproduce the giant 90° Kerr rotation [13] on other specimens: besides the general need for reproducibility, we wanted to establish sample independence and the intrinsic nature of this effect. Second, since Pittini et al. found the 90° Kerr rotation at a magnetic field of 5 T [13], we wanted to investigate what happens to the ‘resonance’ upon increasing the magnetic field
up to 10 T. The latter aspect is particularly intriguing if the 90° Kerr rotation is confirmed to be truly an intrinsic effect. The question was whether above 5 T the Kerr rotation stays constant and saturates to such a maximum value or eventually decreases again.

5.4.1 Previous Magneto-Optical Investigation of CeSb

Magneto-optical measurements of the polar Kerr effect had already been performed on CeSb by Reim et al. in 1986 [8,12] and Pittini et al. in 1996 [13]. Their results are shown in Figs. 5.29a and 5.29b, respectively. The astonishing value of -14° in θ_k at 0.5 eV found by Reim et al. was at that time the highest Kerr rotation ever observed. Because of the low-energy limit of 0.5 eV in the spectrometer of Reim et al., this minimum in the Kerr rotation could not be completely resolved (Fig. 5.29a).

Fig. 5.29: a) Polar Kerr rotation and ellipticity of CeSb measured by Reim et al. [8,12] at 2 K and 5 T, and b) measured by Pittini et al. [13] at 1.5 K and 5 T.

Ten years later and using a spectrometer with an extended spectral range down to 0.23 eV, Pittini et al. found the even more astonishing result of a 90° Kerr rotation (with a 'jump' of 180°) at 0.46 eV (Fig. 5.29b). The two measurements were performed at the same externally applied magnetic field (5 T) and at the temperatures of 2.0 K (Reim et al.) and 1.5 K (Pittini et al.). The latter authors
explained the large differences with the previous investigation of the former by the improved resolution of their spectrometer on both energy and rotation scales. The small temperature difference in experimental conditions is not relevant, since the magnetisation of CeSb at 2 K is already saturated (Figs. 2.10 and 2.11).

Pittini et al. ascribed this 'resonance' at 0.46 eV to a paramagnetic 4f \rightarrow 5d transition located at 0.54 eV. The magneto-optical response associated with this transition was claimed to be enhanced by the vicinity of a strong 5p (Sb) \rightarrow 5d (Ce) transition and by the low values of the optical constants in the region of the minimum in \( R(\omega) \) at the onset of the plasma-edge [2,13].

5.4.2 New Results and Discussion

Figure 5.30 shows our reflectivity measurements on our samples compared with the equivalent measurements of Pittini [2] on his specimen. In both samples, \( R(\omega) \) is metallic-like, as indicated by the sharp rise at about 0.45 eV defining plasma-edge behaviour. We find a few differences between the two specimens, especially between 0.4 and 1.5 eV. In Pittini's measurement there are two sharp structures centred at 0.8 and 1.2 eV, which in our new measurements, are rather broad and tend to merge into a single structure. Moreover, it is worth noting that the original \( R(\omega) \) spectrum of Pittini's sample displays a sensible lower \( R(\omega) \) in the mid-infrared and infrared spectral range, in contrast to our brand-new crystals. This latter feature indicates incipient overdamped-like behaviour of the reflectivity and consequently an enhanced scattering rate of the free electrons. Interestingly enough we also verified that exposure to air for a few minutes considerably damages the crystal. Our air-contaminated CeSb crystal presents similar features in the \( R(\omega) \) spectrum as that reported by Pittini [2]. The \( R(\omega) \) on CeSb measured by Reim et al. is found in Ref. [12] and is in good agreement with our results.

Figure 5.31 shows the results of polar Kerr rotation \( \theta_K \) in the spectral range from 0.23 to 0.80 eV at 1.5 K and a magnetic field of 5 and 10 T. No magnetic field dependence can be recognised between 5 and 10 T (i.e., the magnetisation is saturated). Going from lower to higher energies, we observe a first maximum of +3.0° at 0.26 eV, followed by a very deep minimum of -16.9° at 0.46 eV. At higher energies, \( \theta_K \) increases, reaching the value of +0.8° at 0.79 eV.

We point out that this result is in very good agreement with the first magneto-optical measurement on CeSb performed by Reim et al. at 2 K and in a magnetic field of 5 T (Fig. 5.29a). At 0.5 eV (the low-energy limit of their spectrometer) a Kerr rotation of -14° was measured, while in our new
measurements we find values of $-9.6^\circ$, $-12.7^\circ$, and $-16.0^\circ$ at 0.52, 0.50, and 0.48 eV, respectively. At higher energies, our $\theta_k$ curve crosses the x-axis at 0.66 eV and reaches the value of $+0.8^\circ$ at 0.79 eV, again in very good agreement with the results of Reim et al. (Fig. 5.29a). Moreover, when comparing our measurements

(Fig. 5.31) and those of Reim et al. (Fig. 5.29a) with the results of Pittini et al. (Fig. 5.29b), we note that the latter found a minimum of $-13.2^\circ$ at 0.72 eV (Fig. 5.29b). Pittini et al. also claimed [13] that this minimum at 0.72 eV is similar to that of $-14^\circ$ found by Reim et al. [8,12] at 0.50 eV (Fig. 5.29a). Instead, our value and that of Reim et al. of $\theta_k$ at 0.72 eV is close to 0°, corresponding to more than 13° difference with that of Pittini et al. (Fig. 5.29b). Furthermore, at 0.5 eV, Pittini et al. found a Kerr rotation of about $+55^\circ$, with a huge difference of more than 65° compared with our new measurements (Fig. 5.31) and those of Reim et al. (Fig. 5.29b).

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**Fig. 5.30:** Reflectivity at 300 K for the new and Pittini's [2] CeSb sample.
Fig. 5.31: Polar Kerr rotation at 1.5 K and at 5 and 10 T for the new CeSb. The figure also displays the measurements on the same new sample obtained after 26 hours of air contamination. The inset shows our measurements of Pittini's sample, displaying the 90° Kerr rotation. The black dots in the main figure represent successive measurements of Pittini's sample (old) after air deterioration and surface polishing (see text).

Such differences cannot be explained by the improved resolution of the spectrometer on the energy and rotation scales, as believed by Pittini et al. [13]. In
fact, we measured the Kerr rotation at the fixed energy value of 0.5 eV, as a function of monochromator slit opening, and we found a change of less than one degree (Fig. 5.32) when reducing the slit opening from 2 mm (the experimental conditions of Reim et al. [8,12]) to 0.5 mm (our experimental conditions).

In the inset of Fig. 5.31, we show our results obtained on Pittini's specimen, noting that the sample was quickly inserted into the sample chamber of the $^3$He cryostat directly from the dry-box. The main feature (i.e. the 90° Kerr rotation and the 180° 'jump' occurring at 0.46 eV) could be reproduced although some differences with the old measurements (Fig. 5.29b) are evident: the Kerr rotation value at 0.3 eV is already higher than $-20^\circ$ while in Ref. [13] it is less than $-10^\circ$, and at 0.65 eV we still have a value of about $+40^\circ$ while in Ref. [13] $\theta_K$ it is close to 0°.

After having performed magneto-optical measurements (inset Fig. 5.31), the sample of Pittini et al. was re-analysed with optical reflectivity measurements. We were unable to reproduce our measurements on the new sample or repeat the original curve of Pittini (Fig. 5.30) [2]. The sample seems to have very great sensitivity to exposure to air and, even after polishing and quick transfer into the optical spectrometers, we did not find a systematic and reproducible optical response. Moreover, consecutive measurements of $R(\omega)$ in the same experimental conditions gave quite different results, indicating an instability of the sample or at least of its surface.

The sample of Pittini et al. was finally re-measured with the magneto-optical set-up for the second time in the same experimental conditions as the first measurement (i.e., inset of Fig. 5.31). To our great surprise, the magneto-optical response was completely different (full dots in Fig. 5.31). Pittini's sample was too thin for further cleaving. Therefore, we cannot exclude \textit{a priori} that polishing,
previously performed for $R(\omega)$ measurements, damaged the surface, leading to a different magneto-optical signal [8]. However, this result does not change the fact that the 90° Kerr rotation [13] does not seem to be a general intrinsic feature of CeSb; rather it reinforces the view that such a giant Kerr rotation was peculiar to Pittini's sample.

We speculate that sample deterioration (because of ageing or contamination effects) may explain this variety in data. In this respect, we purposely contaminated our new CeSb sample with air, in order to study ageing and degradation effects on the Kerr rotation signal. Although the reflectivity spectra were greatly affected by air contamination, we did not find any effects on $\theta_k$. Indeed, even after 26 hours’ exposure to air, we reproduced, with a difference of less than one degree, our magneto-optical measurements on the new sample over the whole energy range 0.3 - 0.7 eV (Fig. 5.31). For measurements on the air-contaminated sample, the surface was neither cleaved nor polished. Since from both crystallographic and stoichiometric points of view, the sample of Pittini et al. is of comparable quality as ours, we attribute the extraordinary magneto-optical effects to strange and still puzzling surface effects which may explain why the 90° Kerr rotation could not be reproduced anymore, neither on the new sample or on the old sample with a polished surface.

We believe that this Kerr rotation resonance is the consequence of the interplay between a (bare) interband transition at higher frequencies and the free electron plasma response alone. Pustogowa et al. [33] identify the general origin of giant Kerr rotations as due to mainly non-magnetic optical properties, in particular from the ratio of the dominant interband resonance frequency to the free charge carrier plasma frequency [33]. In the same spirit of Ref. [33], we calculated the optical and magneto-optical responses of CeSb according to § 3.2.5, using the set of parameters of Tab. 5.8. We considered a three-component picture, consisting of a Drude term for the free charge carriers and two harmonic oscillators for the optical transitions. Figures 5.33a, 5.33b and 5.33c show the results of the

<table>
<thead>
<tr>
<th>Drude</th>
<th>$\omega_p = 1.6$ eV</th>
<th>$\Gamma = 0.07$ eV</th>
<th>$f^+ = 1$, $f^- = 1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1st h.o.</td>
<td>$\omega_{p1} = 4.2$ eV</td>
<td>$\gamma_1 = 2.00$ eV</td>
<td>$\omega_1 = 1.6$ eV</td>
</tr>
<tr>
<td>2nd h.o.</td>
<td>$\omega_{p2} = 11.2$ eV</td>
<td>$\gamma_2 = 5.00$ eV</td>
<td>$\omega_2 = 4.4$ eV</td>
</tr>
</tbody>
</table>

Tab. 5.8: Fitting parameters for CeSb.
calculated $R$, $\sigma$, and $\theta_K$ compared with the experimental points, respectively. The main features of both reflectivity and optical conductivity are qualitatively and quantitatively very well reproduced over the whole spectral range. The small shift towards higher energies of the reflectivity minimum in the calculated curve with respect to the measurements at $T = 300$ K and $B = 0$ is in agreement with the
reflectivity at 2 K and 5 T extracted from the magneto-optical spectrometer (dots in Fig. 5.33a).

Regarding the calculations of Kerr rotation and ellipticity (Figs. 5.33c and 5.33d), we note that, in this case, some values obtained for $\theta_K$ do not satisfy eq. (3.60). This means that a mistake is being made in the calculation. In any case, by means of geometrical calculation, it can be shown that this mistake is only a few percent of the calculated point value. Also in the case of CeSb, our goal was mainly to discuss the essential features of the experimental data of $\theta_K$ with a minimum set of components and a realistic set of parameters in the fitting procedure.

The calculated Kerr rotation shows the $-17^\circ$ Kerr rotation and reproduces quite well the experimental points over the spectral range 0.23 - 0.8 eV. Figure 5.33d shows $\theta_K$ and $\varepsilon_K$ calculated over the whole spectral range, always with the same set of parameters as in Tab. 5.8. The calculation demonstrates once again that, also in the case of CeSb, the large Kerr rotation in the reflectivity minimum arises from the interplay between the Drude component and an interband transition at higher energies. Looking at the fitted parameters of Tab. 5.8, we note that this transition should be centred at $\sim$1.6 eV. Furthermore, because of the necessary polarisation (in this case the polarisation is much higher than in the other materials), we believe that $f$-states must be involved (i.e. $4f \rightarrow 5d$ electronic transitions).

Lastly, we point out that the model also describes very well the behaviour of CeSb at 10 T. In fact, as already shown in § 3.2.5, when the electronic interband transition interplaying with the Drude component has an intrinsic polarisation (as in the case of CeSb), the dependence of $\theta_K$ and $\varepsilon_K$ on the externally applied magnetic field intensity is very small. In fact, if in the calculation, with the set of parameters as in Tab. 5.8, we set the magnetic field value at 10 T, the resulting Kerr rotation and ellipticity show a difference of less that 0.1° over the whole spectral range. This result is in very good agreement with the experimental response. Furthermore, in CeSb, the magnetisation saturates at $\sim$4.5 T (Fig. 2.10); this means that, at least from the magnetic macroscopical point of view, there is almost no difference between 4.5 and 10 T. In our model, oscillator strengths $f^+$ and $f^-$ are directly related to the electronic structure of the material and to the relative occupation of the energy levels. The lack of difference in the $\theta_K$ measured points between 5 and 10 T demonstrates, as shown by the model, that the electronic structure does not change either.
Kerr effect measurements were also performed on a CeSb thin film sample oriented along the [100] plane and deposited on a sapphire substrate. The sample was grown by H. Meffert in the Johannes Gutenberg University of Mainz (Germany). Preliminary results at 2 K and 5 T show a negative peak of $-13^\circ$ in $\theta_k$ at 0.36 eV and a shape very similar to our new measurements on CeSb bulk samples at 2 K and 5 T (Fig. 5.31).
APPENDIX

A5.1 Off-Diagonal Conductivity in the Plasma-Edge Region: the Case of EuGd₃O

This section shows how sensitive, in the plasma-edge region, the off-diagonal components of the conductivity tensor are to even small differences in optical functions $n$ and $k$.

Let us consider the case of EuGd₃O ($x = 1.6\%$) already discussed in § 5.3. The reflectivity of this material presents small differences in the region where the reflectivity minimum occurs when it is measured with and without an externally applied magnetic field (Fig. 5.23). Figure 5.34 shows $n$ and $k$ calculated by Kramers-Kronig transformations of reflectivity data measured with (thick lines) and without (thin lines) a magnetic field. The corresponding off-diagonal conductivities calculated from $\theta_k$ and $\varepsilon_k$ as in Fig. 5.22 and from $n$ and $k$ with (thick lines) and without (thin lines) a magnetic field are shown in Figs. 5.35a and 5.35b, respectively. In Fig. 5.35b, the $n$ and $k$ curves for $B = 0$ (thin lines) clearly lead to the presence of a diamagnetic structure centred at $0.58$ eV in $\sigma_{1xy}$ and $\sigma_{2xy}$, for which, however, we do not have
any clearcut evidence in Fig. 5.35a, where $\sigma_{1xy}$ and $\sigma_{2xy}$ are calculated from $n$ and $k$ curves with a magnetic field (thick lines). We argue that, in the second case (Fig. 5.35b), the structure arising in the off-diagonal conductivity is an artefact due to lack of consistency when calculating the off-diagonal conductivity (using $B = 0$ values for $n$ and $k$) and that it does not correspond to a real interband transition.

It should be noted that this structure corresponds in both sign and shape to the structure found at 2.77 eV by Pittini et al. in LaSe [1] (§ 5.1.1, Fig. 5.1b).

![Off-diagonal conductivity calculated from reflectivity curves measured a) with, and b) without externally applied magnetic field (see text). Insets: off-diagonal conductivity shown between 0.2 - 1.0 eV.](image)

**Fig. 5.35:** Off-diagonal conductivity calculated from reflectivity curves measured a) with, and b) without externally applied magnetic field (see text). Insets: off-diagonal conductivity shown between 0.2 - 1.0 eV.
REFERENCES

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6. CONCLUSIONS

Fascinating results were revealed in the magneto-optical spectra of the materials analysed in this work. The most interesting feature is without doubt the sharp, narrow Kerr effect structure arising in the spectral region where the reflectivity minimum occurs. With the support of the Drude-Lorentz model, which in this work is applied to magneto-optics, it is shown that this structure is the consequence of the presence of both the Drude component (free electrons) and a harmonic oscillator (bound electrons) at higher energies. In order to achieve large Kerr effects, one of these two components must be polarised, and the shape of the magneto-optical structure itself may then distinguish between polarisation of the harmonic oscillator (LaX, GdS and CeSb) and polarisation of the free electrons (Gd-doped EuO). Furthermore, simulation of a Kerr effect measurement for the case of pure EuO gave an indication of a possible large Kerr effect centred at 0.06 eV. Since pure EuO is not metallic and consequently no Drude component is present, this result is actually a generalisation of the concept of interplay between the Drude and Lorentz components: the occurrence of a steep, deep minimum in the reflectivity curves is, in principle, enough to give rise to large Kerr effects in the spectral region where the reflectivity minimum occurs. The magnitude and
line-shape depend on polarisation of the Drude component and/or of the Lorentz oscillators, which lead to the formation of the minimum itself.

This result is of great importance in the field of magneto-optical Kerr effect spectroscopy because it points, *inter alia*, to a new approach to the search for materials with large Kerr rotations, which are of interest, e.g., for new possible methods of cheaper and more compact data storage in computer memories.

As task for the future, materials showing giant (or even colossal) magnetoresistance, like some hexaborides and a series of manganese oxides, have recently attracted our interest. These materials show drops of several orders of magnitude in resistivity and metallic behaviour upon the onset of ferromagnetic ordering. We are presently investigating europium hexaboride (EuB₆), which orders ferromagnetically under $T_C = 16$ K and shows plasma-edge behaviour with a recently discovered [1] giant blueshift in the reflectivity minimum concomitant with the onset of magnetic ordering. The measurement of excitation spectrum with magnetic field dependence should shed light on the interplay between magnetic ordering and related giant magnetoresistance and itinerant properties (like, e.g., mean free path, scattering relaxation rate, etc.). Moreover this work should reveal important information on the spin-charge-orbital coupling of this material. The behaviour of the plasma-edge in an external magnetic field could corroborate or question models, based on the moment polarisation for the ferromagnetic phase transition [2]. Our final goal is to possibly establish the existence of a scaling or simply a trend between temperature and magnetic field for a relevant energy scale of the material (plasma frequency, scattering rate or frequency phonon modes). This would allow a direct comparison between the *dc* magnetoresistance and the dynamics of the charge excitation spectrum.

REFERENCES


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September 1993 – June 1994  Diploma work supervised by Prof. Dr. Franco Marabelli on “Study of the Optical Gap in Copper Oxide Monocrystals” at the University of Pavia (Italy)


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March 1996 – Autumn 1999

Ph.D. work supervised by PD Dr. Leonardo Degiorgi in the group of Prof. Dr. Peter Wachter at the ETH Zürich on “Magneto-Optical Kerr Effect in the Plasma-Edge Region: Evidence for Interplay between Drude Term and Interband Transitions”
a) publications


b) conferences

• 22-25 April 1996, Stresa (I), Conference of the European Physical Society, EPS'96 (Poster).

• 19-22 August 1996, Zurich (CH): Conference on “Strongly Correlated Electron Systems”, SCES'96 (Member of the Organising Committee).

• 6-10 July 1997, Ascona (CH): Conference on “Low Energy Electrodynamics in Solids”, LEES’97 (Member of the Organising Committee - Poster).

