Doctoral Thesis

A highly sensitive $^{199}\text{Hg}$ magnetometer for the nEDM experiment

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A highly sensitive $^{199}$Hg magnetometer for the nEDM experiment

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Abstract

The search for a neutron electric dipole moment (nEDM) is closely linked to extensions of the Standard Model, since a nonzero EDM implies CP-violation. The so far best upper limit on the nEDM was published in 2006 by the RAL-Sussex-ILL collaboration ($|d_n| < 2.9 \times 10^{-26} e \text{cm}, 90 \% \text{C.L.}$). An international collaboration is searching for the nEDM at the Paul Scherrer Institute aiming at a sensitivity of $5 \times 10^{-27} e \text{cm}$ at $95 \% \text{C.L.}$ as an intermediate goal and $5 \times 10^{-28}$ at $95 \% \text{C.L.}$ as a final goal.

A crucial tool in the nEDM apparatus is a $^{199}$Hg cohabiting magnetometer, which monitors the magnetic field and allows one to correct for field drifts. The sensitivity of the nEDM apparatus depends among others on the sensitivity of the $^{199}$Hg magnetometer. There are several ways to improve the magnetometer. A feasible one is the increase of polarisation of the $^{199}$Hg vapour. Finding a way how to do this was part of my work at Paul Scherrer Institute (PSI). For this purpose a test setup was built in which polarisation and spin precession of $^{199}$Hg atoms can be observed. In the context of the studies, different volumes and wall coatings of the polarisation chamber of the $^{199}$Hg magnetometer were investigated.

With an increased sensitivity, the systematic effects have to be controlled on the same level. Using a mercury co-magnetometer the largest systematic effect is the geometric phase effect of $^{199}$Hg which is responsible for a false EDM signal. Planning and realisation of a measurement of the $^{199}$Hg geometric phase effect and a subsequent analysis of the taken data was also part of my work.
Zusammenfassung

Die Suche nach dem elektrischen Dipolmoment des Neutrons (nEDM) ist eng verbunden mit den Erweiterungen des Standardmodells, da ein von Null verschiedenes EDM CP-Verletzung impliziert. Die bislang beste Obergrenze des nEDMs wurde 2006 von der RAL-Sussax-ILL Kollaboration publiziert (|d_n| < 2.9 \times 10^{-26} \text{cm}, 90 \% \text{C.L.}). Eine internationale Kollaboration hat sich zum Ziel gesetzt das nEDM mit einer verbesserten Sensitivität von 5 \times 10^{-27} \text{cm} bei 95 \% \text{C.L.} als mittelfristigem und 5 \times 10^{-28} \text{cm} bei 95 \% \text{C.L.} als längerfristigem Ziel.

Ein wesentlicher Bestandteil des nEDM Apparates ist ein $^{199}\text{Hg}$ Co-Magnetometer, das das magnetische Feld misst, um dadurch Felddrifts zu korrigieren. Um den EDM-Apparat zu verbessern ist auch eine Verbesserung der Sensitivität des $^{199}\text{Hg}$ Magnetometers nötig. Die Sensitivität des $^{199}\text{Hg}$ Magnetometers hängt linear von der Polarisation des $^{199}\text{Hg}$ Dampfes ab. Die Suche nach einem Weg die Polarisation zu verbessern war Teil meiner Arbeit am Paul Scherrer Institut (PSI). Dafür war der Bau eines Testaufbaus nötig, in dem die Polarisation und die Spinpräzession von $^{199}\text{Hg}$ Atomen beobachtet werden kann. Im Rahmen meiner Studien wurden verschiedene Volumina und verschiedene Wandbeschichtungen für die Polarisationskammer des $^{199}\text{Hg}$ Magnetometers untersucht.

Bei einer verbesserten Sensitivität ist auch die Kontrolle von systematischen Effekten auf entsprechendem Niveau nötig. Der grösste systematische Effekt ist der Effekt der geometrischen Phasen von den $^{199}\text{Hg}$ Atomen, der ein falsches EDM-Signal erzeugt. Die Planung sowie die Durchführung der Messung der geometrischen Phasen von $^{199}\text{Hg}$ sowie die Analyse der genommenen Daten waren ebenfalls Gegenstand meiner Arbeit.
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1 Introduction

Particle physics is that branch of physics that deals with elementary particles, their interactions, and their symmetries. It describes the innermost structure of matter and thus gives an insight into the evolution of the universe. An open question in the origin of the universe is why there is much more matter than antimatter. This phenomenon is known as the baryon asymmetry. If right after the Big Bang matter and antimatter were produced in the same amounts, one would expect a complete cancellation of both leaving behind a sea of photons. However, this is not the case. From astrophysical observations [Kom11] we know that the baryon asymmetry is

\[ \eta = \frac{n_B - n_{\bar{B}}}{n_\gamma} = 6.3 \times 10^{-10}, \]

(1.1)

where \( n_B \) is the baryon number, \( n_{\bar{B}} \) the antibaryon number and \( n_\gamma = n_B + n_{\bar{B}} \) the number of the relic photons. We therefore need a mechanism that produces more matter than antimatter or transforms antimatter into matter somehow. In 1967, Sakharov postulated three necessary conditions for baryogenesis, the creation of matter [Sak67]:

- Baryon number violation,
- C and CP violation,
- Thermal non-equilibrium,

where C denotes charge conjugation and CP the product of charge conjugation and parity. The Standard Model (SM) predicts only a baryon asymmetry of \( \eta \sim 10^{-18} \) [Rio99], a discrepancy of \( 10^8 \) with observations. Additional sources of CP violation in extensions of the SM might help in solving this discrepancy. As for the baryon asymmetry, CP violation is a requirement for non-zero electric dipole moments (EDM) of particles. Hence, a non-zero EDM indicates a source of CP violation.

1.1 Neutron electric dipole moment and CP violation

The existence of a nEDM is closely linked to the discrete symmetries of nature: charge conjugation (C), parity (P), and time reversal symmetry (T). A neutron in a magnetic and electric field having a non-zero EDM would violate P and T symmetry [Pos05]. The corresponding Hamiltonian is:

\[ \mathcal{H} = -\mu \frac{\vec{\sigma} \cdot \vec{B}}{|\vec{\sigma}|} - d \frac{\vec{\sigma} \cdot \vec{E}}{|\vec{\sigma}|} \]

(1.2)

where \( \mu \) and \( d \) are the magnetic and electric dipole moment of the neutron respectively, \( \vec{\sigma} \) the spin of the neutron, \( \vec{B} \) the magnetic field and \( \vec{E} \) the electric field. While the spin \( \vec{\sigma} \) and the magnetic field \( \vec{B} \) are axial vectors, the electric field \( \vec{E} \) is a polar vector. The first part of the Hamiltonian
Figure 1.1: A non-zero EDM of the neutron in a magnetic and electric field violates the parity symmetry (P) as well as the time-reversal symmetry (T). While the first term in Eqn. 1.2 is unchanged for both transformations, the second term changes sign and thereby violating both symmetries.

remains unchanged under P and T transformation, whilst the second part with the electric field changes the sign. The violation under P and T symmetry is therefore obvious. Fig. 1.1 illustrates the behaviour of the Hamiltonian under P- and T-transformation. The Hamiltonian is not an eigenfunction of the P and the T operator and violates thus P and T symmetry. The first measurement of the nEDM was proposed in 1950 by Ramsey and Purcell [Pur50] and performed in 1951. At that time there was no experimental evidence that interactions among particles violate parity. The goal of their experiment was actually to prove its conservation and the expected null-result was therefore not a big surprise. In 1956 parity violation was discovered by Wu [Wu57] in the weak interaction and Ramsey’s experiment became of interest again. The result of Ramsey’s experiment was published in 1957 [Smi57]. This was the starting point of a series of EDM-experiments which lasts up to now. Since 1950, various experiments have been performed, searching for the neutron EDM, but only upper limits have been found and published so far (see Fig. 1.2). In order to construct a symmetry that is conserved in all interactions, Landau proposed that CP symmetry should be conserved [Lan57]. This means that only the product of C and P is conserved and not each symmetry itself. The combined symmetry is therefore less constrictive. But this solution held only seven years. In 1964, CP symmetry was found to be violated in the system of the neutral kaons in the weak interaction. However, one can still construct an even weaker symmetry which is conserved, the CPT symmetry [Lue57]. So far, this symmetry was not found to be broken. Under the assumption of CPT conservation, the nEDM violates CP, knowing that it violates T symmetry.

1.1.1 CP violation in the weak interaction

The Standard Model contains a CP violating term in the weak interaction. The Cabibbo-Kobayashi-Maskawa-matrix (CKM-matrix) describes the mixing of quark flavours. The single matrix elements are transition amplitudes from one quark flavour into another one. The stan-
Figure 1.2: The upper limits of the nEDM experiments since 1951 (taken from [Pen08]): The first experiments were using neutron beams. Technical difficulties and nonnegligible systematics, amongst others the motional magnetic fields, forced to use a more appropriate method. Ultracold neutrons (UCN) in storage chambers turned out to be a promising solution. [Sha68]
The standard parametrisation of the CKM-matrix is [Nak10]

\[ V_{\text{CKM}} = \begin{pmatrix} c_{12}c_{13} & s_{12}c_{13} & s_{13}e^{-i\delta} \\ -s_{12}c_{23} - c_{12}s_{23}s_{13}e^{i\delta} & c_{12}c_{23} - s_{12}s_{23}s_{13}e^{i\delta} & s_{23}c_{13} \\ s_{12}s_{23} - c_{12}c_{23}s_{13}e^{i\delta} & -c_{12}s_{23} - s_{12}c_{23}s_{13}e^{i\delta} & c_{23}c_{13} \end{pmatrix} \] (1.3)

where \( s_{ij} = \sin(\theta_{ij}) \) and \( c_{ij} = \cos(\theta_{ij}) \). The CKM-matrix can be parametrised by 3 mixing angles \( \theta_{12}, \theta_{23}, \theta_{13} \) and a complex phase \( \delta \). The phase is responsible for CP violation. Valence quarks can also possess EDMs on their own stemming from loop diagrams of third order and higher [Cza97]. The loop diagrams allow the change of flavors and the matrix elements of CKM-matrix appear in the calculations. Therefore the quark EDMs depend on the CP violating phase \( \delta \). Finally, the EDMs of the up- and the down-quark enter the formula of the neutron EDM in the following way [Hec01]:

\[ d_n = \frac{1}{3}(4d_d - d_u) \] (1.4)

The weak interaction in the standard model predicts an neutron EDM of \( \sim 10^{-32} \) e·cm [Dar00].

1.1.2 CP violation in the strong interaction

The Standard Model contains CP violating terms also in the strong interaction, however so far CP violation in the strong interaction has never been observed. The Lagrangian Quantum Chromodynamics (QCD) also contains a CP violating term [Pos05]:

\[ \mathcal{L}_\theta = \frac{g_s}{32\pi} \tilde{G}_{\mu\nu}^a G^{a\mu\nu} \] (1.5)

where \( \tilde{G}_{\mu\nu}^a G^{a\mu\nu} \) is the tensor product of the gluon fields, \( g_s \) the interaction constant of the strong interaction and \( \bar{\theta} \) the physical value of the scaling angle \( \theta \). This term represents the interaction of gluons with the vacuum. \( \bar{\theta} \) includes all the phases of the quark mass matrix \( M_q \)

\[ \bar{\theta} = \theta + \text{Arg Det}(M_q). \] (1.6)

The angle \( \bar{\theta} \) can take values from 0 to 2\( \pi \) and regulates the CP violation in the Lagrangian term in Eqn. 1.5. Additionally, \( \theta \) is linked to the nEDM via the following equation [Pos05]:

\[ d_n \sim e \frac{\bar{\theta}m_s}{m_n^2} \sim \bar{\theta} \cdot (6 \times 10^{-17} \text{ e·cm}), \] (1.7)

where \( m_s \) is the reduced quark mass in the two-flavor QCD and \( m_n \) the neutron mass. The most recent measurement of the nEDM results in [Bak06]

\[ d_n = (0.2 \pm 1.5_{\text{stat}} \pm 0.7_{\text{syst}}) \times 10^{-26} \text{ e·cm}, \] (1.8)

which can be interpreted as an upper limit of

\[ d_n \leq 2.9 \times 10^{-26} \text{ e·cm (90\% C.L.)}. \] (1.9)

From Eqn. 1.7 and Eqn. 1.9, the following constraint on \( \bar{\theta} \) can be given:

\[ \bar{\theta} \leq 5 \times 10^{-10} \] (1.10)

The question why \( \bar{\theta} \) is so small is known as the strong CP problem. A possible solution to this problem is the Peccei-Quinn theory [Pec77] that introduces a scalar field that corresponds to a
massless scalar particle called the axion. The axion cancels the $\bar{\theta}$-term through an axion-gluon anomaly diagram. The corresponding Lagrangian is then

$$L_{\theta_{\text{axion}}} = \frac{1}{f_a} \phi_a \frac{\alpha_s}{8\pi} \tilde{G}^a_{\mu \nu} G^{a \mu \nu},$$

where $f_a$ is the decay constant and $\phi_a$ is the axion field. The axion relaxes to $\langle \phi_a \rangle = \bar{\theta} \cdot f_a$ cancelling thereby the $\bar{\theta}$-term.

The CP violation created by weak section of the Standard Model is too small to explain the baryon asymmetry. There are various extensions of the Standard Model such as Multi-Higgs models [Des94], Super Symmetric (SUSY) models [Kra07] and Left-Right Symmetric models [He88] which contain a larger amount of CP violation. These models will among other create a larger baryon asymmetry as well as larger EDMs. Predictions of these models for the nEDM are shown in Fig. 1.3. The search for the nEDM is therefore a test of extensions of the SM. Especially the next generation of nEDM experiments, aiming at a sensitivity of a few times $10^{-28} e\,\text{cm}$, are of interest, since various supersymmetric models predict nEDMs in this range [Dub11]. A zero result from these experiments would thus rule out many supersymmetric models or heavily constrain their parameter space.

### 1.2 How to measure a nEDM

The method applied in nEDM experiments exists since 1950, when it was invented by Norman F. Ramsey [Ram50]. It is therefore called Ramsey’s method of separated oscillatory fields. First, polarised neutrons are aligned to a magnetic field $\vec{B}$. I denote this direction here as the $z$-direction and the plane perpendicular to it as the $xy$-plane. An additional magnetic field, oscillating at resonance in the $xy$-plane, is gated on for a certain time which is also denoted as a spin-flip pulse. During this time, the neutron spins are flipped into the $xy$-plane. For a certain period of time, the spins precess freely around $\vec{B}$ at the corresponding Larmor frequency. Afterwards, the oscillatory field is gated on again, keeping the phase relation to the initial phase. The spins are thus flipped in the negative $z$-direction.

A crucial point is the timing of the second pulse. A 10 MHz atomic clock is used to apply the pulse exactly in the phase to the first pulse after the precession time. The method can be regarded as a clock comparison experiment with the precessing neutrons as one clock and the atomic clock as the other one. A slight shift of the neutron frequency will lead to a phase difference relative to the flip frequency. The second spin flip would then not be complete and a certain amount of neutrons would be in the spin up state, revealing the frequency shift. The determination of the precession frequency from the number of neutrons in the spin up and spin down state is described in Sec. 2.2.4.

The described procedure is known as a Ramsey cycle and is shown in Fig. 1.4. When an additional electric field $\vec{E}$ parallel to $\vec{B}$ is applied, the frequency is shifted and can be expressed via the Hamiltonian in Eqn. 1.2:

$$h \nu_{\uparrow \uparrow} = |2\mu_n B + 2d_n E|.$$  \hspace{1cm} (1.12)

For $\vec{E}$ antiparallel to $\vec{B}$, the expression changes accordingly to:

$$h \nu_{\uparrow \downarrow} = |2\mu_n B - 2d_n E|.$$  \hspace{1cm} (1.13)

From the two frequencies, the electric dipole moment can be calculated easily:

$$d_n = \frac{h(\nu_{\uparrow \uparrow} - \nu_{\uparrow \downarrow})}{4E}.$$  \hspace{1cm} (1.14)
Figure 1.3: Extensions of the Standard Model and their predictions for the nEDM. The dashed line marks the current upper limit. The plot is taken from [Pen00] and was slightly adapted. (The current upper limit is inserted.)
Figure 1.4: Ramsey’s method of separated oscillatory field: steps 1 to 4 show the motion of a spin in a magnetic field. The red curve on the right illustrates an additional oscillatory field used to flip the spin into and out of the $xy$-plane. It is only in use during the two spin flips and else is gated off. These so-called spinflip pulses are phase-coherent. All four steps together are called a Ramsey cycle. The figure is taken from [Har08].
1.3 Magnetometry in nEDM experiments

There are several experiments worldwide searching for the neutron electric dipole moment. As the experiments become more and more sensitive, the requirements concerning magnetic field quality increase. The expression for the nEDM in Eqn. 1.14 has to be adapted when the magnetic field is not the same for $\vec{B} \uparrow \uparrow \vec{E}$ as for $\vec{B} \uparrow \downarrow \vec{E}$. Here, I refer to $B_{\uparrow \uparrow}$ in the parallel case and to $B_{\uparrow \downarrow}$ otherwise. The complete formula for the nEDM is then

$$d_n = \frac{2\mu_n (B_{\uparrow \uparrow} - B_{\uparrow \downarrow}) + h(\nu_{\uparrow \uparrow} - \nu_{\uparrow \downarrow})}{4E}.$$  (1.15)

Any change in the magnetic field between the polarity change of the electric field mimics, according to Eqn. 1.15, a false EDM, as long as the magnetic field is unknown. In order to reduce or correct for these field drifts, many measures were taken, such as multi-layer shielding against external magnetic fields, active shielding, use of double chambers, and use of magnetometers [Pen08], [Khr97]. Possibly the most striking success was achieved when a cohabiting magnetometer was implemented for the very first time [Har99](see Fig. 1.5). The RAL/Sussex/ILL collaboration followed an idea of Norman F. Ramsey [Ram84], as field fluctuations could not be entirely corrected with Rb-magnetometers located next to the precession chamber. A cohabiting or co-magnetometer is a polarised spin-population of atoms, that measures the magnetic field inside the same volume and at the same time as the precessing neutrons. Candidates for a possible co-magnetometer should have the following features:

- Long spin coherence times: A useful co-magnetometer should show spin coherence times of the order of the free neutron precession, which is in the range of 100 s for today's nEDM experiments [Khr97]. $^1S_0$ diamagnetic atoms with nuclear spin $1/2$ are preferably used, since their magnetic moments are small and show thus less interaction with magnetic perturbations.

- Negligible permanent EDM: A permanent electric dipole moment of the cohabiting atoms would create a false-EDM signal. Luckily, permanent EDMs of atoms are small due to the Schiff screening [Sch63], but as experiments become more and more sensitive, EDM upper limits of the magnetometer atoms need to be below the aimed sensitivity of the nEDM experiment.

- No influence on neutron precession: Implementing a co-magnetometer should not disturb the neutron precession, e.g. with additional spin-flips or with a large neutron absorption cross-section of the cohabiting atoms.

There are only three stable isotopes that might reasonably work as a co-magnetometer: $^3$He, $^{129}$Xe, and $^{199}$Hg [Khr97].

$^3$He as a co-magnetometer

A measurement of the spin coherence time of $^3$He in our collaboration resulted in 1 h [Kra12]. Additionally, the intrinsic EDM can be estimated to $d(^3\text{He}) \sim (3 \times 10^{-5}) \cdot d_n$ [Dzu07]. However, a resonance frequency close to that one of the neutron as well as a large neutron absorption cross-section of about 5333 barn (for a velocity of 2200 m/s) [NIS11] might cause some problems. Nevertheless, $^3$He will be used as a co-magnetometer in the nEDM experiment at SNS, Oak Ridge [SNS11].
The plot shows a time series of the raw neutron frequency and the same timeseries corrected with the Hg frequency over a period of a day (run 2999 and 3000, 2nd of May 2008). The magnetic field drifts are reduced from 50 − 100 pT (raw data) to 3 pT (normalised data).

$^{129}$Xe as a co-magnetometer

For $^{129}$Xe, spin coherence times of about 1000 s can be obtained [Pav03]. However, the most recent result of the EDM of $^{129}$Xe was $d(^{129}$Xe) = (0.49 ± 3.3_{stat} ± 0.1_{syst}) × 10^{-27} e cm [Ros01]. Using it as a co-magnetometer is only possible up to a sensitivity of $\sim 10^{-26} e cm$, unless its EDM is not further constrained.

$^{199}$Hg as a co-magnetometer

$^{199}$Hg is the only co-magnetometer in nEDM experiments that has been used successfully [Gre98], [Har99]. It frequently showed spin coherence times between 100 s and 200 s. The last measurement of the HgEDM showed a value of $d(^{199}$Hg) = (0.49 ± 1.29_{stat} ± 0.76_{syst}) × 10^{-29} e cm, which resulted in an upper limit of $|d(^{199}$Hg)| ≤ 3.1 × 10^{-29} e cm (95% C.L.) [Gri09]. With the measurement cells of the HgEDM, spin coherence times of 240 s were reached as best values [Rom03]. However, the cells were filled with 200 − 300 mbar of N$_2$ (90 %) and CO (10 %) as a buffer gas. Concerning the neutron precession, the disturbance is very small, since the resonance frequency for $^{199}$Hg ($\sim 8$ Hz at 1 $\mu$T) is far enough away from that of the neutrons ($\sim 30$ Hz at 1 $\mu$T). It can be further reduced by a clever choice of the pulse envelope.

From the present point of view, two co-magnetometer systems seem to be appropriate for a neutron EDM experiment: $^3$He and $^{199}$Hg. The advantage of $^3$He compared to $^{199}$Hg is its large spin coherence time. But as long as the duration of the free precession does not exceed 200 s, an implementation of $^{199}$Hg is favourable, since the technical realisation (optical pumping without discharge and direct optical readout with a photomultiplier) is easier to realise.

However, $^3$He is foreseen as a co-magnetometer in a cryogenic nEDM-experiment at ORNL [Pen08] which uses superfluid $^4$He for production and storage of UCN. A small concentration
\( \sim 10^{-10} \) of polarised \(^3\)He is injected into the superfluid \(^4\)He where its spin precession is detected by a SQUID system.

A disadvantage of adding a second spin-species to the neutrons for monitoring the magnetic field is that additional systematic effects occur (see chapter 5).
2 The nEDM experiment at PSI

The most recent upper limit of the nEDM of $2.9 \times 10^{-26} e\text{cm}$ (90% C.L.) was published in 2006 by the RAL-Sussex-ILL collaboration [Bak06]. The experiment located at ILL used the storage of ultracold neutrons (UCN, see Sec. 2.1) at room temperature. The storage method is used since the beginning of the eighties when it superseded experiments using a beam of cold neutrons [Pen08]. Uncontrollable systematic effects in the beam experiments were dominating over the statistical uncertainties and storage experiments become more favourable. Another advantage of storage experiments compared to beam experiments is that much longer precession times can be obtained which compensates well for the much lower neutron density (a few 100 s for the storage method compared to a few milliseconds for the beam method). The precession time $T$ enters the sensitivity of the experiment as follows:

$$\sigma(d_n) = \frac{\hbar}{2\alpha EN^{1/2}} \sqrt{N},$$

(2.1)

where $\alpha$ is the visibility of the neutrons (neutron polarisation after the Ramsey cycle), $E$ the electric field, and $N$ the number of neutrons (a derivation of the formula is given in Sec. 2.2.4).

However, the sensitivity of the RAL-Sussex-ILL room temperature experiment was finally limited by the UCN counting statistics and a further improvement would mean increasing the number of UCN. The RAL-Sussex-ILL collaboration decided to pursue a new technique with a completely new apparatus, the cryo EDM [Cry11]. Our collaboration plans to proceed in three phases, which includes an inital experiment with the former apparatus borrowed from RAL-Sussex-ILL.

- Phase I (2006 - 2009): The goal of the first phase was to take over the apparatus and to operate it at ILL (Grenoble, France) in order to get some know-how in the operation. Additionally, some parts were rebuilt or added. The most prominent of them are a new storage chamber for the UCN [Kuz09] and Cs-magnetometers developed by the university of Fribourg [Gro06] for detecting magnetic inhomogeneities. Finally, our collaboration took a sample of EDM-data for the very first time in 2008 [Kne09].

- Phase II (2009 - 2013): In the second phase, the apparatus was set up at the new UCN source at Paul Scherrer Institute (PSI, Villigen, Switzerland). The expected UCN density at PSI is $1000 \text{UCN/cm}^3$ [Ang09] compared to $\sim 50 \text{UCN/cm}^3$ [Ste89] at ILL. Major changes compared to Phase I are the use of a superconducting magnet for the polarisation of the UCN (see Sec. 2.2.1), Cs magnetometers (see Sec. 2.4.1) as well as a new HV system and new designed electrodes [Zen10]. The experiment aims at a sensitivity of $5 \times 10^{-27} e\text{cm}$. Beside the measurements with the Sussex-apparatus, the design of a new apparatus is in progress.

- Phase III (2013 - 2020): In the third phase, a measurement of the nEDM with a completely new apparatus is planned. This experiment aims at a sensitivity of $5 \times 10^{-28} e\text{cm}$. The
Table 2.1: Materials and their Fermi potentials: Many of them are used as storage materials in UCN physics. (Values taken from [May98] and [Goe12])

<table>
<thead>
<tr>
<th>Material</th>
<th>$V_F$ (neV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al</td>
<td>54</td>
</tr>
<tr>
<td>$^{58}$Ni</td>
<td>335</td>
</tr>
<tr>
<td>Fe</td>
<td>210</td>
</tr>
<tr>
<td>Cu</td>
<td>168</td>
</tr>
<tr>
<td>PTFE (Teflon, PF$_2$)</td>
<td>123</td>
</tr>
<tr>
<td>dPS ($C_8D_8$)</td>
<td>170</td>
</tr>
<tr>
<td>Stainless Steel (type T316)</td>
<td>185</td>
</tr>
<tr>
<td>BeO</td>
<td>261</td>
</tr>
<tr>
<td>C (True diamond)</td>
<td>305</td>
</tr>
<tr>
<td>Deuterated CVD diamond or DLC</td>
<td>220</td>
</tr>
<tr>
<td>Perfluoro polyether (Fomblin oil)</td>
<td>107</td>
</tr>
<tr>
<td>H$_2$O</td>
<td>-14.7</td>
</tr>
<tr>
<td>Ti</td>
<td>-48</td>
</tr>
</tbody>
</table>

main improvements of the new apparatus will be a double-chamber, a five-layer magnetic shield, and a large area gradiometers.

2.1 Ultracold neutrons - the new source at PSI

Fermi realised that slow neutrons which are scattered by an ensemble of nuclei are refracted like light and that these nuclei seem to have a potential that attracts or repulses neutrons. Neutrons incident on a surface at a glancing angle $\theta$ with an energy $E$ will be totally reflected when the following condition is fulfilled [Fer46]:

$$\sin \theta \leq \sin \theta_c = \left( \frac{V_F}{E} \right)^{1/2},$$

(2.2)

where $\theta_c$ is the critical angle and $V_F$ is the Fermi potential, an effective potential produced by the nuclei of the material. The condition for neutrons to be totally reflected under all angles of incidence is

$$E \leq V_F.$$  

(2.3)

Zeldovich pointed out in 1959 that neutrons with energies below the Fermi potential could be stored in suitable containers [Zel59]. Typical materials used for the storage of neutrons are given in Tab. 2.1, values for the Fermi potential are on the order of 100 neV. Neutrons with energies less than 300 neV, which corresponds to a temperature of about 3 mK, are called ultracold neutrons (UCN). In this case, the neutron no longer interacts with a single nucleus but a whole ensemble of nuclei. Some years after Zeldovich’s idea, Shapiro was the first to experiment with UCN. He proposed to use UCN for future measurements of the nEDM [Sha68].

At PSI in Villigen, Switzerland, a new spallation-based UCN source has been built [Ang09]. The most important components of the source are shown in Fig. 2.1. The core of the source is a target that consists of 760 tubes of zircaloy$^1$ filled with lead. Protons with an energy of 590 MeV hit the target and smash the lead nuclei. In this process called spallation, about 10 free neutrons

$^1$Zircaloy is an alloy with more than 90% zirconium. The small neutron absorption cross-section makes it transparent for neutrons. Therefore, it is an ideal shell-material for lead tubes in a spallation source.
per incident proton are produced [Kir10]. The proton beam with an intensity of more than 2 mA is produced in the ring cyclotron at PSI and is guided to the target. It is used in a pulsed mode with a duty cycle of 1%. During the pulse, the beam is deflected by a strong kicker magnet. In operation, a pulse between 4 s and 8 s with a corresponding waiting time can be used.

Once the neutrons are produced, they are moderated to thermal energies by heavy water (D_2O). The target is located in the centre of a moderator vessel filled with heavy water (D_2O), which is also used for cooling. A solid D_2 crystal in a second vessel above the target is held at a temperature of about 5 K by supercritical helium. In this crystal the thermal neutrons are further moderated to cold neutrons. The cold neutrons are then downscattered to UCN energies by exciting phonons in the crystal. The reverse process is highly suppressed because of the low temperature of the material. The UCN which leave the D_2 in the vertical direction are further slowed down by gravity and can be stored in the UCN storage volume of \( \sim 1.6 \text{ m}^3 \) [Ang09]. The UCN are guided via the neutron guides to the experimental facilities.

## 2.2 UCN in the nEDM experiment

### 2.2.1 Polarisation of UCN

Before the UCN can be used in the nEDM experiment, they have to be polarised. The polarisation is needed for Ramsey’s method of separated oscillatory fields (see Sec. 1.2) and is done by a superconducting magnet on the horizontal neutron guide. The magnet produces a 5 T magnetic field in the direction of the guide (see Fig. 2.2, upper part). The corresponding potential \( V = -\vec{\mu}_n \vec{B} \) is 300 neV. This potential barrier blocks one spin state whilst the other spin state passes through (see Fig. 2.2, lower part). The polarisation afterwards is almost 100% whilst a polarisation with an iron foil, as it was done at ILL, only amounts to 90% [Roc09]. Guiding fields installed after the magnet guarantee that the ensemble of UCN keeps the polarisation until they enter the experiment. The necessary condition for spin transport is adiabaticity:

\[
\frac{|\partial \vec{B}/\partial t|}{|\vec{B}|} \ll \gamma_n |\vec{B}|, \tag{2.4}
\]

where \( \partial \vec{B}/\partial t \) is the change of the magnetic field in time in the rest frame of the UCN, \( \vec{B} \) the magnetic field and \( \gamma_n \) the gyromagnetic factor of the neutron. It means that the field changes are slow enough, that the spins of the UCN can follow the field lines.

### 2.2.2 Storage of UCN in the nEDM apparatus

In the following, a description of the apparatus is given in some detail (see Fig. 2.3). A switch connects the guide from the source with the vertical guide to the experiment and the UCN enter the experiment from below. They are stored in the precession chamber, where the Ramsey cycles are preformed, as described in Sec. 1.2. The chamber consists of an upper and a lower electrode, which are separated by an insulator ring. The upper electrode is charged via a high voltage (HV) feedthrough while the lower electrode is at ground. Thus a strong electric field in the range of 12 kV/cm is produced inside the chamber. During the Ramsey cycle, the chamber is closed by the UCN-shutter located below the lower electrode. The chamber itself sits in a vacuum tank that can be evacuated to a pressure of some \( 10^{-6} \text{ mbar} \). During the measurement, the chamber is filled with \( \approx 5 \times 10^{-3} \text{ mbar} \) of He/Ne to increase HV-stability. On the outside wall of the vacuum tank several coils are mounted. The main coil is for the holding field \( B_0 \), that can be parallel or anti-parallel to the electric field. Three split coil pairs are used for spin-flipping the UCN and the \(^{199}\text{Hg}\) atoms (the \(^{199}\text{Hg}\)-magnetometer is described in Sec. 2.4.2.), and 33 trim coils to
Figure 2.1: UCN source at Paul Scherrer Institute: The proton beam is guided onto the lead target and produces free neutrons in a spallation process. The neutrons are moderated in the heavy water and finally downscattered in the D\textsubscript{2}-crystal to UCN. Afterwards, they are stored in the storage volume for the next Ramsey cycle. Through the neutron guides, they are guided to the nEDM experiment. Figure taken from [Kir10].
Figure 2.2: The superconducting magnet produces a 5 T field in the direction of the neutron guide (upper part) thus producing a potential barrier which allows only UCN with a spin parallel to the magnetic field to pass (lower part).
reduce magnetic inhomogeneities (see Sec. 2.3.2). The vacuum tank is surrounded by a four-layer Mu-metal shield in order to shield the inside from external magnetic fields. The switch below the vacuum tank not only deflects neutrons up the vertical guide during filling but also connects the precession chamber with the UCN detector while counting UCN, or can be set into a position to deflect neutrons coming from the source directly into the detector, depending on its position. Below the switch are a thin magnetised foil and a spin flipper for spin analysis and the UCN detector that counts the UCN coming from the experiment.

In order to store UCN in the chamber, the electrodes are coated with diamond-like carbon (DLC) and the insulator ring is coated with deuterated polystyrene (dPS). Both materials have large Fermi potentials (see Tab. 2.1) and thus are suitable for storing UCN.

There are however several loss mechanisms for UCN in a chamber [Gol91]:

- \(\beta\)-decay: A natural limit on the storage of UCN is set by the lifetime of the free neutron. A value of \(881.5 \pm 1.5\) s is stated by the Particle Data Group [Nak10]. However, measured storage times are much smaller due to additional terms.

- Upscattering: First there is the upscattering of the UCN. Inelastic scattering on the wall material or on residual gas transfers energy to the neutron, whose energy is then no longer
in the UCN-regime. It immediately leaves the storage volume as it is no longer reflected on the vessel walls.

- Absorption: A next point are absorptions of UCN by the walls of the containment, by residual gas or by contaminants. A clean surface is therefore essential for long storage times.

- Slits: Finally, UCN can leak out of the container through slits.

For each of these mechanisms a characteristic loss rate $\Gamma$ can be defined. The inverse sum gives the storage time:

$$\tau_s = (\Gamma_\beta + \Gamma_{up} + \Gamma_{abs} + \Gamma_{slit})^{-1}. \quad (2.5)$$

The loss of UCN is energy-dependent (except the one for $\beta$-decay). UCN with higher energy are lost more often than those with lower energies. Very often a function with two exponentials is chosen to approximate these energy dependent losses:

$$N(t) = N_1 \exp(-t/\tau_{s,fast}) + N_2 \exp(-t/\tau_{s,slow}), \quad (2.6)$$

where $N(t)$ is the number of UCN after a time $t$, $\tau_{s,fast}$ is the decay time for UCN with higher energy and $\tau_{s,slow}$ is the decay time for UCN with lower energy. Longer storage times are of importance, since it allows longer Ramsey cycles, which makes the experiment more sensitive. A recent measurement at PSI resulted in $\tau_{s,fast} = 56 s \pm 14 s$ and $\tau_{s,slow} = 182 s \pm 33 s$ [Col12] compared to $\tau_{s,fast} \approx 40 s$ and $\tau_{s,slow} \approx 190 s$ at ILL [Kne09].

Not only a long storage time is of importance, but also a high visibility $\alpha$, since it also affects the sensitivity of the experiment. The visibility is defined as follows:

$$\alpha(t) = \frac{N_\uparrow(t) - N_\downarrow(t)}{N_\uparrow(t) + N_\downarrow(t)}, \quad (2.7)$$

where $N_\uparrow(t)$ is the number of UCN with spin up after a time $t$ and $N_\downarrow(t)$ is the number of UCN with spin down after $t$. The visibility $\alpha$ decays exponentially with time, since reflections at the walls and magnetic inhomogeneities cause the ensemble of UCN to depolarise. The characteristic time, in which the polarised ensemble decays, is called relaxation time $T_r$. One distinguishes between the longitudinal relaxation time $T_1$, which describes the decay of the polarisation of an ensemble aligned along a magnetic field, and the transverse relaxation time $T_2$, which describes the decay of the polarisation of a precessing ensemble perpendicular to a magnetic field. In the nEDM-experiment, the latter case is present and we use $T_2$. The formula for $\alpha$ is then

$$\alpha(t) = \alpha_0 \exp(-t/T_2), \quad (2.8)$$

where $\alpha_0$ is the visibility at the beginning of the Ramsey cycle. A first measurement at PSI resulted in $\alpha_0 = 0.790 \pm 0.004$ and $T_2 = 556 s \pm 15 s$ compared to $\alpha_0 \approx 0.8$ and $T_2 \approx 400 s$ at ILL [Kne09].

### 2.2.3 Spin analysis and detection of UCN

After the second pulse of the Ramsey cycle, the spin states of the neutrons are analysed as shown in Fig. 2.4. For EDM measurements, it is important to know the number of UCN for both spin states (see Sec. 2.2.4). When the UCN-shutter opens for emptying the precession chamber, the switch directly connects the chamber to the detector. For spin analysis a spin flipper and a 25 $\mu$m thick aluminium foil with a thin layer of iron is used. The iron layer has a thickness of 400 nm and is magnetised by an iron yoke and permanent magnets around the neutron guide. The field
inside the iron foil is about 2 T, which produces a potential large enough, that only UCN with spin up are able to pass. UCN with spin down are blocked by the magnetic barrier. First, the UCN with spin up are counted and the spin flipper just above the foil is off. Afterwards, it is switched on allowing the other spin state to pass. The neutrons fall into the detector, which is located roughly 1.6 m below the precession chamber.

The detector was developed at LPC Caen [Rog09] and consists of nine separate light guides with a thin layer doped with $^6$Li in the upper part of the guides, the scintillator. The subdivision is made to allow high count rates, since each channel detects only a fraction of the total rate. Each channel is capable of handling count rates up to a few $10^5$ UCN/s. Typical measured values are $10^5$ UCN/s for each channel. The detector was developed at LPC Caen [Rog09] and consists of nine separate light guides with a thin layer doped with $^6$Li in the upper part of the guides, the scintillator. The subdivision is made to allow high count rates, since each channel detects only a fraction of the total rate. Each channel is capable of handling count rates up to a few $10^5$ UCN/s. Typical measured values are $10^5$ UCN/s for each channel. The neutrons undergo the following reaction [Kno00]:

$$n + ^6\text{Li} \rightarrow ^3\text{H} + ^4\text{He} + 4.78 \text{ MeV (kinetic energy)}$$

The produced $^3$H and $^4$He-ions excite cerium atoms, which emit photons in the violet spectral range. The light is guided to the photomultipliers. A dedicated signal processing system finally discriminates all neutron signals and sends the total number of counts per precession cycle to the main computer.

### 2.2.4 Extraction of the precession frequency

So far, only the counts of UCN for different spin states $N_\uparrow$ and $N_\downarrow$ are known after one cycle. When performing a Ramsey cycle and changing the frequency of the applied pulses from one cycle to the next, the counts of UCN with spin up and spin down change according to an interference pattern (see Fig. 2.5), here referred to as the Ramsey pattern. At resonance frequency, there should be in the ideal case no UCN with spin up. Nevertheless, it turns out that this is not the case, since the polarisation of the neutrons is not perfect. However, being close to the resonance frequency, one can find an analytical approximation for the neutron counts as a function of the applied frequency:

$$N(\nu) = \bar{N}(1 \mp \alpha \cos\left(\frac{\pi\nu - \nu_0}{\Delta\nu}\right)),$$

where $\bar{N} = (N_\uparrow + N_\downarrow)/2$, $\alpha$ is the visibility, $\nu$ is the applied rf frequency of the spin flip pulse, $\nu_0$ is the resonance frequency and $\Delta\nu$ is the width of the resonance. The negative sign refers to UCN with spin up and the positive one to spin down. The width of the resonance is

$$\Delta\nu = 1/(2(T + 2\tau\text{rf}/\pi)).$$

where $\tau\text{rf}$ is the duration of the spin flip pulses and $T$ the free precession time. There are three free parameters in Eqn. 2.9: $\bar{N}$, $\alpha$ and $\nu_0$, from which the last one is used to calculate the EDM. To determine them, one needs at least three independent measurements, from which one knows the UCN counts and the applied rf frequency of the spin flip pulse. Then, a fit of the UCN counts in the interference pattern using Eqn. 2.9 is performed over all cycles of a run. The RAL-Sussex-ILL collaboration chose the applied rf frequency alternately at four points:

$$\nu_0 + \Delta\nu/2 \pm \delta\nu,$$

and

$$\nu_0 - \Delta\nu/2 \pm \delta\nu,$$

where $\delta\nu$ is arbitrary but must hold $\delta\nu \ll \Delta\nu$. These points are chosen since they are the steepest and thus most sensitive points in the Ramsey pattern [May98]. In the online analysis, four successive Ramsey cycles were used for a fit to determine the free parameters.
Figure 2.4: Spin analyser and UCN detector of the nEDM apparatus. Below the switch, a neutron guide leads to the detector. In between is a magnetised iron foil, which only allows UCN with spin up to pass. A spin flipper makes a detection of the UCN with spin down possible.
Figure 2.5: The number of UCN counts with spin up (triangle up) and spin down (triangle down) depends on the applied rf frequency of the spin flip pulse. The resonance frequency is determined by fitting the interference pattern (=Ramsey pattern) to a cosine-curve.

Of great interest is the estimation of the intrinsic sensitivity of the method. This is described by the uncertainty of the neutron frequency estimation. For a purely statistical error of the neutron counts, \( \sigma_N = \sqrt{N} \) holds. This translates into an error of the frequency as follows:

\[
\sigma_\nu = \frac{1}{\left| \frac{dN}{d\nu} \right|_{\text{max}}} \sigma_N, \quad (2.13)
\]

where \( \left| \frac{dN}{d\nu} \right|_{\text{max}} \) is the slope of the Ramsey pattern at the steepest point. Calculating this from Eqn. 2.9 results in

\[
\left| \frac{dN}{d\nu} \right|_{\text{max}} = \frac{N\pi\alpha}{\Delta \nu} \approx 2\pi\alpha\bar{N}T, \quad (2.14)
\]

where the approximation \( \Delta \nu \approx 1/(2T) \) has been used. Substituting Eqn. 2.14 in Eqn. 2.13 finally gives us

\[
\sigma_\nu = \frac{1}{2\pi\alpha T \sqrt{N}}, \quad (2.15)
\]

Now, this can be translated to the sensitivity of the nEDM. This is linked to its corresponding frequency difference as follows:

\[
d_n = -\frac{\hbar(\nu_{\uparrow\uparrow} - \nu_{\uparrow\downarrow})}{4E}, \quad (2.16)
\]

where \( d_n \) is the nEDM, \( \hbar \) is the Planck constant, and \( E \) is the electric field applied. Since \( \nu_{\uparrow\uparrow} \) and \( \nu_{\uparrow\downarrow} \) stem from two different measurements, their errors add linearly according to the Gaussian error propagation:

\[
\sigma(d_n) = \frac{\hbar \sigma_{\nu_{\uparrow\uparrow}}}{4E} + \frac{\hbar \sigma_{\nu_{\uparrow\downarrow}}}{4E}. \quad (2.17)
\]

Since \( \sigma_{\nu_{\uparrow\uparrow}} = \sigma_{\nu_{\uparrow\downarrow}} \), it simplifies to

\[
\sigma(d_n) = \frac{\hbar \sigma_\nu}{2E} = \frac{\hbar}{2\alpha ET \sqrt{N}}. \quad (2.18)
\]
This formula shows the purely statistical sensitivity of the experiment. In order to reduce it, the performance parameters $\alpha, E, T,$ and $\bar{N}$ have to be increased. The experiment at the Institut Laue Langevin (ILL) in Grenoble, France, was restricted by the limited UCN density, which in the precession chamber was 1-2 UCN/cm$^3$ [Roc09],[Bak06]. A conservative estimate from a Monte Carlo simulation gives a number of $N = 350'000$ UCN in the experiment at PSI [Zsi12], which is a factor of 25 more than at ILL [Har99]. Reasonable values for the other parameters are $\alpha = 0.75, E = 12$ kV/cm and $T = 150$ s. This results in a purely statistical sensitivity of $4 \times 10^{-25} e\text{cm}$ per cycle. Assuming 400 nights of measurements, this allows us to measure the nEDM with a sensitivity of $4 \times 10^{-27} e\text{cm}$. However, it is necessary to have the systematics under control to a comparable level.

## 2.3 Magnetic environment

As already mentioned in Sec.1.3, drifts in the magnetic field have an influence on the nEDM, since a change of the field could mask an EDM-signal or even create a false one. I will briefly recall this issue here. The contribution of the magnetic field drifts to the nEDM enters as follows:

$$d_n = \frac{2\mu_n(B_{\uparrow\uparrow} - B_{\uparrow\downarrow}) + h(\nu_{\uparrow\uparrow} - \nu_{\uparrow\downarrow})}{4E}. \quad (2.19)$$

Aiming at a sensitivity of $4 \times 10^{-25} e\text{cm}$ with an electric field of $1.2 \times 10^4 \text{V/cm}$, a condition can be obtained for the drift of the magnetic field:

$$B_{\uparrow\uparrow} - B_{\uparrow\downarrow} \ll \frac{h\Delta\nu}{2\mu_n} = \frac{4Ed_n}{2\mu_n}. \quad (2.20)$$

Taking $\mu_n = -1.91 \times \mu_N$ with $\mu_N = \frac{eh}{2\mu_0c}$, an upper limit on the tolerable magnetic field fluctuations of 190 fT can be stated between two estimates of $\nu_{\uparrow\uparrow}$ and $\nu_{\uparrow\downarrow}$. Either it is possible to keep them much smaller or a reference system monitors the fluctuations, which can be corrected for. A second requirement of the magnetic field is the homogeneity. Inhomogeneous fields result in low $T_2$-times and thus in a low $\alpha$ (see Sec. 2.2.2), which makes the measurement less precise. Additionally, inhomogeneities are a source of systematic errors (see chapter 5). Therefore, a lot of effort is put into the control and the monitoring of magnetic fields.

### 2.3.1 Reduction of external magnetic fields

To allow measurements in a highly stable and homogeneous field, the influence of external fields have to be reduced as much as possible.

**Mu-metal shield**

The vacuum tank is surrounded by four cylindrical layers of Mu-metal to shield the inside of the tank from external fields. Mu-metal is an nickel-iron alloy (80%/20%) and shows a permeability of 60'000 to 250'000 [Vac12]. The shielding factor is a measure of how much the magnetic field decreases inside the shield compared to outside. The shielding factors have been measured at PSI, where external coils were used to produce an external magnetic field of known size [Col10]. The field inside was measured with a fluxgate magnetometer. Values from this measurement and from Kuzniaks calculation [Kuz09] using Dubbers formula [Dub86] are given in Tab. 2.2.
Table 2.2: The shielding factors have been measured at ILL as well as calculated. The coordinate system is introduced in Fig. 2.6. The large difference between the measured and calculated value for the comes from holes in the Mu-metal shield. There are holes in the Mu-metal at the positions of the $^{199}$Hg-magnetometer system, the HV system, the neutron guide, and the vacuum tube. The calculated values are taken from [Kuz09], the measured values are from [Col10]

**Surrounding field compensation - SFC**

In addition to the Mu-metal shield, an external coil system was added in order to compensate for external fields and field gradients as well as correct for field drifts. The SFC consists of three large rectangular coil pairs that are located around the experiment, one coil pair for each direction of the coordinate system (see Fig. 2.6). The coils have a length of about 8 m and a width of about 6 m. The centres of the three coil pairs do not exactly match the centre of the experiment. The spatial offsets are given in Tab. 2.3.

<table>
<thead>
<tr>
<th>Coil pairs</th>
<th>x (cm)</th>
<th>y (cm)</th>
<th>z (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>X</td>
<td>-23</td>
<td>0</td>
<td>-7</td>
</tr>
<tr>
<td>Y</td>
<td>11</td>
<td>3</td>
<td>-19</td>
</tr>
<tr>
<td>Z</td>
<td>11</td>
<td>1</td>
<td>-16</td>
</tr>
</tbody>
</table>

Table 2.3: The SFC coil pairs show some small offsets from the centre of the experiment. The values above are taken from [Col10]

There are two modes in which the SFC can be run [Fra10]. In the passive mode there is a constant current flowing through the coils compensating for the earth’s field. Each coil is driven by its own power supply. This mode does not take drifts or other changes into account, e.g. a truck passing by on the road 10 m away. In the active mode, a feedback system with ten fluxgate magnetometers controls the currents for each coil. The fluxgate magnetometers are mounted at several places close to the experiment for an efficient regulation of the current.

### 2.3.2 Creation of magnetic fields inside the chamber

The Mu-metal shield and the SFC are elements to prevent external fields from disturbing the configuration of magnetic fields inside the precession chamber. For this reason, the creation of magnetic fields used for the measurement must take place within the Mu-metal shield. Different coils, wound on the outside wall of the vacuum tank, are used to generate these fields. The following list gives an overview of the different coils present.

**$B_0$-field**

For the spin precession in the chamber, a holding field called $B_0$ is needed. This is produced by a so-called $\cos(\theta)$-coil, that consists of about 50 frames with equidistant space. It produces a field of $\sim 1 \mu$T in the vertical direction.
Figure 2.6: The surrounding field compensation system (SFC) consists of three large coil pairs (one for each axis), which are installed around the experiment for reducing and stabilising external magnetic fields.
**RF coils**

For the Ramsey cycle, oscillating magnetic fields are used for the spin flips of the UCN and the $^{199}$Hg atoms. For this reason, rf coils are installed on the vacuum tank. One split coil pair produces a linear oscillating field of about 30 Hz for the spin flip of the UCN. Additionally, a second split coil pair and a saddle coil produce a circular rotating field of about 8 Hz for the spin flip of the $^{199}$Hg atoms. Although a linear oscillating field for the mercury would do, it affects the initial position of the spins of the UCN a little bit, which, at this time, are not yet flipped. The spins of the UCN would be tilted relative to the vertical direction, leading to a false effect in the EDM measurement.

**Trim coils**

An inhomogeneous magnetic field reduces the $T_2$-time and thus make the measurement of the precession frequency less sensitive. It is as well a source of systematic errors. In order to remove inhomogeneities, a system of 33 trim coils are installed to homogenise the field as much as possible. Most of the coils are saddle coils and solenoids attached on the vacuum tank. Large inhomogeneities occur at places where holes are left in the Mu-metal shield. In these holes, different inserts like the UCN guide, the Hg system, and the HV feedthrough are connected to the vacuum tank. At these places, “chimney” coils are wound around each insert to compensate for the inhomogeneity. With the trim coils, gradients can be set to investigate the performance under large gradients. In chapter 5, large vertical gradients are applied to measure the geometric phase effect of $^{199}$Hg.

### 2.4 Monitoring magnetic fields

In addition to the creation of homogeneous and stable magnetic fields, we are currently using two types of magnetometers to monitor magnetic fields inside the experiment.

#### 2.4.1 The Cs magnetometers

For monitoring magnetic fields in the vacuum tank, we use caesium magnetometers which can be installed on the top and below the bottom electrode on special plates. They allow a measurement of the magnitude of magnetic fields at a certain position as well as field gradients between two different places. They have been developed by the Fribourg Atomic Physics group and show a proven sensitivity of $14 \text{ fT}/\sqrt{\text{Hz}}$ at a bandwidth of 1 kHz. For a detailed discussion see [Gro06], here just a short summary is presented.

They consist of glass bulbs of 2 cm in diameter filled with $^{133}$Cs vapour. The glass bulb is placed in a cylindrical aluminium housing with a height of 8 cm and a diameter of 8 cm for electromagnetic shielding. A circularly polarised light beam from a tunable laser traverses the vapour of caesium atoms. The caesium has a nuclear spin of 7/2 and an electronic spin of 1/2. Therefore the total spin of a caesium atom is $F = 3$ or $F = 4$. The laser beam is tuned to the $(6S_{1/2}, F = 4 \rightarrow 6P_{1/2}, F = 3)$ hyperfine transition of the D1 line at 894 nm and locked for stable operation. An overview of the energy levels of $^{133}$Cs is given in Fig. 2.7.

The optical pumping produces a macroscopic net magnetisation along the light beam, which is tilted with respect to the $B_0$ field at an angle of $45^\circ$. The light serves not only for optical pumping but also as probe beam. The magnetisation precesses around the magnetic field $B_0$ on a cone with the Larmor frequency $\omega_L = \gamma B_0$ where $\gamma = 3.4986 \text{ Hz/nT}$ is the gyromagnetic ratio of caesium [Van89]. The precession is driven at resonance by a transverse rf field, which periodically changes the projection of the magnetisation vector on the direction of the light beam.
Figure 2.7: For optical pumping of $^{133}$Cs, the transition $(6S_{1/2}), F = 4 \rightarrow (6P_{1/2}), F = 3$ of the hyperfine structure is used. In a magnetic field, the levels of the hyperfine structure split further into Zeeman sublevels. Figure is taken from [Gro05].

This is observed by a change of the absorption on the photodiode. Finally the phase difference between the rf field and the signal is measured and the frequency and thus the magnetic field is determined. A sketch of a caesium magnetometer and its working principle is shown in Fig. 2.8.

Recent investigations on the reading of the magnetic field with Cs magnetometers showed, that the measured value depends on the orientation of the magnetic field [Wei10]. If $B_0$ is perfectly aligned to the $z$-direction, the correct field is measured. An arbitrary field, that is tilted towards the $z$-direction, produces a shift in the resonance frequency of the rf field while leaving the phase untouched and thus a slightly changed value is measured. This effect is called the projection phase error, which results in the reading of a wrong magnetic field. In order to remove this effect, Cs magnetometers have been developed with the rf field parallel to the light beam. Due to the cylindrical symmetry of this magnetometer configuration, the effect is no longer present.

2.4.2 The $^{199}$Hg co-magnetometer

As already mentioned in Sec. 1.3 and Sec. 2.2.2, the experiment contains a $^{199}$Hg cohabiting magnetometer [Gre98]. It is based on free spin precession of polarised $^{199}$Hg atoms. The measurement of the field is done in the same volume as the measurement of the neutron EDM and is therefore called cohabiting. The smallness of the magnetic moment of the atom compared to other diamagnetic atoms as well as the absence of a quadrupole moment results in long spin relaxation times (100s and longer). Even more, the simplicity of optical pumping (no spin exchange or discharge of the vapour is needed) and the easy handling at room temperature makes it highly favourable as a magnetometer.

The setup of the $^{199}$Hg magnetometer is shown in Fig. 2.9. Mercury vapour is produced in the Hg source, where HgO is heated up to 180 °C. At this temperature HgO dissociates to metallic Hg and atomic oxygen which then forms molecular oxygen O$_2$. The Hg vapour is guided through a tube to the polarisation chamber which is mounted below the precession chamber. There, the mercury is polarised by light ($\lambda = 254$ nm) coming from a microwave-driven discharge lamp filled with $^{204}$Hg. The light is linearly polarised with a polariser cube and afterwards turned to circularly polarised light by a quarter waveplate. The polarisation chamber consists of a glass tube with 29 cm length and 7 cm diameter. Originally, the walls were coated with Fomblin oil,
Figure 2.8: Sketch of a caesium magnetometer and its working principle. The dashed box contains the measuring unit with the Cs cell, the polarised light beam and the rf field. The feedback loop with the lock-in amplifier, the PID control for the rf coils and the frequency counter is separated from the Cs cell. Figure taken from [Gro06]. For the new generation of Cs magnetometers, the rf field is parallel to the light beam.

A fluorinated vacuum oil is used to prevent the $^{199}$Hg atoms from depolarising on the wall. The process of optical pumping is described in chapter 3 in more detail. During each measurement cycle, mercury accumulates in the polarisation chamber since there is an ongoing flux from the source to the chamber. At the beginning of each cycle, the polarised mercury that has been accumulated during the previous cycle is released into the precession chamber. In the precession chamber, the mercury is spin-flipped into the $xy$-plane by an rf-pulse. Afterwards, it precesses freely around the $B_0$-field at about 7.7 Hz. A second $^{204}$Hg lamp, placed outside the shield at the centre of the vacuum tank, shines light through the precession chamber 4 cm above the centre. The light gets circularly polarised when traversing the polarisation optics located on the outside of the vacuum tank. The light intensity is measured by a photomultiplier located on the opposite side of the shield. Some of the light is absorbed by the precessing mercury atoms. Since the absorption of light depends on the orientation of the spin, a modulation of the brightness is visible due to the precession. The precession frequency is obtained from the oscillating signal of the photomultiplier.

The light intensity (DC-level) is measured three times during a measuring cycle. I refer to as $I_1$, $I_2$ and $I_3$. $I_1$ is measured before the mercury is filled into the precession chamber, $I_2$ is measured with the mercury in the chamber just after the filling and finally $I_3$ is measured before the end of the precession time. A certain amount of light is absorbed by the mercury atoms and $I_2$ is reduced compared to $I_1$ according to

$$I_2 = I_1 e^{-n_{\text{Hg}} \sigma l}, \quad (2.21)$$

where $n_{\text{Hg}}$ is the mercury particle density, $\sigma$ is the absorption cross-section for light from a $^{204}$Hg lamp and $l$ is the path the light traverses through the precession chamber. The absorption $A$ is
Figure 2.9: The $^{199}$Hg magnetometer in the nEDM experiment. For details, see text.
then
\[ A = \frac{I_1 - I_2}{I_1} = 1 - e^{-n_{\text{Hg}}\sigma t}. \] (2.22)

The absorption of light depends on the orientation of the spin and the effective cross-section oscillating between \( \sigma(1-P) \) (spins parallel to the light) and \( \sigma(1+P) \) (spins antiparallel to the light), where \( P \) is the polarisation of the mercury atoms. The amplitude of the precession signal is
\[ a_s = I_1 e^{-n_{\text{Hg}}\sigma(1-P)t} - I_1 e^{-n_{\text{Hg}}\sigma t} = I_1 e^{-n_{\text{Hg}}\sigma t}((e^{-n_{\text{Hg}}\sigma t} - P - 1), \] (2.23)
and depends on the different absorption values. Replacing the exponential expression by absorption \( A \) gives:
\[ a_s = I_1 ((1 - A)((1 - A)^{-P} - 1)). \] (2.24)

Since the amplitude of the signal and the absorption are measured, the polarisation can be calculated:
\[ P = -\frac{\log(1 + \frac{a_s}{I_1(1-A)})}{\log(1 - A)}. \] (2.25)

Knowing that \( a_s \ll I_1 \), the polarisation can be written as
\[ P \approx \frac{a_s}{I_1 A(1 - \frac{1}{2})}. \] (2.26)

The precession signal is not constant but decays with time. For small absorptions, \( a_s \) is proportional to \( A \) and \( P \). One can therefore write:
\[ a(t) = a_s \exp(-t/\tau), \] (2.27)
where \( \tau \) is the decay time. There are two mechanisms which contribute to \( \tau \).

- **Depolarisation:** Mercury atoms are depolarised due to wall collisions, residual gas collisions and magnetic inhomogeneities. The polarisation decays exponentially with the depolarisation time \( T_2 \).

- **Leakage:** The precession chamber is not entirely gas tight and the mercury atoms leak out of the chamber with a certain rate and a characteristic loss time \( T_{\text{loss}} \) can be defined. With the measurement of \( I_3 \), \( T_{\text{loss}} \) can be determined: \( T_{\text{loss}} = T/\ln(\frac{I_2-I_1}{I_3-I_1}) \).

\( T_2 \) and \( T_{\text{loss}} \) enter \( \tau \) as follows:
\[ \tau = \left( \frac{1}{T_2} + \frac{1}{T_{\text{loss}}} \right)^{-1}. \] (2.28)

\( T_{\text{loss}} \) is on the order of several 100 s and \( T_2 \) can be well above 100 s in a well prepared system.

**A metallic \(^{199}\text{Hg} \) source**

Recently, a source with metallic \(^{199}\text{Hg} \) has been developed in order to study the effect of \( \text{O}_2 \) on \( \tau \). For long leakage times on the order of several 100 s, \( \tau \approx T_2 \), where \( T_2 \) is the polarisation lifetime of the \(^{199}\text{Hg} \) atoms. When starting the operation of the magnetometer after pumping down from atmospheric pressure, \( T_2 \) is initially between 10 s and 30 s, which is unacceptably small for EDM runs with a precession time of 150 s. The reason for the bad polarisation lifetime is thought to be the presence of polar molecules and radicals, which are on the inside walls of the precession chamber. The RAL-Sussex-ILL collaboration discovered a way to increase \( T_2 \) to 100 s and higher by performing discharge cleanings. In this procedure, the precession chamber is filled with \( \text{O}_2 \).
at a pressure of about 1 mbar. Then, an electric field is applied until electrical discharge occurs
and some active oxygen is formed. The active oxygen probably neutralises polar molecules on
the wall and removes them. The polarity of the electric field is changed several times during the
procedure. The whole process takes about half an hour. A more detailed description of discharge
cleaning is given in [May98].

During their measurements, the RAL-Sussex-ILL collaboration observed a correlation between
the absorption \( A \) of the probe light and \( T_2 \) [May98]. The more mercury enters the precession
chamber, the larger \( T_2 \). A possible explanation for this behaviour might be the presence of oxygen,
which is formed as a by-product in the HgO source. The oxygen might help to increase \( T_2 \) in a
way similar to that of the active one used in discharge cleanings. Investigations on this correlation
were done using a metallic Hg source. A small bead of metallic \(^{199}\)Hg was filled in a second source
container, which was then connected to the experiment instead of the HgO source. Due to the low
pressure in the experiment, a steady evaporation from the bead feeds the polarisation chamber
with mercury similar to the HgO source. The vapour pressure at room temperature of metallic
Hg is \( 2.1 \times 10^{-3} \) mbar [Gra72]. A cooling device controlled the evaporation of mercury and thus
the amount of mercury in the chamber.

The two different sources were tested at ILL. Both sources show the same correlation between
\( T_2 \) and absorption (see Fig. 2.10). The conclusion of the comparison is that \( T_2 \) improves with
the absorption also when oxygen is absent. Therefore, more mercury in the precession chamber
increases \( T_2 \). A possible explanation might be that some of the mercury atoms stick to the
depolarising molecules and neutralise them. A greater amount of mercury would then result in
a better neutralisation and thus a longer polarisation lifetime. The two sources are comparable
in terms of stability. The operation of the metallic source at room temperature results in an
absorption of roughly 30\%.

Figure 2.10: Comparison of Hg sources. The figure shows the dependence
of \( T_2 \) on the absorption of the probe light. Both Hg sources show the same
correlation. The slightly higher \( T_2 \)-values for the metallic source may come
from a longer vacuum exposure in vacuum.
Together with $\tau$, all parameters $A$, $P$, and $a_s$ make a statement on the performance of the magnetometer, but the main interest is the determination of the frequency.

**Sensitivity of the magnetometer**

The magnetometer gives an average precession frequency for each Ramsey cycle. In order to make the most precise determination of the frequency of a cycle, a method is used where the phases at the beginning and at the end of the precession are determined in a fit. The total phase difference is then divided by the precession time [Gre98]. The duration of the fitting windows $t_{fit}$ are in the order of several seconds. The zero crossings from minus to plus are counted and contribute with $2\pi n$ to the total phase. The sensitivity of the frequency can be estimated to be

$$\sigma_f \approx \frac{1}{4T'} \frac{a_n^1}{a_s} \frac{1}{\sqrt{n}} \sqrt{1 + e^{2T'/\tau}}, \quad (2.29)$$

where $T' = T - 2t_{fit}$, $a_n$ the amplitude of the noise, $n$ the number of datapoints used for fitting [Gre98]. The formula is only valid for the frequency extraction described above. The best value obtained for the error of the frequency was $\approx 25 \text{ fT}$ at ILL [Gre98].

Improving the $^{199}\text{Hg}$ magnetometer means increasing the sensitivity. In the following, a brief glimpse of each parameter and its influence on the sensitivity is given.

- $T'$: Making the precession time larger than the 150 s does not make any sense, unless the UCN can be stored longer.

- $n$: Increasing the number of data points by increasing the sampling rate will not improve the sensitivity. The narrow bandwidth of the filter ($\approx 2 \text{ Hz}$) does not allow independent measurements of adjacent points, when the sampling rate is too large.

- $a_s$: The amplitude of the signal is $a_s = I_1 PA(1 - A/2)$. Increasing the intensity of the light $I_1$ is not advisable, since the photomultiplier runs into saturation at some point. Rather promising seems to be the increase of the polarisation $P$. It depends significantly on the wall coating, as seen in Fig. 2.11. Additionally, the strong dependence on the absorption $A$ leads to the assumption that a higher polarisation is reached for smaller amounts of mercury. In fact, mercury vapour is polarised more efficiently at lower densities [Gre98]. A larger polarisation chamber would decrease the density and hence increase the polarisation for a certain amount of mercury.

- $a_n$: Reducing the noise has already been tested at ILL [Roc09]. The relative noise level was found to be $a_n/I_1 = 8 \times 10^{-5}$ compared to $a_{n,\text{shot}}/I_1 = 5 \times 10^{-5}$ for the shot noise. Decreasing the noise further is difficult, since the measured value is already close to shot noise, which sets a natural limit on the noise level.

- $\tau$: The decay time of the signal must be large enough so that the S/N-ratio at the end of the precession is still the same order of magnitude as at the beginning. This is the case for $\tau \approx T$. Usually, $\tau$ is well above 100 s, which is sufficient for a precession time of 150 s. A further increase in $\tau$ improves the sensitivity only slightly.

Among all these possibilities, increasing the polarisation seems to be the most feasible way to make the magnetometer more sensitive. A study on different coating methods and materials as well as investigations on different sizes of the polarisation chamber are part of this thesis.
Figure 2.11: The figure shows the dependence of the polarisation of the mercury vapour on the absorption. Higher polarisations are reached for a newly-coated polarisation chamber. A good wall coating is therefore crucial for a reliable performance. In addition, the strong absorption dependence of the polarisation shows, that mercury at lower densities is better polarised. A larger polarisation chamber might help to increase the polarisation. The data stem from the last measurements at ILL.
3 An effective model for $^{199}$Hg polarisation

$^{199}$Hg is polarised by optical pumping, as already mentioned in Sec. 2.4.2. In this process, unequal populations of magnetic sublevels are produced due to the excitation of the atoms with polarised light from a $^{204}$Hg lamp.

In the following, the subatomic structure of $^{199}$Hg and $^{204}$Hg are discussed. Their energy levels are illustrated in Fig. 3.1. Mercury is diamagnetic and thus no isotope has an electronic spin. $^{199}$Hg has a nuclear spin 1/2. Therefore, the ground state consists of two Zeeman levels, $m_F = -1/2$ and $m_F = +1/2$, which are equally populated at room temperature. In the experiment, we use the transition ($^1S_0$, $F = 1/2$) $\rightarrow$ ($^3P_1$, $F = 1/2$) to polarise the $^{199}$Hg population. The atoms are illuminated by circularly polarised resonant light with a wavelength of 254 nm, denoted as $\sigma^+$ for right handed polarisation. This light has an angular momentum $\Delta m = +1$. Atoms with $m_F = -1/2$ absorb the light by making the transition to $^3P_1$, $F = 1/2$, $m_F = +1/2$ (blue arrow in Fig. 3.1), while atoms with $m_F = +1/2$ can not absorb light because of energy and

---

Figure 3.1: The plot shows the atomic sublevels of $^{199}$Hg (upper part) and $^{204}$Hg (lower part). The plot has been taken from [Bro52] and was slightly adapted. The arrows of the optical pumping cycle have been added. For details see text.
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angular momentum conservation. From this excited state, two decays are possible (red arrows in Fig. 3.1). The decay to the level \( m_F = -1/2 \) just reverses the excitation by emitting a \( \sigma^+ \)-photon, whilst the decay to the level \( m_F = +1/2 \) transfers the angular momentum from the electron to the nucleus by emitting a \( \pi \)-photon, which is linearly polarised. The branching ratio of these two decays are 2/3 for \( m_F = -1/2 \) and 1/3 for \( m_F = +1/2 \) according to the Clebsch-Gordan coefficients. This procedure transfers atoms from the \( m_F = -1/2 \) ground state to the \( m_F = +1/2 \) ground state, which results in a polarisation of the \(^{199}\text{Hg}\) ensemble.

The resonant light stems from a \(^{204}\text{Hg}\) lamp. The \(^{204}\text{Hg}\) atoms are excited by microwaves to the \( 3P_1, F = 1 \) state, which decays again by emitting a photon. A polarisation optic selects the \( \sigma^+ \)-light, which is used for optical pumping (green arrow in Fig. 3.1). The emission line of \(^{204}\text{Hg}\) is only shifted by 96 MHz from that of \(^{199}\text{Hg}\) [Cor77], which is small compared to the Doppler broadened width of the absorption curve, which is \( \approx 1 \) GHz. The overlap is hence excellent, which guarantees an efficient pumping.

Resonant light from a \(^{199}\text{Hg}\) lamp would be less efficient for optical pumping, since a part of the \( \sigma^+ \) light excites also the sublevel \( m_F = +1/2 \) to \( 3P_1, F = 3/2, m_F = +3/2 \), which has no net effect, since this state decays to 100% back to the sublevel \( m_F = +1/2 \). There is no net change in the populations for this ground state.

### 3.1 Evolution of the polarisation

The imbalance of populations for the two ground state levels \( m_F = -1/2 \) and \( m_F = +1/2 \) can be expressed by the polarisation \( P \), which is defined as follows:

\[
P = \frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow},
\]

where \( N_\uparrow \) is the number of atoms for \( m_F = +1/2 \) and \( N_\downarrow \) the number of atoms for \( m_F = -1/2 \). The polarisation is therefore nothing else than the normalised difference between the number of atoms in the \( m_F = +1/2 \) state and the atoms in the \( m_F = -1/2 \) state, where \( N = N_\uparrow + N_\downarrow \).

#### 3.1.1 Optical pumping

For the build-up of the polarisation, one can define a differential equation which describes the change of the spin down population with time:

\[
\frac{dN_\downarrow}{dt} = -\frac{1}{3} \Phi \mathcal{P}_{\text{abs}},
\]

whereas for spin up

\[
\frac{dN_\uparrow}{dt} = -\frac{dN_\downarrow}{dt},
\]

where \( \Phi \) is the initial photon flux (number of photons per second), \( \mathcal{P}_{\text{abs}} \) is the probability that a photon is absorbed by the \(^{199}\text{Hg}\) ensemble and \( N_\downarrow, N_\uparrow \) the number of atoms with spin down, up respectively. The factor of 1/3 is the branching ratio of the decay channels; only one out of three \(^{199}\text{Hg}\) atoms will be in the +1/2 state after having absorbed a photon.

The absorption probability can be found by a geometrical consideration. The area of a \(^{199}\text{Hg}\) atom which is seen by a photon is its effective cross-section \( \sigma \). The beam cross-section of the light traversing the cell shall be denoted as \( A_{\text{lamp}} \). The absorption probability is then

\[
\mathcal{P}_{\text{abs}} = 1 - \exp(-N_{\text{ill},\downarrow} \sigma / A_{\text{lamp}}),
\]
where \( N_{\text{ill},↓} \) is the number of the illuminated atoms with \( m_F = -1/2 \). Expressing \( N_{\text{ill},↓} \) in terms of the total number of atoms in the cell \( N \) with \( m_F = -1/2 \):

\[
N_{\text{ill},↓} = N_↓ \frac{A_{\text{lamp}}}{(V/l)},
\]

where \( V \) is the volume of the cell and \( l \) is the length of the cell, \( P_{\text{abs}} \) changes to:

\[
P_{\text{abs}} = (1 - \exp(-N_↓ \sigma l/V)).
\]

The differential equation then changes to:

\[
\frac{dN_↓}{dt} = -\frac{1}{3} \Phi(1 - \exp(-N_↓ \sigma l/V)).
\]

Using Eqn. 3.1 and the fact, that \( N \) is constant in time, the evolution of the polarisation can be expressed as follows:

\[
\frac{dP}{dt} = -\frac{2}{N} \frac{dN_↓}{dt} = \frac{2}{3} \Phi \frac{(1 - \exp(-N_↓ \sigma l/V))}{N}
\]

Replacing \( N_↓ \) with the polarisation \( P \) results in\(^1\)

\[
\frac{dP}{dt} = \frac{2}{3} \Phi \frac{(1 - \exp(-(1 - P)N\sigma l/(2V)))}{N}
\]

At the beginning, both populations are equal and the polarisation is zero. This implies the boundary condition \( P(t = 0) = 0 \). An example of the evolution of the polarisation with time is plotted in Fig. 3.2. Since no relaxation or depolarisation is taken into account so far, the polarisation reaches 100%.

### 3.1.2 Relaxation rate \( \Gamma_r \)

There are a few mechanisms, which destroy the polarisation of the mercury again (see Sec. 2.4.2). For each of these mechanisms, a relaxation rate \( \Gamma_r \) for the polarisation can be defined. The inverse of the relaxation rate is the relaxation time \( T_r \).

- **Wall collisions:**
  The strongest relaxation mechanism are wall collisions. Dipoles located at the surface of the chamber interact with the magnetic moment of the \(^{199}\text{Hg} \) atoms. For each single wall collisions exists a probability \( \epsilon \) for disorientation of the spin. The relaxation rate for wall collisions is then

\[
\Gamma_{\text{wall}} := \frac{1}{T_{\text{wall}}} = \frac{\epsilon \bar{v}}{\lambda}
\]

where \( \bar{v} \) the averaged velocity and \( \lambda \) the mean free path of the \(^{199}\text{Hg} \) atoms in the cell.

- **Magnetic field inhomogeneities:**
  There is also the possibility that gradients of magnetic fields could produce a spin flip. In this case, a longitudinal relaxation rate \( \Gamma_{1,\text{mag}} \) and a transverse relaxation rate \( \Gamma_{2,\text{mag}} \) can be defined. For aligning the spin along a certain direction, only \( \Gamma_{1,\text{mag}} \) is of interest, \( \Gamma_{2,\text{mag}} \)

\(^{1}\)Eqn. 3.8 and all the subsequent differential equations are solved numerically with a MATLAB program, which uses an already implemented solver for initial value problems. The solver used for this problem is ode23s, a one step solver with an adaptive stepsize. Further informations are provided on the MATLAB homepage [Mat12].
is given for the sake of completeness. The relaxation rates due to magnetic field gradients can be expressed as follows [Cat88]:

\[ \Gamma_{1,\text{mag}} := \frac{1}{T_{1,\text{mag}}} = \frac{8R^4\gamma^2_{\text{Hg}}}{175D} (|\vec{\nabla}B_x|^2 + |\vec{\nabla}B_y|^2), \quad (3.11) \]

and

\[ \Gamma_{2,\text{mag}} := \frac{1}{T_{2,\text{mag}}} = \frac{4R^4\gamma^2_{\text{Hg}}}{175D} (|\vec{\nabla}B_x|^2 + |\vec{\nabla}B_y|^2 + 2|\vec{\nabla}B_z|^2), \quad (3.12) \]

where \( \vec{\nabla}B_x, \vec{\nabla}B_y, \) and \( \vec{\nabla}B_z \) are the gradients over the volume of the polarisation chamber, \( R \) the characteristic length of the chamber, and \( D \) the diffusion constant.

- Residual gas collisions:
  A third possibility is the relaxation due to the residual gas collisions. Similar to the relaxation due to wall collisions, a relaxation rate can be defined:

\[ \Gamma_{\text{gas}} := \frac{1}{T_{\text{gas}}} = \frac{\pi}{\lambda_{\text{gas}}}, \quad (3.13) \]

where \( \lambda_{\text{gas}} \) is the mean free path for a \(^{199}\text{Hg} \) atom in gas.

These three relaxation mechanisms can be summarised in an over-all relaxation rate:

\[ \Gamma_r = \Gamma_{\text{wall}} + \Gamma_{1,\text{mag}} + \Gamma_{\text{gas}}. \quad (3.14) \]

The expressions from Eqn. 3.7 for the evolution change to

\[ \frac{dN_i}{dt} = -\frac{1}{3}\Phi(1 - \exp(-N_1\sigma l/V)) - \frac{1}{2}N_i\Gamma_r + \frac{1}{2}N_\uparrow\Gamma_r. \quad (3.15) \]

A decrease in the \( m_F = -1/2 \) population leads at the same time to an increase in the \( m_F = +1/2 \) population and vice versa. The polarisation is therefore changed by two units, when the spin of a \(^{199}\text{Hg} \) atom is flipped. The expression for the polarisation changes accordingly:

\[ \frac{dP}{dt} = \frac{2}{3}\Phi\left(1 - \exp(-(1 - P)N\sigma l/(2V))\right) - \Gamma_r P. \quad (3.16) \]

The evolution of the polarisation for different relaxation rates \( \Gamma_r \) is illustrated in Fig. 3.2.

### 3.1.3 Depolarisation by reemitted light

Another source of depolarisation is reemitted light in the optical pumping process. A part of this stray light can depolarise the \(^{199}\text{Hg} \) ensemble again [Gre98]. In the next steps, emitted photons can again excite other atoms which afterwards emit light and so on. In the following, I will show that in higher order processes \( \pi \)- and \( \sigma^- \)-photons occur which are responsible for the depolarisation.
Figure 3.2: The plot shows calculated polarisation curves using Eqn. 3.16. The evolution of polarisation depends crucially on the relaxation rate $\Gamma_r$. For no relaxation rate, the polarisation reaches 100%. The following typical parameter settings have been chosen: $\Phi = 4.5 \times 10^{12} \text{s}^{-1}$ [Fer11], $N = 4.0 \times 10^{13}$ atoms, $\sigma = 2.6 \times 10^{-17} \text{m}^2$ [Pen05], $l = 0.287 \text{m}$ and $V = 1.2 \times 10^{-3} \text{m}^3$. 
First order reemission

The emitted photons in optical pumping are in first order $\pi$-photons with a probability of $1/3$ and $\sigma^+$-photons with a probability of $2/3$. Adding the first order reemission terms, Eqn. 3.15 changes to:

$$
\frac{dN_\downarrow}{dt} = -\frac{1}{3}\Phi(1 - \exp(-N_\downarrow\sigma_l/V)) - \frac{1}{2}N_\downarrow\Gamma_r + \frac{1}{2}N_r\Gamma_r
+ \frac{2}{3}\Phi_\pi(1[(1 - \exp(-N_\downarrow\sigma_\zeta/V)) - (1 - \exp(-N_\downarrow\sigma_\zeta/V))])
- \frac{1}{3}\Phi_{\sigma^+}(1)(1 - \exp(-N_\downarrow\sigma_\zeta/V)),
$$

(3.17)

where $\Phi_\pi(1)$ and $\Phi_{\sigma^+}(1)$ are the photon fluxes of the reemitted light:

$$
\Phi_\pi(1) = \frac{1}{3}\Phi(1 - \exp(-N_\downarrow\sigma_l/V))
$$

(3.18)

for $\pi$-light and

$$
\Phi_{\sigma^+}(1) = \frac{2}{3}\Phi(1 - \exp(-N_\downarrow\sigma_l/V))
$$

(3.19)

for $\sigma^+$-light. The parameter $\zeta$ is the mean free path of the light in the chamber. Similar as in the section before, the evolution of the polarisation can be calculated:

$$
\frac{dP}{dt} = \frac{2}{3}\Phi\left(1 - \exp\left(- \left(1 - P\right)\frac{N\sigma_l}{2V}\right)\right) - \Gamma_rP
+ \frac{8}{9}\Phi\left(1 - \exp\left(- \left(1 - P\right)\frac{N\sigma_l}{2V}\right)\right)\left(1 - \exp\left(- \left(1 - P\right)\frac{N\sigma_\zeta}{2V}\right)\right)
- \frac{4}{9}\Phi\left(1 - \exp\left(- \left(1 - P\right)\frac{N\sigma_l}{2V}\right)\right)\left(1 - \exp\left(- \left(1 + P\right)\frac{N\sigma_\zeta}{2V}\right)\right),
$$

(3.20)

where the last two lines represent the reemission effect.

Second order reemission

The result of the first order reemission is $\sigma^+$- and $\pi$-light, which produces reemission of second order.

The obtained $\sigma^+$-light excites again the $m_F = -1/2$ level, which contributes to the pumping. The result is $2/3 \sigma^+$-light and $1/3 \pi$-light as second order. The obtained $\pi$-light can excite the $m_F = -1/2$-level and the $m_F = +1/2$-level likewise. In the first case, $2/3 \sigma^-$-light and $1/3 \pi$-light is produced and in the second case, $2/3 \sigma^+$ and $1/3 \pi$-light.

All the processes, which occur in remission effects, are shown in Fig. 3.3. The differential equation for the spin down population including second order reemission is then:

$$
\frac{dN_\downarrow}{dt} = -\frac{1}{3}\Phi(1 - \exp(-N_\downarrow\sigma_l/V)) - \frac{1}{2}N_\downarrow\Gamma_r + \frac{1}{2}N_r\Gamma_r
+ \frac{2}{3}\Phi_\pi(1[(1 - \exp(-N_\downarrow\sigma_\zeta/V)) - (1 - \exp(-N_\downarrow\sigma_\zeta/V))])
- \frac{1}{3}\Phi_{\sigma^+}(2)(1 - \exp(-N_\downarrow\sigma_\zeta/V))
+ \frac{2}{3}\Phi_{\sigma^+}(2)(1 - \exp(-N_\downarrow\sigma_\zeta/V)) - \frac{1}{3}\Phi_{\sigma^+}(2)(1 - \exp(-N_\downarrow\sigma_\zeta/V))
$$

(3.21)
where $\Phi_{\sigma^-}(2)$, $\Phi_{\pi}(2)$, and $\Phi_{\sigma^+}(2)$ are the corresponding photon fluxes of the reemitted light of second order:

$$\Phi_{\sigma^-}(2) = \frac{2}{3} \Phi_{\pi}(1)(1 - \exp(-N_\downarrow \sigma \zeta / V))$$

(3.22)

for $\sigma^-$-light,

$$\Phi_{\pi}(2) = \frac{1}{3} \Phi_{\sigma^+}(1)(1 - \exp(-N_\downarrow \sigma \zeta / V))$$

$$+ \frac{1}{3} \Phi_{\pi}(1)((1 - \exp(-N_\downarrow \sigma \zeta / V)) + (1 - \exp(-N_\uparrow \sigma \zeta / V))$$

(3.23)

for $\pi$-light, and

$$\Phi_{\sigma^+}(2) = \frac{2}{3} \Phi_{\sigma^+}(1)(1 - \exp(-N_\downarrow \sigma \zeta / V)) + \frac{2}{3} \Phi_{\pi}(1)(1 - \exp(-N_\uparrow \sigma \zeta / V))$$

(3.24)

for $\sigma^+$-light. This translates to a change in polarisation as follows:

$$\frac{dP}{dt} = \frac{2}{3} \Phi \frac{z}{N} - \Gamma_1 P$$

$$+ \frac{8}{9} \Phi \frac{z(1 - \exp(-(1 - P)N\sigma \zeta / (2V)))}{N}$$

$$- \frac{4}{9} \Phi \frac{z(1 - \exp(-(1 + P)N\sigma \zeta / (2V)))}{N}$$

$$+ \frac{20}{27} \Phi \frac{z(1 - \exp(-(1 - P)N\sigma \zeta / (2V))))^2}{N}$$

$$- \frac{8}{27} \Phi \frac{z(1 - \exp(-(1 - P)N\sigma \zeta / (2V))))(1 - \exp(-(1 + P)N\sigma \zeta / (2V)))}{N}$$

$$- \frac{4}{27} \Phi \frac{z(1 - \exp(-(1 + P)N\sigma \zeta / (2V))))^2}{N},$$

(3.25)

where $z = 1 - \exp(-(1 - P)N\sigma l / (2V))$. An example of the evolution of polarisation with reemission is plotted in Fig. 3.4.

Figure 3.3: The plot shows all possible transitions resp. decays between the $(^3P_1, F = 1/2)$ and $(^1S_0, F = 1/2)$ state with $\sigma^+$, $\sigma^-$, and $\pi$-light, where $\pi$-light and $\sigma^-$ light destroy the polarisation of the ensemble again. For decays where $m_F$ does not change, the probability is 1/3 and for decays with $\Delta m = \pm 1$, the probability is 2/3.
3.1.4 Dilution of the polarisation

There is an additional effect which has an influence on the polarisation. So far, the total number of atoms $N$ is constant with time. A change of the polarisation is then only due to the change in $\Delta N = N^\uparrow - N^\downarrow$. However, a constant flow of unpolarised mercury into the polarisation chamber dilutes the polarised mercury. The polarisation can be expressed with $\Delta N$ and $N$ as follows:

$$P = \frac{\Delta N}{N}$$

(3.26)

The total change in time is then

$$\frac{dP}{dt} = \frac{(d\Delta N/dt)N - \Delta N(dN/dt)}{N^2}$$

(3.27)

with $dN/dt = \Gamma_0$, where $\Gamma_0$ is the mercury flux into the polarisation chamber. An additional term occurs due to the dilution:

$$\frac{dP}{dt} = \frac{1}{N} \left( \frac{d\Delta N}{dt} - \frac{\Delta N}{N} \right) \frac{\Gamma_0}{N}.$$  

(3.28)

The first part contains the terms of Eqn. 3.25. One can assume, that $\Gamma_0$ is constant: $\Gamma_0 = N/t$. The complete expression for the polarisation finally is

$$\frac{dP}{dt} = \frac{2}{3} \Phi \frac{z}{\Gamma_0 t} - \Gamma_t P$$

$$+ \frac{8}{9} \Phi \frac{z(1 - \exp(-(1 - P)\Gamma_0 t \sigma \zeta/(2V)))}{\Gamma_0 t}$$

$$- \frac{4}{9} \Phi \frac{z(1 - \exp(-(1 + P)\Gamma_0 t \sigma \zeta/(2V)))}{\Gamma_0 t}$$

$$+ \frac{20}{27} \Phi \frac{z(1 - \exp(-(1 - P)\Gamma_0 t \sigma \zeta/(2V)))^2}{\Gamma_0 t}$$

$$- \frac{8}{27} \Phi \frac{z(1 - \exp(-(1 + P)\Gamma_0 t \sigma \zeta/(2V)))(1 - \exp(-(1 + P)\Gamma_0 t \sigma \zeta/(2V))))}{\Gamma_0 t}$$

$$- \frac{4}{27} \Phi \frac{z(1 - \exp(-(1 + P)\Gamma_0 t \sigma \zeta/(2V)))^2}{\Gamma_0 t} \frac{P}{t},$$

(3.29)

where $z = 1 - \exp(-(1 - P)\Gamma_0 t \sigma \zeta/(2V))$. The solution is plotted and compared with the case without dilution (Eqn. 3.25) in Fig. 3.4. Finally, the model is used to calculate the polarisation in the test setup (see next chapter). A comparison of the calculation with the polarisation measured with the setup is made in Sec. 4.3.

3.2 Approximation for small $^{199}$Hg densities

For small absorption values, reemission can be neglected. Furthermore, the exponential expressions from Sec. 3.1 can be developed. The expansion up to first order is

$$1 - \exp(-\delta) \approx \delta.$$  

(3.30)

Applying this to the equations above, linear differential equations of first order are obtained. In the following, the polarisation for the two cases with and without dilution are calculated.
Figure 3.4: The calculated polarisation curves include the processes of pumping, relaxation, and reemission. The difference between the curves is due to dilution. The following values have been chosen: \( \Phi = 4.5 \times 10^{12} \text{s}^{-1} \), \( \Gamma_r = 0.01 \text{s}^{-1} \), \( \zeta = 4V/S = 6.4 \text{cm} \), \( N = 4.0 \times 10^{13} \), \( \Gamma_0 = 1.3 \times 10^{11} \text{s}^{-1} \).

### 3.2.1 Solution without dilution

First, the case without the dilution term \(-P/t\) is investigated. Using the first order Taylor expansion, Eqn. 3.16 changes to

\[
\frac{dP}{dt} = \frac{\Phi \sigma l}{3V}(1 - P) - \Gamma_r P. \tag{3.31}
\]

The first term in the equation above can be expressed by a pump rate \( \Gamma_p \):

\[
\Gamma_p = \frac{\Phi \sigma l}{3V}. \tag{3.32}
\]

Eqn. 3.31 can then be rewritten as

\[
\frac{dP}{dt} = \Gamma_p (1 - P) - \Gamma_r P. \tag{3.33}
\]

A further simplification leads to

\[
\frac{dP}{dt} = gP + h, \tag{3.34}
\]

where

\[
g = -\Gamma_p - \Gamma_r \\
h = \Gamma_p \tag{3.35}
\]
Linear differential equations of first order are solvable analytically. The solution

\[ P = \left( \int_{t_0}^{t} h(t') \exp(-G(t')) dt' + C_0 \right) \exp(G(t)) \]  

(3.36)

with

\[ G(t) = \int_{t_0}^{t} g(t') dt'. \]

(3.37)

is derived in Sec. A. Since \( g \) and \( h \) are constant, the integrals in Eqn. 3.36 and Eqn. 3.37 are trivial and the result is

\[ P(t) = \left[ -\frac{h}{g} \exp(-gt) + C_0 \right] \exp(gt), \]

(3.38)

where \( C_0 \) is a free parameter and not yet determined. The boundary condition \( P(t = 0) = 0 \) fixes the parameter to \( C_0 = h/g \). The complete solution is

\[ P(t) = -\frac{h}{g} \left[ 1 - \exp(gt) \right]. \]

(3.39)

The solution can be expressed by the pump rate and the relaxation rate:

\[ P(t) = \frac{\Gamma_p}{\Gamma_p + \Gamma_r} \left[ 1 - \exp(-(\Gamma_p + \Gamma_r)t) \right]. \]

(3.40)

The analytic solution and the numeric solution (from Eqn. 3.21) are plotted in Fig. 3.5 for comparison.

### 3.2.2 Solution with dilution

The case, where the dilution is present, can also be solved analytically. Starting from Eqn. 3.33, the term \(-P/t\) is added:

\[ \frac{dP}{dt} = \Gamma_p (1 - P) - \Gamma_r P - \frac{P}{t}. \]

(3.41)

Similar to Sec. 3.2.1, the equation is

\[ \frac{dP}{dt} = gP + h, \]

(3.42)

where \( g \) and \( h \) are defined as follows:

\[ g := g_0 - \frac{1}{t} = -\Gamma_p - \Gamma_r - \frac{1}{t}, \]

\[ h = \Gamma_p. \]

(3.43)

\( g \) is now depending on \( t \), which makes the integration in Eqn. 3.36 a bit more complicated. First, \( G(t) \) has to be determined:

\[ G(t) = \int_{t_0}^{t} g_0 - \frac{1}{t'} dt' = g_0 t - \log(t) + k, \]

(3.44)

where \( k = \log(t_0) - g_0 t_0 \). Substituting \( G(t) \) into Eqn. 3.36 results in

\[ P(t) = \left( \int_{t_0}^{t} t' \exp(-g_0 t') dt' h \exp(-k) + C_0 \right) \frac{1}{t} \exp(g_0 t) \exp(k) \]

(3.45)
The integral can be solved by partial integration as follows:

\[
P(t) = \left[ \left( - \frac{t \exp(-g_0 t)}{g_0} - \frac{\exp(-g_0 t)}{g_0^2} \right) + \frac{t_0 \exp(-g_0 t_0)}{g_0^2} + \frac{\exp(-g_0 t_0)}{g_0^2} \right] \cdot I(t_0) + C_0 \\
\times \frac{1}{t} \exp(g_0 t) \exp(k).
\]  

(3.46)

The expression can be simplified to

\[
P(t) = -\frac{h}{g_0} - \frac{h}{t g_0^2} + C_1 \frac{1}{t} \exp(g_0 t),
\]  

(3.47)

where \(C_1\) is a free parameter. Knowing, that \(P(t = 0) = 0\) must be satisfied, the result changes to

\[
P(t) = \frac{\Gamma_p}{\Gamma_p + \Gamma_r} \left[ 1 - \frac{1}{t(\Gamma_p + \Gamma_r)} \left[ \exp(- (\Gamma_p + \Gamma_r) t) \right] \right].
\]  

(3.48)

The analytic and numeric results for the two cases with and without dilution are compared in Fig. 3.5. In the case of small mercury densities in the chamber, the approximation is pretty good. For \(N = 4.0 \times 10^{13}\), the results are in agreement within 2%.
4 Investigations of depolarisation mechanisms

A detailed study of depolarisation effects is crucial to an enhancement of the signal. There are also some not yet understood features of the $^{199}$Hg magnetometer which are important to investigate in order to guarantee a stable operation. It was therefore useful to build a dedicated setup to perform test measurements independent from the main experiment. Such a setup can be much simpler and easier to handle than the nEDM apparatus. The following list shows items which can be investigated by such a setup:

- **Anti-relaxation coatings:**
  Coatings prevent polarised mercury from depolarising on the walls [Rob58]. At the moment, Fomblin oil and Fomblin grease are used. The application procedure is rather time consuming and tedious, since it is difficult to achieve a complete coverage of the surface. There are possibly other materials that show longer relaxation times for $^{199}$Hg [Rom03] and are easier to apply.

- **Reemission:**
  A next point is the emission of light. The polarisation decreases with higher absorptions and thus higher $^{199}$Hg densities (see Fig. 2.11). This effect stems from the absorption of reemitted light which depolarises the $^{199}$Hg. A higher mercury density increases the absorption of emitted light. A larger volume of the polarisation chamber for the same amount of mercury would lower the mercury density and thus reduce the effect of the reemission.

- **Dilution:**
  This effect decreases the polarisation due to unpolarised mercury flowing continuously into the polarisation chamber. In order to avoid this dilution, the mercury could be filled into the chamber at once, at the beginning of the polarisation process.

- **E-field dependent effects:**
  An interesting feature is the characteristic behaviour of $T_2$ under repeated polarity changes of the high voltage (HV) [Gre98]. $T_2$ decreases suddenly after changing polarity and recovers slowly afterwards (see Fig. 5.1). This effect is independent of the polarisation but affects the sensitivity of the magnetometer via $T_2$. It has to be studied in order to remove or reduce it.

- **Outgassing:**
  A further aspect is the outgassing properties of surfaces with respect of the $^{199}$Hg performances.
In this work, the first two issues are investigated. The goal of the following studies is to increase the polarisation and hence the amplitude of the signal. Different coatings are tried out in order to find the coating with the lowest relaxation rate and the easiest application. For studying reemission, the signal and the polarisation is measured for different mercury densities and different volumes of glass cells where mercury is polarised.

4.1 A dedicated $^{199}\text{Hg}$ test setup

With the device, it is possible to fill mercury in a cell, to polarise it and to measure the polarisation after a certain time. The setup is based on the working method of the $^{199}\text{Hg}$ magnetometer of the nEDM experiment.

4.1.1 Magnetic Environment

For polarisation of mercury, a weak homogeneous magnetic field is necessary. Therefore the external field has to be highly suppressed. The shielding of the environmental magnetic field is done by three co-centric tubes of mu metal. The single cylinders are 1800 mm, 1750 mm, and 1700 mm long and have a diameter of 450 mm, 400 mm, and 360 mm respectively. Measurements of the field inside give an estimate for the shielding factor of roughly 2000 in the radial direction and 500 in the direction of the axis of the shield. The coordinate system is defined as follows: the horizontal radial direction is the $x$-axis, the vertical radial direction is the $y$-axis and the symmetry axis of the shield is the $z$-axis (see Fig. 4.1).

The easiest way to optically pump $^{199}\text{Hg}$ vapour is to align the pump light along the shield axis. In this configuration, the magnetic holding field has to be parallel to the light beam for
most efficient pumping. The holding field $\vec{B}_0$ is produced by a solenoid wound on a PVC tube. The coil has a length of 1600 mm and consists of 760 windings. A current of 1.6 mA produces a magnetic field of 1 $\mu$T along the axis of the shield. There is a remanent field of about 50 nT when the coil is off. The current needed to produce the field of 1 $\mu$T was determined in situ, by using a fluxgate magnetometer inside the shield to measure the magnetic field.

To obtain a modulation in the light beam, a precession around a magnetic field perpendicular to the $\vec{B}_0$ direction is necessary. Here, a second static field along the $x$-direction is used. I refer to this field as $\vec{B}_1$. The coil producing this field is a saddle coil consisting of ten frames which have three windings each. The coil is fixed on the same PVC tube (see Fig. 4.2). This field is off during optical pumping and switched on for free precession. Then, the spins start precessing around the effective field $\vec{B}_{\text{eff}} = \vec{B}_0 + \vec{B}_1$, which is almost perpendicular to $\vec{B}_0$. The fields were scanned with a fluxgate magnetometer moved on a sledge through the shield. The magnetic field was measured every 10 cm in the $z$-direction and every 6 cm in the $x$- and the $y$-direction. Fig. 4.3, 4.4 and 4.5 show the magnetic field with all coils switched off, with only the $\vec{B}_0$ coil switched on, and with both coils on respectively. Using a second static field for spin precession was already tested in other setups [Chi90]. The important point is the non-adiabatic switch-on of the $\vec{B}_1$ coil. This is made sure by keeping the inductance of the $\vec{B}_1$ coil low enough. If the Larmor frequency is small enough compared to the relative field change perpendicular to the field $\vec{B}_0$, the spins start precessing around the $\vec{B}_{\text{eff}}$ field. This condition can be expressed by the following equation:

$$\omega_L \ll \frac{\partial B_1/\partial t}{B_1}$$

(4.1)

It can be shown that the inductance of the coil is small enough to switch on the field non-
Figure 4.3: The remanent field, when no field coil is on, is approx. 50 nT.

Figure 4.4: The $\vec{B}_0$ coil produces a field of 1 $\mu$T along the shield axis.
Figure 4.5: Switching on the $\vec{B}_1$ coil in $x$-direction in addition to the $\vec{B}_0$ coil produces an effective field $\vec{B}_{\text{eff}}$ that is almost perpendicular to the shield axis.

adiabatically. The time evolution of a current $I$ flowing through the coil obeys [Gre02]:

$$I = I_{\text{max}} \left(1 - e^{-t/(L/R)}\right)$$

(4.2)

where $I_{\text{max}}$ is the maximal current, $L$ is the inductance of the coil and $R$ its resistance. The ratio $L/R$ forms a characteristic time constant, that is a measure how fast the current, and thus the magnetic field, evolves in time. The resistance of the $\vec{B}_1$ coil is 4.0 $\Omega$. The inductance can be calculated by the following formula [Cla10].

$$L = \frac{\mu_0}{\pi} \left[ -l \log \left(1 + \sqrt{1 + \left(\frac{w}{l}\right)^2}\right) - w \log \left(1 + \sqrt{1 + \left(\frac{w}{l}\right)^2}\right) 
+ l \log \frac{2w}{w_r} + w \log \frac{2l}{w_r} + 2\sqrt{l^2 + \sqrt{2} - 2w - 2l}\right]$$

(4.3)

where $l$ is the length of a rectangular current loop, $w$ is the width of the loop and $w_r$ is the thickness of the wire. Putting in the values of the coil, $L = 102$ $\mu$H. For the right-hand side of Eq. 4.1, the values below can be applied.

$$\partial B_1 = 4 \, \mu T$$

(4.4)

$$\partial t = L/R = 2.6 \times 10^{-5} \, s$$

(4.5)

$$B_1 = 4 \, \mu T$$

(4.6)

This results in a value of $3.8 \times 10^4$ Hz for the right-hand side of Eqn. 4.1. The Larmor frequency at $4 \mu T$ is only $\omega_L = 2\pi \times 30 \, Hz = 188.5 \, Hz$. Thus, the inequality above is fulfilled.

**Homogeneity of the fields**

The relaxation of a polarised ensemble depends, among other parameters, on magnetic field gradients (see Sec. 3.1.2). Therefore, it is important to know the size of the gradients of the
holding field $\vec{B}_0$, in which the $^{199}$Hg atoms are pumped, and the precession field $\vec{B}_{\text{eff}}$. The formula for the longitudinal relaxation time $T_{1,\text{mag}}$ and the transverse relaxation time $T_{2,\text{mag}}$ for $\vec{B}_0$ can be taken from Sec. 3.1.2. The formula for $T_{1,\text{mag}}$ and $T_{2,\text{mag}}$ for $\vec{B}_{\text{eff}}$ have to be adapted, since the spins precess around the $x$-component of $\vec{B}_{\text{eff}}$:

$$\frac{1}{T_{1,\text{mag}}} = \frac{8R^4\gamma_{\text{Hg}}^2}{175D} (|\vec{\nabla} B_{\text{eff},y}|^2 + |\vec{\nabla} B_{\text{eff},z}|^2),$$

(4.7)

and

$$\frac{1}{T_{2,\text{mag}}} = \frac{4R^4\gamma_{\text{Hg}}^2}{175D} (|\vec{\nabla} B_{\text{eff},y}|^2 + |\vec{\nabla} B_{\text{eff},z}|^2 + 2|\vec{\nabla} B_{\text{eff},x}|^2),$$

(4.8)

where $R$ is the characteristic length of the cell and $D$ is the diffusion constant. For a cylindrical glass cell with a length of 8 cm and a diameter of 5 cm (see Sec. 4.1.4), the larger of the two dimensions is chosen for a conservative estimate. Therefore $R = 8$ cm as characteristic length.

The diffusion in the low pressure regime is described by $D = \pi \lambda^2/3$ with $\pi \approx 180$ m/s as the average velocity of $^{199}$Hg and $\lambda = 4V/S = 3.8$ cm as the mean free path of the $^{199}$Hg atoms in the glass cell. This results in

$$D = 2.3 \times 10^4 \text{ cm}^2/\text{s}.$$

(4.9)

The gradients can be extracted from the fieldmaps. For $\vec{B}_0$, gradients over the volume of the cell are $\partial B_x/\partial x \approx \partial B_y/\partial y \approx \partial B_z/\partial z \approx 3$ nT/cm and $\partial B_x/\partial z \approx \partial B_y/\partial z \approx 2$ nT/cm.

Using these values in Eqn. 4.7, a value for $T_{1,\text{mag}}$ can be calculated:

$$T_{1,\text{mag}} = 54'000 \text{ s}$$

(4.10)

Since there is no precession around $\vec{B}_0$, a value for $T_2$ is of no interest.

For $\vec{B}_{\text{eff}}$, typical gradients are $\partial B_x/\partial x \approx \partial B_y/\partial y \approx \partial B_z/\partial z \approx 8$ nT/cm, $\partial B_x/\partial z \approx \partial B_y/\partial z \approx 1$ nT/cm, $\partial B_x/\partial x \approx \partial B_y/\partial y \approx 5$ nT/cm, and $\partial B_x/\partial z \approx 0.5$ nT/cm. In Fig. 4.6, the profile of $B_{\text{eff},x}$ along the $x$-direction for different $z$ is shown as an example. With these values, the following $T_{1,\text{mag}}$ and $T_{2,\text{mag}}$ for $B_{\text{eff}}$ can be stated:

$$T_{1,\text{mag}} = 600 \text{ s},$$

(4.11)

and

$$T_{2,\text{mag}} = 200 \text{ s}.$$  

(4.12)

A comparison with $T_{1,\text{mag}}$ for the polarisation chamber of the nEDM-apparatus is made in the following. Assuming $R = 29$ cm as the characteristic length for the polarisation chamber, $D = 6.4$ cm$^2$/s as the diffusion constant, and $\partial B_z/\partial t = 100$ pT/cm over the volume of the chamber as typical gradients for every field component and every direction, we arrive at $T_{1,\text{mag}} \approx 8700$ s. However, measurements for $T_1$ show a value of 100 s. Therefore, $T_1$ must be dominated by wall collisions. From this, a rough estimate for $T_1$ for the glass cell can be made, since $T_1 \sim \lambda$.

The mean free path in the polarisation chamber can be estimated to be 6.4 cm. This results in $T_1 = 60$ s for the cell. For wall collisions, $T_1 = T_2$.

In order to keep this homogeneity, only strictly non-magnetic parts may be used inside the shield. Every single piece was tested, whether it is magnetic or not. These tests were done with a device which is able to detect magnetic impurities with a sensitivity of 30 pT at a distance of 5 cm. A sledge carries a sample piece through a magnetic shield with two Cs magnetometers in the middle. A magnetic sample will then be seen by the Cs magnetometers. With the help of this device, a highly magnetic steel spring (a few nT) was detected in the Teflon valves which disconnect the cell from the vacuum system. The spring has been replaced by a non-magnetic one of CuBe.
Figure 4.6: Magnetic field $B_x$ along the $x$-direction for different $z$-positions. The glass cell has a diameter of 5 cm and is located in the range from $x = -2.5$ cm to $x = 2.5$ cm.

4.1.2 Optical pumping in the setup

The pump light is the same as the readout light and is provided by a $^{204}$Hg-discharge lamp, the same system as in the nEDM-experiment. The lamp is mounted on a table adjustable in all direction for easier alignment. For pumping the $^{199}$Hg vapour and also for the readout of the signal, we need circularly polarised light, either $\sigma^+$ or $\sigma^-$ light. Fig. 4.7 shows the optical system of the setup. The light is first collimated by a lens (focal length 15 cm) and then polarised with a quartz thin film polariser. The incident light is reflected on the polariser under the Brewster angle. Under this angle, the reflected light is linearly polarised. In the specifications from Eksmaoptics, the Brewster angle for the thin film polariser is $55^\circ$ and the degree of linear polarisation is more...
than 99% [Eks11]. Therefore, I assume

\[ P_\pi = 99\%. \]  \hspace{1cm} (4.13)

A quarter-wave plate behind the thin film polariser turns the linearly polarised light into circularly polarised light, which is needed for optical pumping. Assuming that the quarter-wave plate can be adjusted to 1° to the polarisation plane of the linear polarised light, the amount of \( \sigma^+ \)-light is \( \Phi_{\sigma^+} = \Phi_\pi \sin(1^\circ) \) and the amount of \( \sigma^- \)-light is \( \Phi_{\sigma^-} = \Phi_\pi \cos(1^\circ) \). The circular polarisation of the light can be expressed as follows:

\[ P_{\sigma^+} = \frac{\Phi_{\sigma^+} - \Phi_{\sigma^-}}{\Phi_{\sigma^+} + \Phi_{\sigma^-}} = 97\%. \]  \hspace{1cm} (4.14)

Thus, the total polarisation of the light can be expressed as:

\[ P_l = P_\pi \times P_{\sigma^+} = 96\%. \]  \hspace{1cm} (4.15)

The polarisation optics have a diameter of 50 mm and thus are as large as the cell diameter to be able to illuminate the cell as much as possible. For stable positioning, the optical elements are fixed on a optical bench that is placed before the mu shield. The height of the light beam is defined by the position of the glass cell in the shield. Therefore, the optical elements are adjustable to this height. The light is detected by a photomultiplier from Hamamatsu (R431S) at the other end of the shield [Ham11]. The RMS noise in the signal is about 1–2 mV. Measurements for different PM-voltages showed that 400 V is a good value to work with, as the S/N ratio increases up to 400 V and then stays constant until at very high values, when S/N is reduced due to saturation.

4.1.3 Data Acquisition

For the data acquisition, a USB card from National Instruments is used [Ins11]. The card NI-USB 6211 is a 16-bit ADC with a sampling rate of 250 kS/s. For the readout of the PMT, the DC-level is sampled every millisecond and is written to a file. The card possesses also digital output channels which are used for driving the valves and analog output channels of which one is used for driving the \( \vec{B}_0 \) coil. The \( \vec{B}_1 \) coil is driven separately by a power supply of the type PL 303-P from TTI, since it was not possible for the card to deliver a current of 40 mA. The card and the power supply are controlled by a LabView application. The application is structured in cycles, which consist of five steps, which are explained in Fig. 4.8. The duration of every step can be changed as well as the values for the currents in the coils; normally the currents stay unchanged. The amount of mercury in the cell can be controlled by the opening time of the inlet valve. Once the program is started, the cycles run automatically, unless the program is stopped manually. In a second case, the numbers of cycles for one run can be entered in a window. When the number of cycles are accomplished, the program terminates automatically.
Figure 4.8: A cycle in the setup consists of five steps: (a) measurement of the noise of the light intensity level for 5 s, (b) filling the chamber with mercury (rapid change of DC-level from $-3.1$ V to $-2.4$ V), (c) polarisation of the mercury (slow decrease of the DC-level during 30 s), (d) precession of the signal during 60 s, (e) the mercury is released from the cell. A small amount of mercury stays in the cell after every cycle. This explains why the DC-level does not return to the initial value in this plot. However, the measurement is not influenced when this effect does not vary with time.
4.1.4 The $^{199}$Hg cell and the vacuum system

Mercury is filled into a glass cell for optical pumping. Contrary to other experiments [Chi90], [Rom03] the cell is not just filled with a defined quantity of mercury and then closed, it remains connected to the vacuum system. This allows us to pump and vent the cell and hence vary the density of $^{199}$Hg vapour as in the nEDM-experiment. It also allows one to easily connect and disconnect the cells.

An overview of the vacuum system with the glass cell is illustrated in Fig. 4.9. The long tubes with the cell can be disconnected from the vacuum system outside the shield to take out the cell. Before operation, the whole system is evacuated for two days. All the valves except V1 are open.

![Sketch of the vacuum system and the glass cell](image)

Figure 4.9: Sketch of the vacuum system and the glass cell: The mercury vapour is guided from the Hg source via the inlet valve to the glass cell in the shield. After the precession the mercury is released via the outlet tube to the pump. The two long aluminium tubes with the cell can be disconnected at the vacuum clamps at the end of the shield to take out the cell. The whole system can be evacuated to $(1-2) \times 10^{-5}$ mbar.

After two days, the pressure sensors G2 and G3 show a pressure of $(1-2) \times 10^{-5}$ mbar and G1 shows a value below $10^{-6}$ mbar.

During operation, the valve V2 is closed to prevent that all the mercury in the inlet tube is pumped away. Mercury is then delivered from the Hg source and is guided via the inlet tube to the glass cell. The inlet valve is typically opened for two seconds and the cell is filled with $^{199}$Hg vapour. After the precession period the outlet valve opens and releases the mercury from the cell. This takes about two to three minutes. Approximately 10% of the $^{199}$Hg vapour remains in the cell. The mercury which leaves the cell is trapped in a Hg filter system connected to the pump. Charcoal with sulphur additives capture the Hg atoms and prevent them from being released to the environment.

The standard cell is of cylindrical shape (see Fig. 4.10). It has a length of 8 cm and an inner diameter of 5 cm. It is made of borosilicate and was fabricated by a local glassblower [Tra11]. The windows at its ends are quartz plates since it has to be transparent to UV light. These plates have a thickness of 3 mm and a UV-transmission of roughly 90%. The quartz windows are pressed on the cell by a plastic frame that can be removed easily. The cell is sealed with o-rings. The connection pieces are 5 cm long and are connected to Teflon valves above the cell. Two Teflon valves connect the cell with two aluminium tubes (inlet, outlet tube) and with the rest of the vacuum system. The Teflon valves work with pressurised air which moves a piston back and forth.
and opens respectively closes the valve this way [SA11]. The valves are fixed on a sledge which is guided on rails through the mu metal shield. The rails are made of non-magnetic fibreglass plastic and have a length of 1.9 m. The whole setup inside the shield is shown in Fig. 4.11. The exact position of the cell is given by the length of the aluminium tubes that connect the vacuum system with the cell. The sledge we use is a 20 cm long and 10 cm wide breadboard of PVC with wheels. In a first version, the cell itself was fixed on the sledge as well as the Teflon valves. The result was that one of the connectors of the cell broke off due to the mechanical stress of the system. Now, only the valves are fixed whilst the cell hangs freely from the valves. It guarantees that no additional force acts on the glass.

**Pressure in the cell during operation**

In the following I try to give an estimate of the pressure in the cell during operation. The pressure sensor G3 shows values around $p_1 = 10^{-2}$ mbar in the volume of the source. This volume consists of a tube system with an inner diameter of 1 cm and a total length of 100 cm. This results in a volume of $V_1 = 79$ cm$^3$. The cell is pumped out for two to three minutes, therefore I assume a value below $p_2 = 10^{-4}$ mbar. The cell has a volume of $V_2 = 157$ cm$^3$. The aluminium tube, that connects the two volumes, has a length of $l = 90$ cm and an inner diameter of $d = 4$ mm. The conductance of the tube can be calculated as follows [Yos08]:

$$C = 12 \frac{d^3}{l} = 8.5 \times 10^{-3} \text{l/s},$$  \hspace{1cm} (4.16)

where $d$ and $l$ are given in cm and C in liter/s. The formula is only valid in the molecular flow regime.
Figure 4.11: The cell is connected to the Teflon valves which are fixed on a sledge. The sledge can be guided on rails through the mu shield. The dimensions of sledge, rails and cell are chosen in order that the cell is exactly on the axis of the mu cylinder. The cell hangs freely to avoid mechanical stress acting on the glass.
Opening now the inlet valve, the evolution of $p_1$ and $p_2$ can be described by the following differential equations:

$$V_1 \frac{dp_1}{dt} = -C(p_1 - p_2),$$  \hspace{1cm} (4.17)

and

$$V_2 \frac{dp_2}{dt} = C(p_1 - p_2).$$  \hspace{1cm} (4.18)

We define now $\Delta p = p_1 - p_2$. The difference of the two equations above results in:

$$\frac{\Delta p}{dt} = -C \left[ \frac{1}{V_1} + \frac{1}{V_2} \right] \Delta p.$$  \hspace{1cm} (4.19)

This describe the equilibration of the pressure difference. The result is an exponential decay:

$$\Delta p(t) = 10^{-2} \text{ mbar} \exp(-t/\tau_p)$$  \hspace{1cm} (4.20)

with

$$\tau_p = \left( C \left[ \frac{1}{V_1} + \frac{1}{V_2} \right] \right)^{-1} = 6 \text{ s}.$$  \hspace{1cm} (4.21)

For an opening time of $2 \text{ s}$, $\Delta p = 7.2 \times 10^{-3} \text{ mbar}$. The pressure in the cell is thus $p_2 = 1.5 \times 10^{-3} \text{ mbar}$. The equilibrium pressure can be calculated as follows:

$$p_1 V_1 + p_2 V_2 = p_\infty (V_1 + V_2).$$  \hspace{1cm} (4.22)

Since $p_2 \ll p_1$, $p_2$ is negligible and the equilibrium pressure is

$$p_\infty = p_1 \frac{V_1}{V_1 + V_2} = 3.3 \times 10^{-3} \text{ mbar}.$$  \hspace{1cm} (4.23)

A pressure of $1.5 \times 10^{-3} \text{ mbar}$ in the cell corresponds to a particle density of $n = 3.6 \times 10^{13} \text{ cm}^{-3}$. From this a mean free path in the gas can be estimated [Tip09]:

$$\lambda_{\text{gas}} = \frac{1}{\sqrt{2n\pi r^2}}$$  \hspace{1cm} (4.24)

where $n$ is the particle density of the gas and $r$ the radius of a gas molecule. The typical diameter of a two-atomic molecule is $d = 4 \times 10^{-10} \text{ m}$ [Wol93]. A value of $\lambda_{\text{gas}} = 16 \text{ cm}$ is obtained. Compared to the geometrical free path in the cell of $\lambda = 3.8 \text{ cm}$, collisions with residual gas are four times less frequent than wall collisions. Therefore, the wall collisions are dominating over residual gas collisions.

### 4.2 Investigations of wall coatings

An important depolarisation mechanism for $^{199}$Hg atoms are wall collisions. Dipolar interactions of paramagnetic sites on the surface cause $^{199}$Hg vapour to depolarise [Rom03]. For a certain material, there is a spin flip probability per collision. In order to keep this interaction small, the surface is coated with an unpolar material, which results in longer relaxation times. So far, Fomblin oil of type Y has been used as an antirelaxation coating for the polarisation chamber. This fluid is a perfluorinated polyether consists of only carbon, fluorine and oxygen atoms. It was originally used in vacuum pumps, since the C - F bond results in low vapour pressure. However, there are disadvantages as well. One problem with the Fomblin is its oily character. It runs down surfaces leaving uncoated regions after some time, which results in a decrease in relaxation...
<table>
<thead>
<tr>
<th>Coating</th>
<th>Chemical Structure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fomblin oil</td>
<td>$\text{CF}_3-[-\text{O}-\text{CF}_2-\text{CF}_2]_n - [\text{O}-\text{CF}_2]_m - \text{O}-\text{CF}_3$</td>
</tr>
<tr>
<td></td>
<td>$n/m = 20 \ldots 40$</td>
</tr>
<tr>
<td>Fomblin grease</td>
<td>$\text{CF}_3-[-\text{O}-\text{CF}_2-\text{CF}_2]_n - [\text{O}-\text{CF}_2]_m - \text{O}-\text{CF}_3$</td>
</tr>
<tr>
<td></td>
<td>mixed with $[\text{CF}_2-\text{CF}_2]_n$</td>
</tr>
<tr>
<td>Apiezon J oil</td>
<td>based on hydrocarbons</td>
</tr>
<tr>
<td>Paraffin</td>
<td>$\text{C}<em>{32}\text{H}</em>{66}$</td>
</tr>
<tr>
<td>Perfluorinated paraffin</td>
<td>$\text{C}<em>{20}\text{F}</em>{42}$ (80%-90%) + $\text{C}<em>{16}\text{F}</em>{34}$ (10%-20%)</td>
</tr>
<tr>
<td>AquaSil</td>
<td>$\text{CH}_3-(\text{CH}<em>2)</em>{15}-\text{Si}-(\text{OH})_3$</td>
</tr>
<tr>
<td>SurfaSil</td>
<td>$\text{Cl}-[-(\text{CH}<em>3)</em>{2}-\text{Si}-\text{O}-]_n-(\text{CH}<em>3)</em>{2}-\text{Si}-\text{Cl}$</td>
</tr>
<tr>
<td>Black teflon</td>
<td>$[\text{CF}_2-\text{CF}_2]_n$</td>
</tr>
<tr>
<td></td>
<td>mixed with carbon</td>
</tr>
</tbody>
</table>

Table 4.1: An overview of the anti-relaxation coatings for Hg and their chemical structure. One property, which all coatings have in common, is the carbon chain in the center.
times and finally in a lower polarisation of the $^{199}$Hg vapour. In longer measurement periods, recoating the chamber every few weeks is inevitable. Another point is the tedious and lengthy coating process, which lasts for several days. For the upper part of the polarisation chamber, Fomblin grease is used. This grease is an emulsion of Fomblin oil with Teflon powder. The grease, although more sticky than the oil, has the disadvantage that it cannot be distributed homogeneously on the surface of the chamber.

The search for a better coating is quite important, not only in terms of relaxation times but also in terms of stability and application. The materials listed in Tab. 4.1 were tested in the setup. Positive results have been reported by unpolar paraffin or paraffin-like coatings by [Rom03].

Paraffins are used in many other experiments dealing with polarisation of gases and vapours. In the HgEDM experiment in Seattle, dotriacontane which is also a paraffin, is used. Knowing that perfluorinated hydrocarbons like Teflon or Fomblin oil are good relaxation coatings, also perfluorinated paraffin is tested. AquaSil and SurfaSil are hydrocarbons containing silicon atoms and are good candidates as well [Chi90]. They can react directly with the glass and form hence a more stable compound. The ”J” oil from Apiezon is chosen, because it is a well-known vacuum oil that is based on hydrocarbons. The exact composition is not revealed by Apiezon. Finally, black Teflon is Teflon mixed with carbon to 25% to colour the surface black. This is done to avoid reflections of light on the walls. Reflections of light on the wall enhances depolarisation by reemitted light as we will see in the next section.

4.2.1 Measurement of $T_2$ and reflectivity of coatings

The relaxation time is measured with the “relaxation in the dark” method, which was performed by Franzen in 1959 for the first time [Fra59]. In this method, the atoms are first pumped to the maximum polarisation that can be obtained by the system. Then, the reading light is blocked for a certain time $t_{\text{dark}}$, in which the polarisation decays exponentially without being disturbed by optical pumping from the light. After $t_{\text{dark}}$, the polarisation is measured via the level of the reading light. The measurement is repeated for different $t_{\text{dark}}$ and $T_1$ can be extracted by a fit.

In the test setup, I measure the transverse relaxation time $T_2$ instead of the longitudinal relaxation time $T_1$. For the measurement of $T_1$, the light level has to be determined just after the light appears again. For a measurement of $T_2$, the amplitude of the precession signal has to be known, which is half the difference between the two envelope levels of the precession. Drifts of light level are therefore cancelled and $T_2$ can be measured more accurately than $T_1$. The amplitude of the signal is determined for different dark times. $T_2$ is finally determined by a fit (see Fig. 4.12).

Important is, that the lamp is in stable operation to reduce drifts in the light and that the whole measurement is performed with the same absorption, since the polarisation depends on the absorption. An example of a measurement is shown in Fig. 4.13. Since one is interested in the quality of the coatings, $T_2$ has to be dominated by wall collisions. The estimates of Sec. 4.1.4 and Sec. 4.1.1 show that this is the case.

The best would be to always use the same mercury and never open the valves during the measurement once a certain amount of mercury is in the cell. The problem then is that the pressure rises due to outgassing and small leaks, since the cell is not pumped for approximately one hour. If the pressure rises too fast, collisions with the residual gas dominate over wall collisions and may distort the measurement.

For each coating, $T_2$ has been measured twice with a time in between of at least two weeks. From the two measurements, one can also estimate the stability of coatings in time. The results are shown in Tab. 4.2. The best $T_2$ was measured with perfluorinated paraffin from Maflon [Maf10], actually a skiing wax. The J oil of Apiezon was found to be highly magnetic. Therefore
Figure 4.12: The amplitude of the signal as a function of the dark time $t_{\text{dark}}$. Fitting the data with an exponential gives $T_2$.

Figure 4.13: The plot shows the cycles of a $T_2$-measurement. The dark time is varied between 0 s and 60 s. The different dark times are in random order to cancel possible drifts. Important is, that the measurement is performed always at the same absorption and with a constant reading light to guarantee a stable initial polarisation during the whole measurement.
Table 4.2: The tables shows the measured $T_2$-times for the coatings listed in Tab. 4.1. A second measurement has been done for a better comparison and for an estimate of stability.

<table>
<thead>
<tr>
<th>coating</th>
<th>measurement 1 [s]</th>
<th>measurement 2 [s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fomblin oil</td>
<td>28.4 ± 0.5</td>
<td>27.0 ± 0.6</td>
</tr>
<tr>
<td>Fomblin grease</td>
<td>34.7 ± 0.7</td>
<td>30.6 ± 0.5</td>
</tr>
<tr>
<td>Apiezon J oil</td>
<td>no signal</td>
<td>-</td>
</tr>
<tr>
<td>paraffin</td>
<td>26.3 ± 0.4</td>
<td>27.8 ± 2.1</td>
</tr>
<tr>
<td>perfluorinated paraffin</td>
<td>40.5 ± 1.2</td>
<td>34.6 ± 0.6</td>
</tr>
<tr>
<td>AquaSil</td>
<td>22.8 ± 1.0</td>
<td>26.0 ± 0.4</td>
</tr>
<tr>
<td>SurfaSil</td>
<td>32.2 ± 0.4</td>
<td>30.6 ± 0.7</td>
</tr>
<tr>
<td>black teflon</td>
<td>&lt; 5</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 4.3: The table shows the measured reflectivity and transmission for each coating at a wavelength $\lambda = 254$ nm. The substrates were 3 mm thick quartz plates. (Actually the windows for the glass cell.) A thickness for each coating is estimated in the text.

<table>
<thead>
<tr>
<th>material</th>
<th>transmission [%]</th>
<th>reflectivity [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>blank quartz</td>
<td>91.9</td>
<td>3.4</td>
</tr>
<tr>
<td>Fomblin oil</td>
<td>92.6</td>
<td>3.1</td>
</tr>
<tr>
<td>Fomblin grease</td>
<td>78.4</td>
<td>5.3</td>
</tr>
<tr>
<td>Apiezon J oil</td>
<td>14.7</td>
<td>4.6</td>
</tr>
<tr>
<td>paraffin</td>
<td>59.2</td>
<td>5.9</td>
</tr>
<tr>
<td>perfluorinated paraffin</td>
<td>69.5</td>
<td>7.0</td>
</tr>
<tr>
<td>AquaSil</td>
<td>91.4</td>
<td>3.6</td>
</tr>
<tr>
<td>SurfaSil</td>
<td>91.1</td>
<td>3.7</td>
</tr>
<tr>
<td>black teflon</td>
<td>-</td>
<td>10.1</td>
</tr>
</tbody>
</table>

no signal was seen. Also black Teflon showed a very short $T_2$ of less than 5 s. A possible reason might be that there are “contaminants” in the material which causes the $^{199}$Hg atoms to depolarise much faster. However, the black teflon piece was found to be non-magnetic.

4.2.2 Transmission and reflectivity of UV light

Important properties of wall coatings are also the transmission and the reflectivity of UV light. The pump light has to pass twice a quartz plate with an thin layer of coating on it. It is obvious that a high transmission value results in stronger pumping, since more light enters the cell. Even more important is the reflectivity of the coated surface. In chapter 3, we have seen that a part of the reemitted light depolarises the $^{199}$Hg vapour. The transmission and the reflectivity of the coated quartz windows have been measured in a wavelength range from 200 nm to 400 nm (see Tab. 4.3). The measurements with the UV spectrometer is described in appendix C. The reflectivity depends slightly on the wavelength. This is shown in Fig. 4.14.

Knowing the reflectivity $R$ of the surface, it is possible to calculate the enhanced light flux due to the reflections on the surface. Assuming an initial light flux of $\Phi_0$ coming from reemission, the reflected light flux $\Phi_1$ can then be expressed as

$$\Phi_1 = R\Phi_0 \exp(-\zeta \ldots),$$

(4.25)
where $\zeta$ is the mean free path of the light in the cell, and $\zeta_{\text{gas}}$ is the mean free path of the light in the $^{199}\text{Hg}$ vapour. The reflected light can be reflected again and so on. The light flux after the $i$th reflection can be expressed as

$$\Phi_i = R \Phi_{i-1} \exp(-\zeta \ldots). \quad (4.26)$$

The total light flux is then given by the sum of the partial fluxes after each reflection:

$$\Phi_{\text{tot}} = \Phi_0 (1 + R \exp(-\zeta \ldots) + (R \exp(-\zeta \ldots))^2 + \ldots). \quad (4.27)$$

The infinite sum can be rewritten as:

$$\Phi_{\text{tot}} = \frac{1}{1 - R \exp(-\zeta \ldots)} \Phi_0. \quad (4.28)$$

The largest value for $R$ was 10.1% for black Teflon. From this, a maximal value for $\Phi_{\text{tot}} \leq 1.1 \Phi_0$ can be estimated. The change of the reemission terms in Sec. 3.1.3 are thus within 10%.  

The transmission and the reflectivity are a function of the thickness of the coating. In the following, I give estimates on the thicknesses for each coating. Concerning the Fomblin oil and the Fomblin grease, the density is $\rho = 2.0 \text{g/cm}^3$ [Fom12]. I was able to distribute an amount of $m = 20 \text{mg}$ of the oil or the grease on a quartz plate having a diameter of $d = 5 \text{cm}$ diameter. The thickness is then

$$\Delta l = \frac{m/\rho}{\pi(d/2)^2} = 5 \mu\text{m}. \quad (4.29)$$

No density could be found for the "J" oil of Apiezon, but since it can be smeared on the surface as the Fomblin oil and the Fomblin grease, the thickness of the layer can be estimated to be roughly the same. SurfaSil and AquaSil form one layer of molecules. The thickness is then smaller than
1 nm. An estimate of the thickness for the paraffins is hardly possible, since only a part of the paraffin used in the coating process is deposited on the surface of the glass cell. The rest is pumped away. However, since maximal 10 mg is used, the thickness of the layer must be thinner than for Fomblin oil.

4.3 Depolarisation due to reemission

A second depolarisation mechanism is the absorption of reemitted light. A part of the reemitted light destroys the polarisation again. This effect depends crucially on the density of the $^{199}\text{Hg}$ vapour. Therefore, lowering the density by increasing the cell volume might result in a higher polarisation.

For the following tests, different cells are used which have the same diameter but have different lengths. The lengths of the four cells are 5 cm, 8 cm, 12 cm, and 16 cm. For each cell, the precession amplitude is measured for different absorption values. The light level $I_1$ enters the amplitude of the signal twice, once as light flux in optical pumping and once in the readout of the signal. To remove the dependence of $I_1$ on the readout, the amplitude is divided by $I_1$. The influence on optical pumping still remains. Therefore, it is important to have a stable $I_1$ in order to keep the power of optical pumping the same. The result is shown in Fig. 4.15. The longer cells show higher values of polarisation. The polarisation can be calculated using Eqn. 2.25 in Sec. 2.4.2:

$$P = -\frac{\log (1 + \frac{a}{I_1(1 - A)})}{\log(1 - A)}. \quad (4.30)$$

The polarisation is shown in Fig. 4.16 as a function of the absorption. For a certain absorption and thus for a certain amount of mercury, the polarisation increases for longer cells. The effect
doubles from the shortest cell to the longest cell. Finally, the polarisation can be plotted as a

Figure 4.16: Polarisation as a function of absorption for different cell lengths. One clearly sees that the degree of polarisation increases with the cell length for a certain absorption.

function of the $^{199}$Hg density (see Fig. 4.17). The polarisation curves for the different cell lengths fall close together for small density values (smaller than $1.5 \times 10^{11}$ atoms/cm$^3$). The influence of the wall relaxation must be small, since it would have been observed as higher polarisation for longer cells for a certain density. But this is not the case. For larger densities, the polarisation is even smaller than for longer cells. This might come from the longer path of the depolarising light in the cell.

For each cell, $T_2$ has been measured in a separate measurement. The important quantities for each cell are given in Tab. 4.4.

<table>
<thead>
<tr>
<th>length [cm]</th>
<th>volume [cm$^3$]</th>
<th>mean free path $\lambda$ [cm]</th>
<th>$T_2$ [s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>98</td>
<td>3.3</td>
<td>20.5</td>
</tr>
<tr>
<td>8</td>
<td>157</td>
<td>3.8</td>
<td>28.4</td>
</tr>
<tr>
<td>12</td>
<td>236</td>
<td>4.1</td>
<td>37.2</td>
</tr>
<tr>
<td>16</td>
<td>314</td>
<td>4.3</td>
<td>43.8</td>
</tr>
</tbody>
</table>

Table 4.4: Characteristic values for each cell. For the mean free path, the formula $\lambda = 4V/S$ was used. All $T_2$-times were measured with Fomblin oil as coating and at an absorption of 30%.

Comparison of the data with the model

The polarisation curves for the different cell lengths were cross-checked with the model in chapter 3. The values for the parameters were taken from Tab. 4.4. The light flux was regarded as constant for all cells, $\Phi = 4.5 \times 10^{12}$ s$^{-1}$. The result of the calculation is shown in Fig. 4.18. With
Figure 4.17: Polarisation as a function of the $^{199}\text{Hg}$ density for the different cell lengths. The polarisation curves for the different cell lengths fall close together for small density values (smaller than $1.5 \times 10^{11}$ atoms/cm$^3$). For larger densities, the longer cells show smaller polarisation values. A possible explanation might be that depolarising light has a longer path in the cell.

Figure 4.18: Calculated polarisation for the different cells. The calculations were done with the model described in chapter 3. The parameters used in were taken from Tab. 4.4.
longer cells, higher polarisation curves are calculated. However, the curves are less steep than the measured ones. The effect of the reemission, that the polarisation decreases with the absorption, is not observed. The reason for the decrease with higher absorption is that the efficiency of optical pumping is reduced with higher mercury densities, since the flux of the pump light decreases over the length of the cell. The following possibilities were investigated to see whether they can explain the discrepancy between data and model:

- **Change of light absorption cross-section:**
  A change of the light absorption cross-section only increases or decreases the efficiency of optical pumping. The curves are shifted vertically. A density-effect does not appear.

- **Different cross-section for $\sigma^+$, $\pi$, and $\sigma^-$-light:**
  Increasing the cross-sections for $\pi$- and $\sigma^-$-light affects the steepness only little. Having cross-sections for $\sigma^-$- and $\pi$-light ten times larger than the cross-sections for $\sigma^+$-light, the polarisation decreases for absorption values below 10% and stays constant for larger absorptions.

- **Depolarisation due to collisions with other $^{199}$Hg atoms:**
  Another possibility is that the mercury depolarises due to collisions with other $^{199}$Hg atoms. In the following, I estimate the mean free path of a $^{199}$Hg atom in the $^{199}$Hg vapour.

  \[
  \lambda_{Hg} = \frac{1}{\sqrt{2n\pi(d/2)^2}},
  \tag{4.31}
  \]

  where \( n = 4 \times 10^{11} \text{ cm}^{-3} \) for the $^{199}$Hg density and \( d = 3 \times 10^{-10} \text{ m} \). The mean free path in the vapour is then \( \lambda_{Hg} = 6.3 \text{ m} \), which is much larger than the mean free path in the residual gas. Depolarisation due to collisions with other $^{199}$Hg atoms can thus be excluded.

Finally, I can say that increasing the cell volume helps to increase the polarisation of the $^{199}$Hg vapour. A next possibility which I did not try is to go to reemission of even higher order. However, higher order effects are smaller and smaller and would at some point be negligible. It is not clear how much the reemission effects contribute to the decrease of polarisation.
5 Systematics correlated to the $^{199}$Hg magnetometer

For the measurement of the nEDM, the control of systematic effects on the same level as the statistical sensitivity is necessary. In phase II, an overall systematic error of $1.3 \times 10^{-27} e\,\text{cm}$ is aimed at, which would allow us to measure the nEDM with an overall sensitivity of $5 \times 10^{-27} e\,\text{cm}$. A list of all the systematic errors is shown in Tab. 5.1.

The first three effects are related to the mercury geometric phases. The geometric phase effect is the shift of the precession frequency when a magnetic field gradient and an electric field are present. The frequency shift depends on the electric field and thus generates a false EDM. The effect occurs for both mercury and UCN, but for UCN the effect is negligible (see Sec. 5.1.2). The direct false EDM due to the mercury geometric phase does not appear in the list of systematic effects, since a sophisticated extraction of the nEDM from the data removes this effect (see Sec. 5.1.4). However, we will see, that local dipole fields and transverse fields do not allow a complete removal of the geometric phase effect.

Additional systematic effects related to the $^{199}$Hg magnetometer are those due to light shifts. The precession frequency is shifted due to interactions of the $^{204}$Hg probe light with the precessing $^{199}$Hg atoms. Even more, a light shift correlated to the electric field would result in a false EDM signal. This light shift is referred to as direct light shift.

In this chapter, a measurement of the geometric phase effect of $^{199}$Hg is described and corrections for the related systematics are given. From the same data, an upper limit on the false EDM coming from direct light shifts is calculated.

5.1 Geometric phase effect

5.1.1 Introduction

Geometric phases were thoroughly discussed for the first time in 1984 by M. V. Berry and thus are sometimes also referred to Berry’s phases [Ber84]. They are a quantum mechanical phenomenon, that manifests itself in measurable quantities. A brief glimpse of the origin of these phases is presented here [Ber84]. Assume a quantum system with a Hamiltonian $\mathcal{H}[\mathbf{R}(t)]$ and a wavefunction $\psi$, that describes the system. $\mathbf{R}$ is a set of parameters changing slowly and continuously with time. The whole system is now moved along a closed circuit called $C$. Therefore, $\mathbf{R}(t)$ starts at a certain $\mathbf{R}_0$ and returns to $\mathbf{R}_0$ after a time $T$. The time evolution of the wavefunction $\psi$ is given by the Schrödinger equation:

$$\mathcal{H}(\mathbf{R}(t))|\psi(t)\rangle = i\hbar \frac{d}{dt}|\psi(t)\rangle. \quad (5.1)$$

Additionally one can develop the wavefunction $\psi$ in a basis of eigenstates as follows:

$$\mathcal{H}(\mathbf{R}(t))|n(\mathbf{R}(t))\rangle = E_n(\mathbf{R}(t))|n(\mathbf{R}(t))\rangle, \quad (5.2)$$
Table 5.1: List of known systematic effects appearing in the nEDM-experiment. It shows the shift in terms of $d_{\text{false}}$ and its error estimated by the RAL-Sussex-ILL collaboration [Bak06]. The errors in the last column are estimated for phase II by our collaboration [Kne09].

where $|n(R(t))\rangle$ are the eigenstates and $E_n(R(t))$ are the eigenenergies at time $t$. Substituting 5.2 into 5.1, we arrive at

$$|\psi(t)\rangle = \exp\left(-\frac{i}{\hbar} \int_0^t dt' E_n(R(t'))\right) \exp(i\gamma_n(t)) |n(R(t))\rangle,$$  

(5.3)

where the first exponential term is the well-known time evolution factor and the second term is the one containing the geometric phase $\gamma_n(t)$. When the system is transported over the whole circuit we arrive at

$$|\psi(T)\rangle = \exp\left(-\frac{i}{\hbar} \int_0^T dt E_n(R(t))\right) \exp(i\gamma_n(C)) |\psi(0)\rangle,$$  

(5.4)

where $\gamma_n(C)$ is the Berry phase depending on the circuit $C$.

### 5.1.2 Geometric phase of $^{199}$Hg in the nEDM experiment

In the nEDM experiment, the magnetic field is not completely homogeneous. Field gradients are present at any point of the chamber. Due to the gradients and a superimposed electric field, the precessing particles acquire a phase, which results in a shift of the precession frequency. A detailed discussion of geometric phases of particles in traps is given in [Pen04] using the Bloch equations.

When a particle is precessing in a constant magnetic field $B_{0z}$ and in addition, a magnetic field $B_{xy}$ in the $xy$-plane is rotating at angular speed $\omega_r$, the precession frequency is shifted away from the Larmor frequency as follows:

$$\Delta \omega = \frac{\omega_{2y}^2}{2(\omega_0 - \omega_r)},$$  

(5.5)
where $\omega_0 = -\gamma B_{0z}$, $\omega_{xy} = -\gamma B_{xy}$, and $\omega_r = v_{xy}/R$ with $v_{xy}$ as the particle velocity in the $xy$-plane and $R$ as the trap radius. There are two magnetic fields pointing in the $xy$-plane. The first one stems from magnetic inhomogeneities. Vertical gradients produce a remaining field in the $xy$-plane due to the Maxwell equation $\nabla B = 0$. The equation in cylindrical coordinates is

$$\frac{1}{r} \frac{\partial}{\partial r} (r B_r) + \frac{1}{r} \frac{\partial B_\phi}{\partial \phi} + \frac{\partial B_z}{\partial z} = 0.$$  \hspace{1cm} (5.6)

Since we have cylindrical symmetry, we end up with

$$\frac{\partial}{\partial r} (r B_r) = -r \frac{\partial B_z}{\partial z}. \hspace{1cm} (5.7)$$

Assuming now a constant gradient in $z$-direction, a simple expression can be found for the field in the $xy$-plane. Integration over $r$ results in the radial component

$$B_r = -\left( \frac{\partial B_{0z}}{\partial z} \right) \frac{r}{2}. \hspace{1cm} (5.8)$$

We can write this in vectorial notation using $B_{0xy}$ as the radial field:

$$B_{0xy} = -\left( \frac{\partial B_{0z}}{\partial z} \right) \frac{r}{2}. \hspace{1cm} (5.9)$$

Additionally to the static fields, there occur motional magnetic fields. The particles moving with a velocity $v$ experience an additional magnetic field in their rest frame when an electric field $E$ is present:

$$B_v = \frac{v \times E}{c^2}. \hspace{1cm} (5.10)$$

Substituting now Eqn. 5.9 and Eqn. 5.10 in Eqn. 5.5, we can express the frequency shift in terms of the electric field $E$:

$$\Delta \omega = \gamma^2 \left( -\frac{r}{2} \frac{\partial B_{0z}}{\partial z} + \frac{v \times E}{c^2} \right) \frac{2(\omega_0 - \omega_r)}{\omega_0^2 - \omega_r^2} = c_0 + c_1 E + c_2 E^2, \hspace{1cm} (5.11)$$

where $c_0$, $c_1$ and $c_2$ are the corresponding coefficients. While the $E$-even terms do not change under HV polarity change, the $E$-odd term changes the sign and thus gives rise to a false EDM signal. This term results in a frequency shift between $B_0$ and $E$ parallel (denoted as ↑↑) and antiparallel (denoted as ↑↓). Taking into account all the possible orbits and both senses of circulation, the frequency difference is then

$$\Delta \omega_{↑↑} - \Delta \omega_{↑↓} = -2\gamma^2 \left( \frac{R \partial B_{0z}}{2} \frac{B_{0z}}{\partial z} \right) \frac{v_{xy} E}{c^2} \frac{1}{\omega_0^2 - \omega_r^2} \frac{|\omega_r|}{\omega_0^2 - \omega_r^2}. \hspace{1cm} (5.12)$$

The two spin species in the experiment behave differently. Whilst the spins of UCN follow the magnetic field lines, mercury atoms average over the $z$-component of the magnetic field. The UCN show values of $\omega_r/\omega_0 \ll 1$. This regime is called the adiabatic regime. The expression above can then be simplified to

$$\Delta \omega_{↑↑} - \Delta \omega_{↑↓} = -\frac{\partial B_{0z}/\partial z}{B_{0z}^2} \frac{v_{xy}^2 E}{c^2} \frac{1}{\omega_0^2 - \omega_r^2} \frac{|\omega_r|}{\omega_0^2 - \omega_r^2}. \hspace{1cm} (5.13)$$

This corresponds to a false EDM of

$$d_{f,UCN} = -\frac{\hbar}{4} \left( \frac{\partial B_{0z}/\partial z}{B_{0z}^2} \frac{v_{xy}^2 E}{c^2} \frac{1}{\omega_0^2 - \omega_r^2} \frac{|\omega_r|}{\omega_0^2 - \omega_r^2} \right). \hspace{1cm} (5.14)$$
Assuming values of $\partial B_0 z / \partial z = 10 \text{ pT/cm}$, $B_0 z = 1 \mu T$, and $v_{xy} = 2.4 \text{ m/s}$, the resulting false EDM amounts to $d_{f,UCN} = -1.05 \times 10^{-27} \text{ e cm}$.

The co-habiting $^{199}\text{Hg}$ atoms are situated in the near nonadiabatic regime with $\omega_r / \omega_0 \geq 1$. The formula for the $^{199}\text{Hg}$ frequency shift therefore slightly differs compared to that for UCN:

$$\Delta \omega_{\uparrow\uparrow} - \Delta \omega_{\uparrow\downarrow} = \frac{\gamma_{^{199}\text{Hg}} E}{c^2} R^2 \left( \frac{\partial B_0 z}{\partial z} \right) \left[ 1 - \frac{\omega_r^2}{\omega_0^2} \right]^{-1}. \quad (5.15)$$

This shift can be expressed as a false EDM

$$d_{f, \text{Hg}} = \frac{\hbar}{8} \left( \frac{\partial B_0 z}{\partial z} \right) \frac{\gamma_{^{199}\text{Hg}}^2 R^2}{c^2} \left[ 1 - \frac{\omega_r^2}{\omega_0^2} \right]^{-1}. \quad (5.16)$$

It is remarkable that the false EDM for $^{199}\text{Hg}$ is independent of the velocity of the atoms, but instead shows a dependence on the trap radius $R$. Assuming again $\partial B_0 z / \partial z = 10 \text{ pT/cm}$, $B_0 z = 1 \mu T$, and $R = 23.5 \text{ cm}$, the resulting false EDM is $1.16 \times 10^{-26} \text{ e cm}$. The effect for $^{199}\text{Hg}$ atoms is one order of magnitude larger than for UCN. A false $^{199}\text{Hg}$ EDM is transferred to the nEDM, since the UCN frequency is corrected for magnetic field drifts with the $^{199}\text{Hg}$ frequency (see Sec. 5.1.4).

However, only uniform vertical gradients have been considered so far. A different approach considers $B_{bxy}$ and $B_v$ as arbitrary weak fields in the $xy$-plane. A frequency shift can then be derived using correlation functions [Lam05], [Bar06]. Based on this, an expression for the geometric phase false EDM for particles in the nonadiabatic regime has been stated recently [Pig12]:

$$d_{f, \text{Hg}} = -\frac{\hbar \gamma_{^{199}\text{Hg}}^2}{2c^2} \langle x B_x + y B_y \rangle. \quad (5.17)$$

where $B_x$ and $B_y$ are the fluctuating fields and the brackets denote the average over the precession chamber. Assuming cylindrical symmetry, the fields are

$$B_x = -\frac{x \partial B_0 z}{2 \partial z} \quad (5.18)$$

and

$$B_y = -\frac{y \partial B_0 z}{2 \partial z}. \quad (5.19)$$

Substituting the expressions above in Eqn. 5.17, the result is the same as the solution obtained from the Bloch equations. The approach with the transverse fields is more general and holds for any kind of inhomogeneities.

5.1.3 Measurement and data taking

The shift in the $^{199}\text{Hg}$ precession frequency due to the geometric phase effect can be measured. The frequency shift and thus the corresponding false EDM can be determined as a function of the applied vertical magnetic field gradient and the electric field strength. A correction for systematic effects related to the geometric phase can be derived.

For one run (several 100 cycles) of the measurement, a certain $B_0$-field configuration was set and a certain gradient was applied. Then, the difference between the precession frequencies for an electric field parallel and anti-parallel to $B_0$ was measured. The applied high voltage (HV) for both polarities was 100 kV and the polarity was changed every 20 cycles. An important point is the sudden decrease of $T_2$ after a polarity change of HV and the slow recovery afterwards (see [Gre98]). A more frequent change would have decreased $T_2$ too fast and long runs would not have
been possible. The field fluctuations from one cycle to the next were about 3 pT, the sensitivity of the magnetometer over a cycle was about 200 fT. Since the field fluctuations are the limiting factor, an increase in statistics reduces the total error. Therefore, one is interested in as many data points as possible. For this reason, the free precession time was chosen to be only 40 s, the cycle length was 96 s.

The magnetometer ran for two and a half weeks for the measurement of the geometric phase in April/May 2011. There were two weeks of preparation and optimisation before, and about three weeks after the measurement, when tests with buffer gas were performed. During the entire two months, the magnetometer ran smoothly. Towards the end of this mercury campaign, $T_2$-values of more than 200 s were reached (see Fig. 5.1).

In order to produce vertical gradients, the trimcoils TTC (top transversal coil) and BTC (bottom transversal coil) were used in anti-Helmholtz configuration [Que09](see Fig. 5.2). The gradients have to be large enough to make the effect visible. A simple estimate shows that a gradient of 4 nT/cm produces a shift of $\sim$ 3 pT. Therefore, gradients were chosen in the range 0 . . . 4 nT/cm. In order to determine the gradients, Cs-magnetometers were placed above and below the precession chamber as shown in Fig. 5.2 and Fig. 5.3, four on the top electrode and eight below the bottom electrode. Already at the beginning of the measurement campaign, Cs 4, Cs 13, and Cs 18 did not work properly and no reasonable data were recorded. However, the remaining Cs magnetometers worked fine during the whole measurement period.
Figure 5.2: The main parts of the apparatus used for the measurement of the Hg geometric phase. The vertical gradients are produced with the trim coils. Cs-magnetometers on the top electrode and below the bottom electrode measure the local magnetic fields, which finally allows a determination of field gradients. For the geometric phase measurement, a small polariser cube with integrated quarter waveplate is put on the lower end of the Hg polarisation system instead of the elsewise used polarisation optics (see also Fig. 2.9).
5.1.4 Results and discussion

Finally, one is interested in the Hg frequencies during one run, that includes 20 to 30 polarity changes of the HV. A clear correlation is shown between the shifts in the Hg frequency and the HV polarity in Fig. 5.4. The time series is subdivided into parts where the drifts are approximately linear. The minimal length of such a subpart was chosen to contain 4 HV polarity changes. In each subpart, the data are fitted with a linear regression (see Fig. 5.5). The residuals of the fit are the shift in frequency due to the geometric phase. Then, the residuals of one run are divided into two groups for positive HV and negative HV resulting in a distribution for each polarity, which gives a mean value and an error for both groups. Finally, the two frequency shifts for positive and negative HV can be transformed to a false EDM for every applied gradient and for both $B_0$-directions.

Two methods are used to calculate the average gradient over the chamber as a function of the current in TTC, where the same current but of opposite sign is present in BTC.

- The average of the three magnetometers Cs 2, Cs 3, and Cs 5 on top and the average of five magnetometers Cs 12, Cs 15, Cs 17, Cs 19, and Cs 21 below are calculated. The difference of the upper and lower average is then divided by the vertical distance between the upper and lower Cs magnetometers.

- Another way to determine the average gradient is to use the field maps of TTC and BTC. The field of every trim coil was previously mapped for a standard current of 25 mA. From these maps, a determination of the magnetic field at any point inside the chamber is possible. A superposition of the TTC-field, the BTC-field, and the $B_0$-field is then easily provided.
Figure 5.4: The time series of the Hg frequency with a magnetic gradient of 3.5 nT/cm and an electric field of 8.3 kV/cm (run 1973, 5th of May 2011). The frequency is clearly correlated to the polarity of the HV.

Figure 5.5: Linear fitting of parts of the Hg frequency time series removes drifts. The residuals are used to calculate the mean values and the errors of the frequency shift. The black lines show the drift over the whole run.
<table>
<thead>
<tr>
<th>Method</th>
<th>Function</th>
<th>Field</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs magnetometers</td>
<td>$G = 158 \frac{\text{pT}}{\text{mA} \cdot \text{cm}} \times I + 217 \frac{\text{pT}}{\text{cm}}$</td>
<td>$B_0 \uparrow$</td>
</tr>
<tr>
<td>3 top and 5 bottom</td>
<td>$G = -159 \frac{\text{pT}}{\text{mA} \cdot \text{cm}} \times I - 4 \frac{\text{pT}}{\text{cm}}$</td>
<td>$B_0 \downarrow$</td>
</tr>
<tr>
<td>Field maps</td>
<td>$G = 127 \frac{\text{pT}}{\text{mA} \cdot \text{cm}} \times I + 109 \frac{\text{pT}}{\text{cm}}$</td>
<td>$B_0 \uparrow$</td>
</tr>
<tr>
<td></td>
<td>$G = -127 \frac{\text{pT}}{\text{mA} \cdot \text{cm}} \times I - 267 \frac{\text{pT}}{\text{cm}}$</td>
<td>$B_0 \downarrow$</td>
</tr>
<tr>
<td>Cs magnetometers (cross check)</td>
<td>$G = 159 \frac{\text{pT}}{\text{mA} \cdot \text{cm}} \times I + 281 \frac{\text{pT}}{\text{cm}}$</td>
<td>$B_0 \uparrow$</td>
</tr>
<tr>
<td>2 top and 2 bottom</td>
<td>$G = -160 \frac{\text{pT}}{\text{mA} \cdot \text{cm}} \times I + 73 \frac{\text{pT}}{\text{cm}}$</td>
<td>$B_0 \downarrow$</td>
</tr>
</tbody>
</table>

Table 5.2: This table shows the gradient $G$ as a function of the current $I$ in TTC. The same current, but of opposite sign, is applied to BTC. The values obtained from Cs magnetometers and from fluxgate field maps differ by 25%.

Since the field depends linearly on the current in the coils, the field and thus also the average gradient of every magnetic configuration is computable.

The dependence of the gradients on the current in TTC is shown in Fig. 5.6. The results are summarised in Tab. 5.2. The two methods show different results. A possible discrepancy might be that the centre of mass of the three upper Cs-magnetometers is not entirely above the five lower ones, which would result in a gradient, which is not entirely vertical. There is a way to check this: The Cs 5 and Cs 2 are completely above Cs 17 and Cs 12, which allows us to calculate an entirely vertical gradient, which is located in the middle of the precession chamber. The cross check is in good agreement with the first method. An additional check shows that the field map is not in good agreement with the field values measured by the Cs magnetometers. The differences are up to a few nT.

The time stability of the gradients is of interest too. Over several 100 cycles, a typical stability is $10 - 20 \text{pT/cm}$. The typical error of a cycle is $50 - 100 \text{pT/cm}$. Therefore, the drifts of gradients are negligible for the average gradient of one run. The stability of the gradient of run 1950 for 620 cycles is shown in Fig. 5.7. The distribution of gradients is in good approximation gaussian (see Fig. 5.8).

Finally, one is interested in the false EDM related to the applied gradient. The measured values of the false EDM are plotted in Fig. 5.9 as a function of gradients obtained from the Cs magnetometers. Fitting the data points to a straight-line going through the origin, we obtain

$$d_{f,Hg}/(\partial B_{0z}/\partial z) = (9.94 \pm 0.25) \times 10^{-28} \text{e cm}/(\text{pT/cm})$$ (5.20)

for $B_0$ down and

$$d_{f,Hg}/(\partial B_{0z}/\partial z) = (-9.69 \pm 0.33) \times 10^{-28} \text{e cm}/(\text{pT/cm})$$ (5.21)

for $B_0$ up. An average is calculated from the absolute values above as follows:

$$d_{f,Hg}/(\partial B_{0z}/\partial z)_{av} = (9.82 \pm 0.29) \times 10^{-28} \text{e cm}/(\text{pT/cm})$$ (5.22)

Additionally, the dependence of the false EDM vs gradient is also plotted in Fig. 5.10 using the gradients obtained from the field maps. The fitted model is still the same. The corresponding
Figure 5.6: The plot shows the dependence of the gradients on the applied current in TTC. The same current, but of opposite sign, is applied to BTC. Gradients have been determined by a fit from the data points with the Cs-magnetometers (solid line) as well as calculated from the field maps (dash-dotted line). The data from the cross-check (2 Cs top, 2 Cs bottom) are in good agreement with the first method (3 Cs top, 5 Cs bottom). They are not plotted here, since they practically coincide with the already plotted lines obtained from the caesium data.
Figure 5.7: Stability of the gradient for run 1952, 29th of April (one magnetic field setting). The stability of the gradients over a period of a run (one day) are in the range of 10 to 20 pT/cm for all the runs. The change of the gradient at cycle 220 is most probably due to an external field change.

The result is

\[ \frac{d_f}{Hg} \left( \frac{\partial B_0 z}{\partial z} \right) = (1.22 \pm 0.04) \times 10^{-27} \text{ cm/(pT/cm)} \] (5.23)

for \( B_0 \) down and

\[ \frac{d_f}{Hg} \left( \frac{\partial B_0 z}{\partial z} \right) = (-1.25 \pm 0.03) \times 10^{-27} \text{ cm/(pT/cm)} \] (5.24)

for \( B_0 \) up. Again, the average value is calculated:

\[ \frac{d_f}{Hg} \left( \frac{\partial B_0 z}{\partial z} \right)_{av} = (1.24 \pm 0.04) \times 10^{-27} \text{ cm/(pT/cm)} \] (5.25)

The discrepancy of the gradients is transformed to a discrepancy of the false EDM. Finally, the comparison with the theoretical value is made. The result obtained from Eqn. 5.16 is expressed in the same form as the results from the measurement:

\[ \frac{d_f}{Hg} \left( \frac{\partial B_0 z}{\partial z} \right)_{th} = 1.16 \times 10^{-27} \text{ cm/(pT/cm)} \] (5.26)

The value above is not in agreement with the two experimental results. A possible explanation is the inhomogeneity of the magnetic field gradient which directly affects the geometric phases.

**Corrections for the geometric phase related systematics**

A shift in the neutron precession frequency correlated with the change of the \( E \)-field corresponds to a nEDM according to Eqn. 1.14. The situation is a bit different when the neutron frequency is corrected with the precession frequency of the \(^{199}\text{Hg} \) atoms. The nEDM can then be extracted via
Figure 5.8: The histogram shows the fluctuations of the gradient from cycle 300 to cycle 550. The distribution of gradients is most probably gaussian. The parameters from the fit are $\mu = 3.422 \text{nT/cm}$ and $\sigma = 1.6 \text{pT/cm}$. The mean value is $\langle g \rangle = 3.421 \text{nT/cm}$ and the standard deviation is $\sqrt{\text{Var}(g)} = 1.7 \text{pT/cm}$. 

$\chi^2/d.o.f. = 1.41$
Figure 5.9: Geometric phase false EDM versus applied gradient. The dashed lines are obtained by fitting Eqn. 5.16 to the data. The solid lines stem from theory (Eqn. 5.16). The gradients in this plot are determined using the Cs magnetometers.
Figure 5.10: Geometric phase false EDM versus applied gradient as in Fig. 5.9. The gradients are average values over the precession chamber and are calculated from the fluxgate magnetometer maps.

The geometric phase effect. This procedure is described in [Bak06]. The ratio of the frequencies can be calculated using the energy equation

\[ h \nu = |2 \mu B \pm 2dE|. \]  

The dependence of the frequency ratio on the nEDM to first order is

\[ \frac{\nu_n}{\nu_{\text{Hg}}} = \left| \frac{\gamma_n}{\gamma_{\text{Hg}}} \right| + 2 \frac{d_n + |\gamma_n/\gamma_{\text{Hg}}|d_{\text{Hg}}}{h\nu_{\text{Hg}}} E. \]  

Hence, the measured nEDM contains \( d_n \) and \( d_{\text{Hg}} \). The geometric phase effect produces a non-negligible \( d_{f,\text{Hg}} \), which enters the measured nEDM as follows:

\[ d_{\text{meas}} = d_n + |\gamma_n/\gamma_{\text{Hg}}|d_{f,\text{Hg}}. \]  

It follows that, the geometric phase false EDM of \(^{199}\text{Hg}\) contributes to the measured nEDM as

\[ d_{n\leftrightarrow f,\text{Hg}} = |\gamma_n/\gamma_{\text{Hg}}|d_{\text{Hg}} = \frac{h}{8} \left| \frac{\gamma_n\gamma_{\text{Hg}}}{\gamma_{\text{Hg}}} \right| \left( \frac{\partial B_{0z}}{\partial z} \right) \frac{R^2}{c^2} \left[ 1 - \frac{\omega_0^2}{\omega_f^2} \right]^{-1}. \]  

Since \( \omega_0/\omega_f \approx 0.07 \), the last term is \( \approx 1 \). Therefore, it is neglected in the following.

Since the UCN have very little kinetic energy, the centre of gravity is \( \Delta h = 0.28 \text{ cm} \) below the one of \(^{199}\text{Hg}\) atoms, which corresponds to the centre of the precession chamber. This means, UCN and mercury average differently the magnetic field over the volume of the precession chamber. Due to this, a vertical gradient shifts the frequency ratio \( \nu_n/\nu_{\text{Hg}} \) away from \( \gamma_n/\gamma_{\text{Hg}} \) according to

\[ R_a = \frac{\nu_n}{\nu_{\text{Hg}}} \left| \frac{\gamma_{\text{Hg}}}{\gamma_n} \right| = 1 \pm \Delta h \left( \frac{\partial B_{0z}/\partial z}{B_{0z}} \right) \]  

(5.31)
where the $+,-$ corresponds to $B_0$ down,up respectively. The measured nEDM can be expressed
via Eqn. 5.30 and Eqn. 5.31 in terms of $R_a - 1$ as follows:

$$d_{\text{meas}} = d_n \pm \frac{\hbar}{8} |\gamma_n \gamma_{Hg}| \frac{R^2 B_{0z}}{c^2 \Delta h} (R_a - 1).$$

(5.32)

The two lines, with positive slope for $B_0$ down and negative slope for $B_0$ up, intersect at a point
where no geometric phase is present. The RAL-Sussex-ILL collaboration took EDM data at
different values for $R_a - 1$ and for both $B_0$ directions. They plotted EDM vs $R_a - 1$ as shown
in Fig. 5.11 and calculated the intersection point of the two fitted lines. The EDM value at the
crossing point refers to the true EDM $d_n$.

However, various systematic effects shift these lines and produce an additional EDM signal.
Local dipole fields and transverse quadrupole fields are related to the geometric phase. Therefore,
corrections for them shall be discussed here briefly. A local dipole moment produces an additional
gradient independent of the trim coil setting and the $B_0$ direction. This creates an offset in terms
of an EDM independent of $R_a - 1$ and the lines in the EDM vs $R_a - 1$ graph are shifted upwards
(see Fig. 5.12). Assuming a dipole at a distance $z_0$ below the electrode, the offset amounts to

$$d_{\text{dip,Hg}} = \frac{\hbar}{8} \left\langle \left( \frac{\partial B_{0z,\text{dip}}}{\partial z} \right) \left( 1 + \frac{R^2}{z_0^2} \right) \right\rangle \frac{\gamma_{Hg} R^2}{c^2},$$

(5.33)

where the gradient of the dipole is enhanced about a factor of $1 + R^2/z_0^2$ [Har06]. The correction
for the nEDM caused by a local dipole is then

$$d_{\text{dip}} = \frac{\hbar}{8} \left\langle \left( \frac{\partial B_{0z,\text{dip}}}{\partial z} \right) \left( 1 + \frac{R^2}{z_0^2} \right) \right\rangle \frac{\gamma_n \gamma_{Hg} |R|^2}{c^2}.$$ 

(5.34)

Another systematic related to the geometric phase is created by fields in the $xy$-plane, that
are not due to a vertical gradient. The most prominent example is the field of a quadrupole
moment aligned to the $z$-direction. The two spin species behave differently for such a quadrupole
field, as long as it is much smaller than $B_0$. The UCN are so slow that they follow the field lines
and precess around $\vec{B}_{0z} + \vec{B}_q$. The frequency is shifted as follows:

$$\nu_n = \frac{1}{2\pi} \gamma_n |(\vec{B}_{0z} + \vec{B}_q)| = \frac{1}{2\pi} \gamma_n B_{0z} \sqrt{1 + \frac{B_q^2}{B_{0z}^2}},$$

(5.35)

The $^{199}$Hg atoms move so fast that transverse field components are averaged out and only the
$z$-component is seen by the atoms. The frequency remains untouched by the quadrupole field:

$$\nu_{Hg} = \frac{1}{2\pi} \gamma_{Hg} B_{0z}.$$ 

(5.36)

Using now the formula for $\nu_n$ and $\nu_{Hg}$, Eqn. 5.31 changes to

$$R_a = 1 \pm \Delta h \left( \frac{\partial B_{0z}}{\partial z} \right)_{av} \frac{B_{0z}}{B_{0z}^2} + \frac{B_q^2}{B_{0z}^2},$$

(5.37)

where the root of Eqn. 5.35 has been expanded, since $B_q \ll B_{0z}$. This additional term might shift
the lines in Fig. 5.12 to the right. If this term is the same for $B_0$ up and $B_0$ down, no false EDM
is created. If $B_{q,\uparrow} \neq B_{q,\downarrow}$, $R_a$ is shifted differently thus creating a false EDM via the mercury
geometric phase. The measured EDM depending on $R_a - 1$ is then:

$$d_{\text{meas}} = d_n - \frac{d_{Hg}}{(\partial B_{0z}/\partial z)_{av}} \frac{\gamma_n |B_{0z}|}{\gamma_{Hg} \Delta h} \left( (R_a - 1) - \frac{B_{q,\uparrow}^2}{2B_{0z}^2} \right),$$

(5.38)
Figure 5.11: The graph shows the EDM as a function of the frequency ratio $R_a - 1$. Each data point corresponds to a different gradient and thus to a different $R_a - 1$. At the crossing point of the two fitted lines, no geometric phase is present and the EDM value corresponds to the true $d_q$. Plot taken from [Bak06].
Figure 5.12: Upper part: A local dipole on the wall produces a false EDM $d_{\text{dip}}$, which is independent of $R_a - 1$. The two original lines (solid) are shifted vertical. At the crossing point of the shifted lines (dashed), the false EDM due to the dipole is present. Lower part: A quadrupole field in the $xy$-plane shifts the original lines (solid) to the right. If the shift is not the same for $B_0$ up as for $B_0$ down, the crossing point of the shifted lines (dashed) are raised or lowered thus creating a false EDM $d_{\text{quad}}$. 
for \( B_0 \) up and

\[
d_{\text{meas}} = d_n + \frac{d_{t,\text{Hg}}}{(\partial B_{0z}/\partial z)} \frac{\gamma_n}{\gamma_{\text{Hg}}} \frac{B_{0z}}{\Delta h} \left( R_a - 1 - \frac{B_{q,\uparrow}^2 - B_{q,\downarrow}^2}{2B_{0z}^2} \right),
\]

for \( B_0 \) down. The resulting false EDM due to the quadrupole difference is then

\[
d_{\text{quad}} = \frac{d_{t,\text{Hg}}}{2(\partial B_{0z}/\partial z)} \frac{\gamma_n}{\gamma_{\text{Hg}}} \frac{B_{0z}}{\Delta h} \frac{B_{q,\uparrow}^2 - B_{q,\downarrow}^2}{2B_{0z}^2}. \]

From the fluxgate magnetometer field map for \( B_0 \) up and \( B_0 \) down, the following value was obtained:

\[
B_{q,\uparrow}^2 - B_{q,\downarrow}^2 = (0.3 \pm 0.5) \text{nT}^2.
\]

For an estimate of the false EDM due to the difference of quadrupole fields, I assume the values \( B_{0z} = 1.02 \, \mu\text{T}, \left| \gamma_n/\gamma_{\text{Hg}} \right| = 3.84 \), and \( \Delta h = 0.28 \) cm (taken from [Bak06]). For \( d_{t,\text{Hg}}/(\partial B_{0z}/\partial z) \), I take the values from Eqn. 5.22 and Eqn. 5.25. The results are listed in Tab. 5.3. The false EDM for the quadrupole difference is roughly three times the value given in Tab. 5.1. Further effort has to be done to determine the transverse fields with higher precision or reduce the fields with trim coils in order to correct for this effect.

### Light shifts

#### 5.2.1 Introduction

Another systematic correction concerning the \(^{199}\text{Hg}\) magnetometer is the light shift, which is a shift in the Larmor frequency due to the interaction of the probe light beam with the precessing \(^{199}\text{Hg}\) atoms. In general, if atoms are illuminated with light, the atomic levels are broadened and shifted due to the light. A quantum mechanical theory for these shifts was developed by Cohen Tannoudji [CT62]. A short description of it is given in [Cor77]. The effective Hamiltonian of the light interaction can be expressed as

\[
\delta \mathcal{H} = \delta \mathcal{E} - i\hbar \frac{\delta \Gamma}{2}
\]

with

\[
\delta \Gamma = \sum_m \frac{\hat{e}^* D|m\rangle\langle m|D\hat{e}}{\epsilon_0 \hbar^2} \int_0^\infty d\omega U(\omega) \frac{\Gamma/2}{(\omega - \omega_{mm})^2 + \Gamma^2/4}
\]
Figure 5.13: $U(\omega)$ is the spectral density of the light source and $(\omega - \omega_{m\mu})/[(\omega - \omega_{m\mu})^2 + \Gamma^2/4]$ is the dispersion curve of the illuminated atoms. The convolution of these two functions create the energy shift due to the light. Maximum light shift occurs for $\omega_{m\mu} - \omega_0 \approx \Delta \omega$ and no light shift is present for $\omega_{m\mu} = \omega_0$. Plot taken from [Cor77] for the broadening of the atomic levels and

$$\delta E = \sum_m \frac{\hat{e}^* D|m\rangle\langle m|D\hat{e}}{2\epsilon_0\hbar} \int_0^\infty d\omega U(\omega) \frac{\omega - \omega_{m\mu}}{(\omega - \omega_{m\mu})^2 + \Gamma^2/4}$$

(5.44)

for the shifting. In the equations above, $\hat{e}$ denotes the operator of the electric field of the light, $D$ the operator of the dipole moment of the illuminated atoms due to the valence electron of the excited state and $m$ all excited states which contribute to the absorption process. $U(\omega)$ is the spectral distribution of the light centred at $\omega_0$ and with a width $\Delta \omega$. The second function in the integral is the absorption function for Eqn. 5.43 and the dispersion function for Eqn. 5.44. $\omega_{m\mu} = (E_m - E_{\mu})/\hbar$ is the transition frequency and $\Gamma$ is the width of the optical absorption line of the precessing atoms. The index $\mu$ denotes the ground state of the atoms. The convolution of $U(\omega)$ and the dispersion function (see Fig. 5.13) is responsible for the energy shift, which is denoted as light shift.

In the nEDM experiment, $^{199}$Hg is illuminated by circularly polarised light from a $^{204}$Hg lamp. The absorption lines of the two isotopes differ slightly thus producing a light shift according to
Eqn. 5.44. In some simple cases, the light shift can be interpreted as the action of a fictitious magnetic field [CT71]. A light beam with polarisation $\sigma^+$ can be described as an additional magnetic field in the direction of the beam. I will refer to this field as $\vec{B}_{\sigma^+}$. A deviation of the beam from the $xy$-plane by an angle $\alpha$ results in a $z$-component $\vec{B}_{\sigma^+}\sin(\alpha)$ parallel to $\vec{B}_0$. The $^{199}$Hg atoms experience thus the field $B_0 + B_{\sigma}\sin(\alpha)$, whilst the UCN only see $B_0$. This results in

$$R_a - 1 = \frac{\nu_n}{\nu_{n_{\perp}} \pm \Delta \nu_{n_{\perp}}} \left| \frac{\gamma_{n_{\perp}}}{\gamma_n} \right| - 1,$$

(5.45)

where $+$ is for $B_0$ up, - for $B_0$ down, and $\Delta \nu_{n_{\perp}} = (1/2\pi)\gamma_n B_{\sigma} \sin(\alpha)$. The shift of $R_a - 1$ in different directions for different $B_0$ directions implies a false nEDM due to the geometric phase

$$d_{\text{lightshift}} = \frac{d_{I_{1,\text{Hg}}}}{\partial B_0/\partial z} \left| \frac{\gamma_n}{\gamma_{n_{\perp}}} \right| \frac{B_0 \nu_n \Delta \nu_{n_{\perp}}}{\Delta \hbar \nu_{n_{\perp}}^2}. \quad (5.46)$$

**Direct light shift**

When measuring the nEDM, an alternating $E$-field is applied in addition to the $B_0$ field. Some years ago, the RAL-Sussex-ILL collaboration observed a dependence of the DC-level $I_1$ on the $E$-field [Doy05]. If $I_1$ is not exactly the same for positive HV as for the negative one, the light shift is different for the two HV polarities, which therefore generates a false EDM signal. This effect is called direct light shift, because it depends directly on $E$. The false HgEDM is given by the difference of the $^{199}$Hg frequency

$$d_{\text{direct, Hg}} = -\frac{\hbar(\nu_{n_{\perp}} + \Delta \nu_{n_{\perp}}) - (\nu_{n_{\perp}} + \Delta \nu_{n_{\perp}})}{4E}. \quad (5.47)$$

Assuming now that $B_0$ is completely stable, the difference in frequency is only due to light shifts. The expression then changes to

$$d_{\text{direct, Hg}} = -\frac{\hbar(\Delta \nu_{n_{\perp}} + \Delta \nu_{n_{\perp}})}{4E}. \quad (5.48)$$

The expression can be transformed to

$$d_{\text{direct, Hg}} = \frac{\hbar \Delta \nu_{n_{\perp}} + \Delta \nu_{n_{\perp}}}{4E}. \quad (5.49)$$

I define $\Delta \nu_{n_{\perp}} := \Delta \nu_{n_{\perp}} + \Delta \nu_{n_{\perp}}$ as a baseline level. This choice of the baseline is completely arbitrary since only the difference between $\Delta \nu_{n_{\perp}}$ and $\Delta \nu_{n_{\perp}}$ matters. Furthermore, the light shift depends linearly on the number of photons of the light beam and thus on the DC-level $I_1$: $\Delta \nu_{n_{\perp}} \sim j_{\text{ph}} \sim I_1$. Using these relations, the expression above changes to

$$d_{\text{direct, Hg}} = \frac{\hbar \Delta \nu_{n_{\perp}}(I_1(HV^-)/I_1(HV^+) - 1)}{4E}. \quad (5.50)$$

The false HgEDM can be transformed to a false nEDM by multiplying with $|\gamma_n/\gamma_{n_{\perp}}|:

$$d_{\text{direct}} = \frac{\gamma_n}{\gamma_{n_{\perp}}} \frac{\hbar \Delta \nu_{n_{\perp}}(I_1(HV^-)/I_1(HV^+) - 1)}{4E}. \quad (5.51)$$
5.2.2 Measurement of the direct light shift

The direct light shift is investigated in the following with data taken during the geometric phase measurement, in April/May 2011. During the measurement, the magnitude of the applied HV was 100 kV for both polarities. The polarity was changed every 20 cycles (about 40 minutes). In every cycle, the DC-level $I_1$ was measured. The idea is now to investigate whether there is a difference in the probe light intensity, which is correlated to the change of the HV polarity. The problem is that the intensity of the light drifts due to thermal instabilities. Therefore, we need a reference system. There is a photodiode fixed next to the cavity of the Hg lamp to monitor the brightness of the lamp. Nevertheless, it showed no correlation to the signal in the PMT. Another possibility to correct for drifts is to split the PMT-signal measured for the positive and the negative HV polarity and build the ratio $R_{DC} = \frac{I_1(HV^-)}{I_1(HV^+)}$ as illustrated in Fig. 5.14. A second potential problem still remains. There is a time difference between two neighbouring HV sequences, since it is impossible to measure for both polarities at the same time. However, Fig. 5.15 shows that this effect is small compared to the fluctuations of the light level. There is obviously no difference between the two curves for $+100$ kV and $-100$ kV.

5.2.3 Results and discussion

For the calculation of $d_{direct}$, $R_{DC} - 1$ is more convenient than $R_{DC}$. For all four runs 1952, 1955, 1961, and 1967, $R_{DC} - 1$ has been calculated and plotted in a histogram in Fig. 5.16. Due to some remaining drifts of the probe light, the peak in the histogram is not symmetric. However, from the histogram, a mean value and an error is obtained:

$$R_{DC} - 1 = 6.3 \times 10^{-5} \pm 4.5 \times 10^{-3} \quad (5.52)$$

The binning in Fig. 5.16 is relatively rough. A finer binning would destroy the shape of the peak, since the limit of bit resolution is reached (see Fig. 5.17). An estimate of the light shift in the experiment has been performed by Pendlebury [Pen05]. The light shift along the beam can be expressed as follows:

$$\Delta \nu_{Hg,\text{total}} = \sigma \times j_{ph} \times 0.0094, \quad (5.53)$$

where $\sigma = 2.64 \times 10^{-13}$ cm is the absorption cross-section of $^{199}$Hg for $^{204}$Hg light, $j_{ph} = 3 \times 10^9$ photons/(cm$^2$s$^2$) the light flux through the chamber, and 0.0094 is the value of the convolution integral in Eqn. 5.44. Due to the redesigned probe light optics, the beam is collimated and three times more light enters the chamber than with the old polarisation optics. The light...
Figure 5.15: DC\textsubscript{1} level in V for positive and negative HV. The data has been taken from the runs 1952, 1955, 1961 and 1967, which is about two full days of measurement. These runs have been chosen because the drift of the lamp was rather low compared to other runs.

Figure 5.16: Histogram of $R_{DC} - 1$: From the distribution, a value and an error is obtained for $R_{DC} - 1$. The distribution is not completely symmetric because of remaining drifts.
Figure 5.17: $R_{DC} - 1$ is plotted as a function of cycles. The bit resolution in the DC-level of the DAQ is clearly visible.

shift in the equation above has to be multiplied with 3. This results in a total light shift of $\Delta \nu_{Hg, total} = 2.23 \times 10^{-5}$ Hz. Now, only the light shift in $z$-direction is of interest. Assuming now that the photomultiplier and the polarisation optics can be adjusted in height better than a 1 cm. The distance between photomultiplier and polarisation optics is roughly 1 m. This implies an angle $\alpha = 10$ mrad between light beam and $xy$-plane, the light shift in $z$-direction is

$$\Delta \nu_{Hg} = \sin(\alpha) \Delta \nu_{Hg, total} = 2.23 \times 10^{-7} \text{ Hz.}$$

(5.54)

Using now Eqn. 5.50, the direct false nEDM can be determined to be

$$d_{direct} = 5.6 \times 10^{-31} \text{ cm} \pm 4.0 \times 10^{-29} \text{ cm}$$

(5.55)

A contribution of $\sigma(d_{direct}) = 4.0 \times 10^{-29} \text{ cm}$ to the systematics is completely negligible, even for phase III of the nEDM project.
6 Conclusions and Outlook

One part of my PhD work was to develop and assemble a test setup for investigations of depolarisation mechanisms of $^{199}$Hg, to take data with the device and to analyse them. A second part was the planning, the data taking and the subsequent analysis for the measurement of the $^{199}$Hg geometric phase. The result is compared with theory and estimates of correlated systematic effects are given.

I had the opportunity to organise shifts for the measurement of the $^{199}$Hg geometric phase and to take data with the nEDM apparatus, with which the best upper limit of the nEDM had been measured.

6.1 Chapter 3: An effective model for $^{199}$Hg polarisation

The presented model describes the evolution of the $^{199}$Hg polarisation with time. All known depolarisation mechanisms have been included. One remarkable result is the dilution of the polarisation, which is strongest at the beginning of optical pumping. After $t \sim T_\text{r}$, the polarisation is 7% lower than without dilution. The dilution becomes negligible when the duration of optical pumping exceeds $T_\text{r}$. The model has been tested with a MATLAB program and compared to the data taken with the test setup. At the moment the model and the data do not agree. In the model, reemission does not produce a decrease of polarisation for higher absorptions.

6.2 Chapter 4: Investigations of depolarisation mechanisms

The test setup was operated successfully under stable conditions for half a year. Typical performance parameters were $T_2 = 30$ s, a polarisation $P = 15\%$, and a S/N-ratio of 50. Tests of depolarisation were performed for different coatings, from which the perfluorinated paraffin showed with $40.5 \pm 1.2$ s the best value for $T_2$. The tests with the different cells show, that the amplitude of the signal as well as $T_2$ increases with the volume of the cell. For an increase of the volume of a factor of 3, the amplitude nearly doubles.

6.3 Chapter 5: Systematics correlated to the $^{199}$Hg magnetometer

The performed measurement of the geometric phase of $^{199}$Hg is in agreement with the theory. The exact size and shape of the gradients remain unclear, since the values obtained from the caesium magnetometers differ a bit from those obtained from the fluxgate magnetometer field map.

Concerning the quadrupole difference, one could think of measuring the transverse fields with higher precision. Vector Cs magnetometers are currently under development and might be the
proper tool to measure these fields with a sensitivity of $B_{\text{quad}} = 0.1 \text{nT}$.

Concerning the direct light shift, this effect is on a negligible level. Even so, a laser is currently set up for the reading light of the $^{199}\text{Hg}$ magnetometer which could be tuned to a frequency, where no light shift occurs at all [Fer11].

6.4 Outlook

For phase III of the nEDM experiment, the new polarisation chamber for $^{199}\text{Hg}$ should fulfill the following features:

- **Coating:**
  Use perfluorinated paraffin.

- **Photodiode as a monitoring tool:**
  Mount a UV photodiode opposite the pump light entrance window to monitor the optical pumping. The diode could also be useful to analyse the degree of polarisation of the pump light.

- **Size:**
  In principle, increasing the volume will help in increasing the polarisation, since one reduces reemission effects and the wall collision rate. However, there are two reasons to make it not too large. The illuminated area $A_{\text{ill}}$ should fulfill the inequality $A_{\text{ill}}/A > \Gamma_{\text{r}}/\Gamma_{\text{p}}$, which guarantees an efficient pumping. Furthermore, the size of the polarisation chamber should be small compared to the size of the precession chamber to get a reasonable amount of mercury into the precession chamber and having still short filling times.

- **$^{199}\text{Hg}$ inlet:**
  Add a shutter between the polarisation chamber and the Hg source, in order to avoid dilution.

- **Polarisation optics:**
  Put the polarisation optics outside the vacuum tank, where a better control of the adjustment is guaranteed.
A Solution method for a linear differential equation

Non-homogeneous linear differential equations of first order possess in general an analytical solution. In the following, a method is described, how to obtain an explicit solution [Wal00]. A non-homogeneous linear differential equation, whose solution is a function $y(x)$, has the form

\[
\frac{dy}{dx} = g(x)y + h(x),
\]  

(A.1)

where $g(x)$ and $h(x)$ are known functions depending on $x$. In order to solve the non-homogeneous equation, a solution of the homogeneous equation

\[
\frac{dy}{dx} = g(x)y
\]  

(A.2)

has to be known first. A solution of this equation is obtained by separation of variables with subsequent integration:

\[
\int_{y_0}^y \frac{dy'}{y'} = \int_{x_0}^x g(x')dx',
\]  

(A.3)

where $x'$ and $y'$ are integration parameters. Solving for $y$ results in

\[
y = C \exp(G(x))
\]  

(A.4)

with

\[
G(x) = \int_{x_0}^x g(x')dx'.
\]  

(A.5)

The solution is a family of curves with a free parameter $C$. A solution of the non-homogeneous equation can be found, when the parameter $C$ is replaced by a function $C(x)$. This method is called variation of constants. Eqn. A.4 changes to

\[
y = C(x) \exp(G(x)).
\]  

(A.6)

Substituting the function above into the non-homogeneous equation, one obtains

\[
C'(x) \exp(G(x)) + C(x) \exp(G(x))g(x) = C(x) \exp(G(x))g(x) + h(x).
\]  

(A.7)

This expression can be solved for $C(x)$:

\[
C(x) = \int_{x_0}^x h(x') \exp(-G(x'))dx' + C_0,
\]  

(A.8)
where $C_0$ is again a free parameter and $x'$ again the integration parameter. Substituting it in Eqn. A.6, one obtains

$$y = \left( \int_{x_0}^{x} h(x') \exp(-G(x'))dx' + C_0 \right) \exp(G(x))$$  \hspace{1cm} (A.9)

as a solution for the non-homogeneous equation. A boundary condition $y(x_0) = y_0$ defines the parameter $C_0$. The solution of a linear differential equation of first order with a boundary condition is unique.
B  Coating methods for the polarisation chamber

This chapter gives a description of the coating methods for the glass cells used in the test setup. The methods may have to be adapted slightly for the polarisation chamber of the main experiment.

B.0.1 Oils and greases

The Fomblin grease and the “J” oil of Apiezon are smeared on the surface by a finger or another suitable tool. Gloves have to be worn for this procedure. The Fomblin oil, less viscous than the Apiezon oil, is applied by placing little drops on the surface, putting the cell in an upright position and letting the drops flow down. Another possibility is to distribute the oil on the surface with a tool. The difficulty in this kind of coatings is to make sure that there are no uncoated areas.

B.0.2 Siliconising fluids

For AquaSil and SurfaSil, the glass cell is immersed in a solution containing these chemicals. Since the agents react chemically with the glass surface, a proper cleaning of the glass surface is therefore crucial. In a first step, the glass is cleaned with soap and water. Afterwards, it is rinsed with distilled water and dried with a hot air gun for some minutes. The solution with the siliconizing fluid has to be prepared in a next step. AquaSil is purchased as a 20% solution in mixture of alcohol. It has to be diluted with water to a 1% solution. SurfaSil has to be diluted with some organic solvent, in this case I use acetone. The cell is then immersed completely in the solution for about five to ten seconds. In this time, a thin layer of the agent covers the surface. Afterwards, the cell is rinsed thoroughly with methanol to remove the remaining, unreacted material. At the end, the cell is air-dried in a fume hood for a day. During the whole process the solution is agitated by a magnetic stirrer. It has to be prepared on the same day the coating is applied, since the solution is not stable over a few days. Furthermore, the agent has a shelf life of only one year. A few precautions are to be taken when dealing with these fluids. Both of them are flammable and SurfaSil even produces hydrochloric acid in small amounts. The whole preparation and coating process has to be performed in a fume hood. Gloves have to be worn to prevent splashes on the skin.

B.0.3 Paraffins

For paraffins, vacuum deposition is used. A special device had to be constructed, which is shown in Fig. B.1. The whole setup is placed in a vacuum vessel. The cell and the paraffin are put on a copper plate. A 10Ω resistance is attached to the bottom of the copper plate, which is heated with a power of 10 W. The amount of paraffin used in one coating process is between five to ten milligrams. It is placed in the middle of the plate. Around the paraffin, a steel tube with an inner
Figure B.1: Vacuum deposition of paraffins: A heater causes the paraffin on the plate to evaporate. The paraffin vapour coats then the cool walls of the glass cell.

diameter 5 cm and a length of 3 cm is placed and thereon, the cell with one quartz window on top. At the edge of the copper plate, a thermosensor PT100 is fixed to measure its temperature. Of course, the temperature at the location of the paraffin will be a few degrees higher than at the edge, but for a rough estimate, it is fine. On top of the vacuum vessel, there is a window to observe the coating process.

First, the whole vessel is evacuated to a pressure of approximately $10^{-5}$ mbar. Once the heater is turned on, the temperature and the pressure rise. When a temperature of $\sim 80 ^\circ C$ is reached, the paraffin melts. The pressure reaches the $10^{-3}$ mbar region and stabilises at this value. After 16 minutes, the size of the paraffin droplets start to decrease and finally disappear after about 20 minutes. This is the length of a typical coating process. The temperature has then reached 160$^\circ$C. The rise of pressure and temperature are shown in Fig. B.2. The heater and the pump are switched off and the vessel is vented within a few minutes. Concerning the perfluorinated paraffin, it behaves practically the same as the normal paraffin, but there is no liquid phase. The powder sublimates slowly at a temperature of 160$^\circ$C. The steel tube inserted between the copper plate and the cell guarantees that the lowest part of the cell is cool enough, that it is covered with paraffin. The thickness of the coating decreases with height. Looking at the upper part of the cell, the coating is invisible and it cannot be guaranteed, that really the whole surface to the upper end is coated. Therefore the cell has to be coated twice; after the first coating it is turned upside down.

The coating is visible as a greyish layer on the glass. It gets thinner towards the middle of the cell. The perfluorinated paraffin looks the same, but the thickness of the coating seems to be randomly distributed. In order to make the homogeneity visible, excess paraffin was used in Fig. B.3.
Figure B.2: Characteristic behaviour of the pressure and the temperature when coating the cell with 5 mg of paraffin. The solid line shows the temperature and the dashed line shows the pressure. The pressure climbs to $10^{-3}$ mbar when heated and stays there. Closing the valve to the pump lets the pressure increase further. The temperature increases steadily to 160°C.
Figure B.3: Homogeneous paraffin coating on a prototype glass cell. In order to make the coating visible, 40 mg of paraffin are used to make the homogeneity of the coating visible.

B.0.4 Black Teflon

Black Teflon is natural Teflon mixed with 25% carbon to make it black. It is a solid tube and would be used for the polarisation chamber in the experiment. In the test setup, a hollow cylinder is placed in the glass cell (see Fig. B.4). The two quartz windows are covered with Fomblin oil. This means that only 80% of the surface is black Teflon. Nevertheless, the relaxation time for black Teflon can be extracted by other measurements. One can measure $T_{\text{mixed}}$ and $T_{\text{Fomblin}}$ and from this, one can determine $T_{\text{blackTeflon}}$.

$$\frac{1}{T_{\text{mixed}}} = \frac{0.2}{T_{\text{Fomblin}}} + \frac{0.8}{T_{\text{blackTeflon}}} \quad (B.1)$$
Figure B.4: The black Teflon cylinder mounted inside the cell. Two holes in the cylinder (only one is visible here) allow the mercury to enter and leave the cell.
C Transmission and reflectivity measurements

The transmission and reflectivity measurements were performed with a UV-VIS-NIR spectrometer Cary 500i from Agilent [Tec11]. The range of wavelengths can be chosen between 180 nm and 1200 nm. For the following measurements, a fixed range between 200 nm and 400 nm was scanned. There are two beams used, one which is guided to the sample and another one as a reference beam. Before the actual measurement, a baseline run has to be performed to calibrate the two beams. The samples are quartz windows used for the glass cell (see Sec. 4.1). The coating is applied directly on the quartz according to the application method described in Sec. B.

The measurement of the transmission is done as follows. The quartz window with the coating is put in the sample beam, whilst the reference beam is untouched. Then the wavelength range is scanned. The stepsize can be chosen freely. For the measurements in Sec. 4.2, 1 nm was taken.

The determination of the reflectivity is done in a similar way. The sample beam is guided to an Ulbricht sphere (see Fig. C.1). This is a hollow sphere with a completely white inner wall and with small holes for the light entrance. The inner wall of an integrating sphere has a reflectivity of nearly 100% over the whole wavelength range and the light is reflected diffusely. The quartz plate is attached from outside on one hole of the sphere. Due to this arrangement, the reflectivity is integrated over all angles. For the baseline measurement, a reflectivity standard is attached at the hole where the samples are located later on. The reflectivity standard is made from the same material as the inner wall of the sphere and thus shows a reflectivity of nearly 100%. The light leaves the integrating sphere through a second hole and is detected by a photodiode.
Figure C.1: View of spectrometer with Ulbricht sphere from top: the reference light beam (right) goes from the light source directly to the photodiode, whilst the sample beam (left) is guided to the Ulbricht sphere in which the beam is reflected many times, until it leaves the sphere again and is guided to the second photodiode. The quartz plate is attached to the Ulbricht sphere from outside on one hole of the sphere.
Bibliography


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Marlon Béla Horras

Education

• 2012 Ph. D. Physics, ETH Zürich
  – Dissertation: A highly sensitive $^{199}$Hg magnetometer for the nEDM experiment

• 2001-2007 Studies of Physics at the University of Basel
  – Master of Science in Physics, 2007, grade 5.9 out of 6
  – Master thesis: Das adiabatische Limit in einer total skalaren, QED-ähnlichen Theorie
  – Bachelor of Science in Physics, 2005, grade 5.9 out of 6

• 2001 Matura: Gymnasium Liestal

Employment

• 2008-2012 Ph. D. Student at Paul Scherrer Institute

• 2007-2008 Teaching associate at University of Basel

Teaching

• 2010 Supervision of students in practica at Technical University of Munich

• 2007-2008 Teaching associate at University of Basel
Conferences

- 1.-4.09.2009 Annual Meeting of the Swiss Physical Society
  (Innsbruck, A): The Hg magnetometer of the nEDM experiment
- 21.-22.06.2010 Annual Meeting of the Swiss Physical Society
  (Basel, CH): The mercury magnetometer in the nEDM experiment (Poster)
- 15.-17.06.2011 Annual Meeting of the Swiss Physical Society
  (Lausanne, CH): A measurement of the Hg geometric phase (Poster)

Schools

- CHIPP PhD Summer School, 1.-7.08.2010, (Zuoz, CH)

Publications

1. An improved Search for the Neutron Electric Dipole Moment
2. New constraints on Lorentz invariance violation from the neutron electric dipole moment
3. Testing isotropy of the universe using Ramsey resonance technique on ultracold neutron spin
5. Neutron to Mirror-Neutron Oscillations in the Presence of Mirror Magnetic Fields (I. Altarev,