Spin Relaxation of ^{199}Hg Vapor on Paraffin-coated Walls

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Abstract

Polarized ^{199}Hg vapor in cells coated with the paraffin $CH_3(CH_2)_{30}CH_3$ is used in searches for T-violating permanent electric dipole moments. A precise measurement of the dipole moment requires that the lifetime of the polarization be as long as possible. Thus, determining the relaxation mechanism may be important in increasing the lifetime of these cells. The dominant relaxation mechanism seems to be a paramagnetic impurity caused by a broken bond between a carbon and a hydrogen in the paraffin walls.

1 Search for T-violation

An important theorem in physics is the CPT theorem, where C is charge conjugation, the operation of changing matter to antimatter, P is parity, the operation of inverting coordinates about a specified origin, and T is time reversal, the operation of reversing a process in time. The CPT theorem states that under all three operations in any order, physical laws remain invariant. CP-violation has been observed which means that T-violation is also expected in some way such that CPT is conserved. However, CP-violation has thus far only been observed in one system, making a measurement of T-violation important for testing whether theories have adequately explained the phenomenon.

The Standard Model and other theories predict that permanent electric dipole moments (EDMs) are sources of T-violation. To see that an EDM violates T symmetry, consider the Hamiltonian of a particle in and electric field $\mathbf{E}$ and a magnetic field $\mathbf{B}$ given by

$$H = -(d\mathbf{E} + \mu\mathbf{B}) \cdot \frac{\mathbf{S}}{S} \tag{1}$$

where d is the electric dipole moment, μ is the magnetic dipole moment, and $\mathbf{S}$ is the spin. Under the T operation, $\mathbf{E}$ does not change sign whereas both $\mathbf{B}$ and $\mathbf{I}$ do. Therefore the Hamiltonian is not invariant under $t \to -t$.

2 Measurement of the EDM

Measuring the dipole moment involves the use of polarized ^{199}Hg vapor in cells coated with paraffin. ^{199}Hg is ideal for several reasons. Its total electronic spin is zero and its nuclear spin is $\frac{1}{2}$, which makes it a simple atom to study. The EDM is also known to be larger in heavy atoms. The mercury atoms are first polarized by sending ciularly polarized light though the cell. If the circularly polarized light, which has spin 1, is the right energy, it will excite the transition from the $m = -\frac{1}{2}$ state to the $m = \frac{1}{2}$ state, or vice versa. The mercury spins are then polarized so that all of their nuclear spins are pointing in one direction. This process is called optical pumping.

In the experiment, there are two ^{199}Hg vapor cells each in an electric and magnetic field. The magnetic field goes through both cells with the same orientation and is perpendicular to the polarization of the spins. The electric field is parallel to the magnetic field in one cell and is antiparallel in the other (see Figure 1). The spins will precess about the fields with the frequencies

$$\omega_{parallel} = \frac{2\mu B + 2dE}{\hbar}, \ \omega_{antiparallel} = \frac{2\mu B - 2dE}{\hbar} \tag{2}$$

giving a total precession frequency difference of $\Delta\omega = \frac{4dE}{\hbar}$.

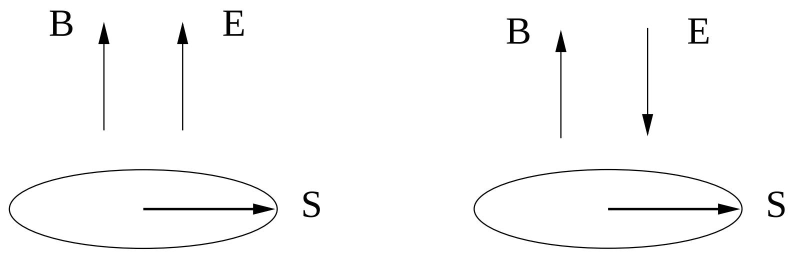

Figure 1: Orientation of fields and spins in the EDM experiment

The difference in the precession frequency is detected by sending linearly polarized light thought the cells. The plane of the linearly polarized light will rotate as it interacts with the spins, and can then be detected to determine the precession frequency. A difference in the precession frequency of the two cells is a signal for an EDM.

3 Measuring the Relaxation Rate of the Polarization

It is important to keep the spins polarized during the measurement of the EDM. The noise limits the precision of the measurement and is inversely proportional to the square root of the lifetime T_1 of the vapor in the cell. The motivation for studying the relaxation of the spins is to determine the mechanism of relaxation and perhaps to use the knowledge to increase the the lifetime of the cell.

The setup for measuring the relaxation rate of the spins is shown below (see Figure 2). A mercury lamp, lit by a microwave cavity, sends light through a linear polarizer. The linear polarizer is at a 45^o angle to a quarter wave plate, which delays one component of the electric field one quarter of a wavelength. This delay produces circularly polarized light, where the electric field rotates in the plane perpendicular to the direction of propagation. The circularly polarized light is then sent through the ^{199}Hg vapor cell to pump the spins.

To measure the polarization of the spins, the quarter wave plate is removed and a modulator is put in its place (see Figure 3). The modulator is a piece of quartz subjected to periodic mechanical vibrations which change its index of refraction. At one point in this mechanical oscillation, circularly polarized light in one direction is allowed through while one-half wave later, the other polarization is let through. If one polarization is dominant, then more light of the same polarization will go through compared to light of the opposite polarization. The difference between the intensities of

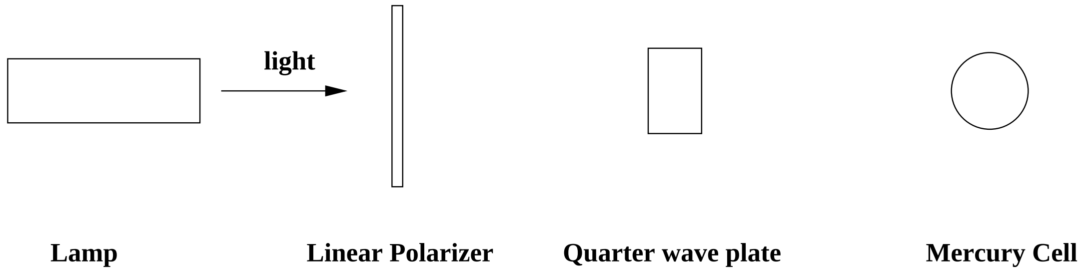

Figure 2: Setup for optical pumping

these two polarizations is proportional to the polarization of the spins. This difference is detected by a lock-in scheme. A lock-in amplifier is locked to the frequency of oscillation of the modulator and reads out the difference in the readings of the photomultiplier.

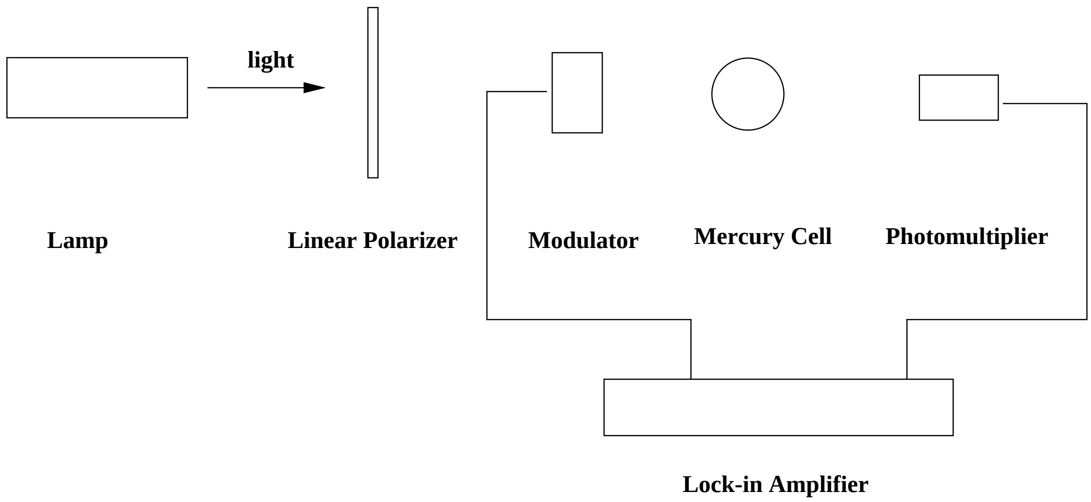

Figure 3: Setup for detecting the polarization of the spins

4 Determining the Mechanism of Relaxation

The relaxation rate had previously been known to be independent of both pressure and density, implying that the interaction between the mercury atoms is not the mechanism of relaxation. The relaxation must be due to the interaction between the vapor and the walls. The paraffin $CH_3(CH_2)_{30}CH_3$ has been found been found so far to be the best substance for increasing the lifetime. One of the reasons for this is that this saturated carbon structure has a low adsorption energy around 0.022 eV. Since this adsorption energy is so low, the time that the mercury sticks to the wall is reduced. The mercury can get kicked out of the potential well by thermal fluctuations with relative ease.

The hypothesis is that the spins are relaxing due a paramagnetic impurity caused by the removal of one of the hydrogens on the end of the carbon chain. There is then an exposed electron at the end of the chain which increases the adsorption energy on the walls causing the mercury to stick for a longer period of time. It also creates a magnetic dipole that can interact with the dipole moment of the mercury. The longer the atom sticks on the walls, the longer it experiences the random fluctuating magnetic fields on the walls and the easier it is for the spin to relax.

Relaxation can be studied as a function of both an external magnetic field and temperature. As a function of magnetic field, there are two main models. The first is the contact interaction given by the Hamiltonian

$$H_c = \alpha \mathbf{K} \cdot \mathbf{S} \tag{3}$$

which results in the form

$$\frac{1}{T_1} \propto \frac{1}{1 + (\omega_S - \omega_K)^2 \left((\alpha/\hbar)^2 + \tau_c^2 \right)} \tag{4}$$

for the relaxation rate where ω_S is the precession frequency of the ^{199}Hg atom, ω_K is the precession frequency of the spin of the electron on the wall, and τ_c is the correlation time, or the characteristic time of the fluctuations of the magnetic field on the walls.

The other mechanism of relaxation is that of a dipole-dipole interaction, which results in the from the interaction of two magnetic dipoles. The Hamiltonian for such an interaction is

$$H_{dd} = \frac{\boldsymbol{\mu}_K \cdot \boldsymbol{\mu}_S - 3(\boldsymbol{\mu}_K \cdot \mathbf{r})(\boldsymbol{\mu}_S \cdot \mathbf{r})}{r^3} \tag{5}$$

which gives a relaxation time

$$\frac{1}{T_1} = \frac{1}{10T_0} \left(\frac{1}{1 + (\omega_S - \omega_K)^2 \tau_c^2} + \frac{6}{1 + (\omega_S + \omega_K)^2 \tau_c^2} + \frac{3}{1 + \omega_K^2 \tau_c^2} \right) \tag{6}$$

Since the precession frequecies are proportional to the magnetic field, the relaxation rate is actually a function of the magentic field in either the case of the contact interaction or the dipole-dipole interaction. A sample of the data taken is shown below (see Fig. 4).

The first notable feature of the graph is that the value does not drop to zero for high fields, but rather flattens out at a value of roughly 0.3. Since the rate drops to zero at high fields for the contact interaction, it is not likely to be the process that dominates. There is a turnover point where the rate drops off called the magnetic

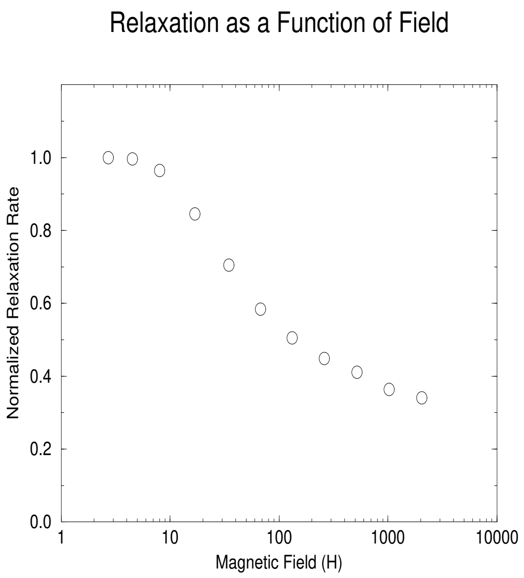

Figure 4: Magnetic field dependence of Relaxation Rate

field decoupling width ΔH. The value of ΔH can be determined by a fit to the rate equation for the dipole-dipole interaction. In equation 6, the precession frequency ω_S of the electron is much larger than that of ω_K, the spin of the mercury. So a plot of the relaxation rate as a function of magnetic field can actually be fitted to

$$\frac{1}{10T_0}\left(\frac{7}{1+\frac{H}{\Delta H}}+\frac{3}{1+\frac{H}{H_1}}\right) \tag{7}$$

where ΔH is about 1000 times H_1 (since ω_S is about 1000 times ω_K). Using this fit, the value of ΔH can be determined. All of the values of ΔH fall between 20-50G, which is consistent with τ_c on the order of 10^{-9} seconds. The theoretical values for τ_c for a proton and an electron are 10^{-6} seconds and 10^{-9} seconds, respectively. Clearly, the data agrees with the hypothesis that the interaction is with an electron rather than a proton, which supports the theory that there is a broken bond in the paraffin.

A plot of the relaxation rate of a single cell as a function of magnetic field for several temperatures is given below (See Fig. 5). It can be seen here that ΔH is independent of temperature. The model that predicts that ΔH is independent of temperature is one where where $\tau_c \ll \tau_s$ where τ_s is the average time that the mercury sticks on the walls. This model also predicts that

$$\frac{1}{T_1} \propto e^{E_a/kT} \tag{8}$$

where $\frac{1}{T_1}$ is the relaxation rate at low magnetic fields. A plot of this data is shown below (see Fig. 6). The adsorption energy E_a can be determined by a fit to this plot and a value of 0.15 eV was found. This energy is far greater than the value predicted by theory for the paraffin that has no broken bonds (0.022 ev). There is some effect that is increasing the adsorption energy and increasing the relaxation rate.

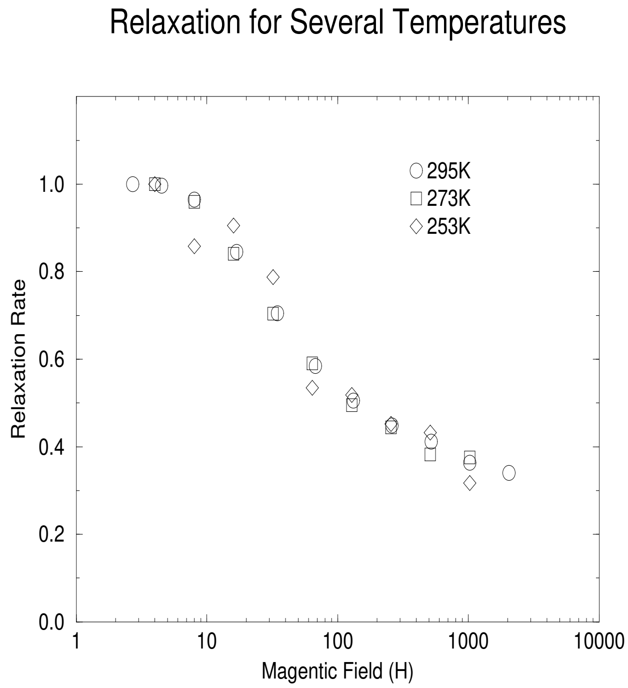

Figure 5: Magnetic field dependence for several temperatures

As a last check for consistency, the sticking time τ_s can be calculated from the adsorption energy using the equation

$$\tau_s = \tau_0 e^{E_a/kT} \tag{9}$$

where τ_0 is the high temperature limit of the sticking time, which is about 10^{-12} seconds. The value calculated is about the same as τ_c ($\approx 1 - 1.5 \times 10^{-9}$ seconds), which might be a slight inconsistency. The assumption made earlier was that the correlation time was less than the sticking time, but it cannot be shown definitively that this is true. More data needs to be taken in order for that to be determined. However, the sticking time need only be about 3 times larger than the correlation time for the theory to be consistent with the data.

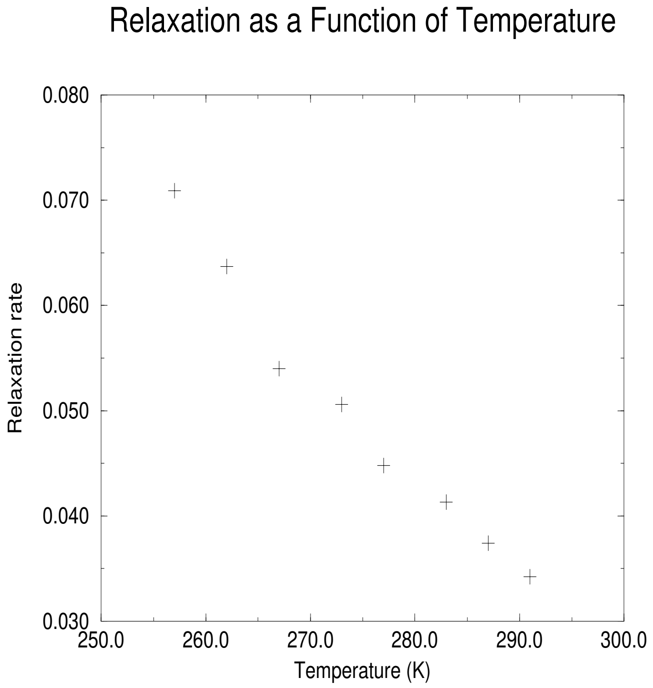

Figure 6: Relaxation rate as a function of temperature

5 Conclusion

All of the data collected seems consistent with the hypothesis that a broken bond is the main mechanism of relaxation. More data needs to be taken in order to make sure that the correlation time is actually greater than the sticking time. The next step would be to figure out what could be done about these broken bonds and to increase the lifetime of the cells.

6 Acknowledgements

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