Improved Limits on the Static Electric Dipole Moment of Cs through Spin-Precession Measurements in Superfluid He-Coated Cells

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Abstract

Vapors of Rb and Cs have recently been shown to persist for several thousand seconds in superfluid ^{4}He coated cells at 2°K, opening the door to a variety of fundamental physics measurements[1]. We propose the development of spin precession measurements with Cs in superfluid coated cells, leading to more precise limits on the electric dipole moment (EDM) of atomic Cs. We conservatively expect a factor of 50 improvement over the current, published limits for Cs, leading to more stringent limits for the electron EDM. The envisioned improvement stems primarily from the extremely long electronic spin coherence times, 100 seconds or longer for Cs, expected in these cells. It should also be possible to utilize superconducting shielding to reduce magnetic field fluctuations (the dominant source of noise in current measurements) by many orders of magnitude. Because Rb can also be loaded into these cells and has a much smaller sensitivity to the electron's EDM than Cs, it can serve as a magnetometer to further reduce systematic errors associated with magnetic field fluctuations. The proposed measurement is an opportunity to investigate the physics of superfluid coated cells and to develop the first useful applications of their properties.

1 Objective

We propose the development of spin precession measurements of atomic Cs in cells coated with superfluid ^{4}He, leading to more precise limits on the electric dipole moment (EDM) in Cs. Measurements in superfluid cells should provide roughly a factor of 50 improvement over the current limits for Cs, leading to improved limits on the electron's EDM. This work will be done in collaboration with Prof. T. Yabuzaki at Kyoto University, who pioneered the production of alkali metal vapors in superfluid ^{4}He-coated vapor cells. In the process of developing this measurement, we will have the opportunity of carefully investigating the physics of these cells, and characterizing the optical pumping process in a unique physical environment.

2 Motivation

At present, the most stringent limits on the magnitude of the EDM of the electron are obtained from atomic beam measurements of atomic Tl[2] and cell experiments on atomic Cs[3]. These measurements provide valuable constraints on suggested extensions to the electroweak standard model by placing limits on the presence of time-reversal (T) non-invariant phases which can give rise to an EDM. For example, models with T non-invariant Higgs couplings and super-symmetric models both introduce possible electron EDM's larger than current experimental upper limits[4]. Measurements in Cs are appealing, in that the theoretical uncertainties involved in producing appropriate limits on the electron EDM are much smaller than the corresponding limits in Tl. This strongly motivates improved experiments in Cs in parallel with measurements in Tl being carried out at Berkeley.

We view this measurement as an opportunity to evaluate the physics of superfluid ^{4}He-coated alkali metal vapor cells at low temperatures, and to develop the first useful applications of their properties. Our approach is complementary to the laser trapping techniques under development in a number of laboratories, in that no external fields are required to maintain the population of atoms under study. Because a wide range of atomic (and molecular) species can be loaded into superfluid He-coated cells, we ultimately envision a series of spin precession measurements on a variety of species, including Tl and possibly isotopes of Radium[5], offering further improvements in limits for electronic and nuclear EDMs. The long coherence time expected in these cells may also make them useful tools for a variety of precision measurements including tests of T invariance in β-decay, searches for spin dependent forces, and tests of the linearity of quantum mechanics. Applications for medical imaging using hyperpolarized gases may be possible as well.

3 Technical Approach

Introduction

For our first EDM measurements, we hope to closely follow the experimental technique developed by Hunter *et. al*[3] to measure the atomic EDM in the ground state of Cs ($6S_{1/2}$,F=4). This sensitive technique uses the electrostatic analog of the Hanle effect to measure spin precession correlated with the direction of an applied electric field. The cryogenic environment and extremely long spin correlation time then provide a number of straightforward and powerful avenues to improve on the systematic errors and the sources of noise associated with these measurements. We present our technical approach in three steps. First, we detail measurements we are currently performing, which will establish the optical pumping and probing techniques we hope to apply to a measurement of the EDM. Then we discuss the introduction of magnetic shielding to provide the necessary field homogeneity and stability for our proposed experiment. Finally, we outline a measurement of the EDM of atomic Cs and discuss various strategies for reducing or characterizing systematic errors.

Generation and Optical Pumping of Cs in Superfluid-Coated Cells

In our proposed measurement, we plan to exploit very recent breakthroughs made in Prof. Yabuzaki's lab[1] in preparing superfluid ^{4}He-coated cells with 10^8 to 10^{10} atoms/cm^3 of Cs trapped in the *vapor* state for time periods of up to an hour. This technique is amazingly simple: a cell filled with 4 to 10 atm of He gas is cooled to 2K in a flowing vapor cryostat. At this temperature, a superfluid He film coats the walls of the cell. A Nd:YLF laser is operated for a few seconds to ablate Cs from a small patch of metallic Cs on the inner face of one of the cell walls. The Cs vapor is then observed to persist for thousands of seconds. Equally important is the fact that the transverse spin coherence time is expected to be longer than 100 seconds[6] (an improvement of more than 3 orders of magnitude over previous electronic coherence times in these experiments!). The first step towards the EDM measurement is a determination of this spin destruction rate, through a measurement of longitudinal relaxation time, T_1, for Rb and Cs in superfluid He-coated cells (T_1 is the decay time of the polarization parallel to the magnetic field).

The degree of Cs polarization is established by a balance between laser optical pumping and relaxation processes in the cell. Optical pumping is reasonably straightforward for alkali metals in this environment. The width of the absorption lines for Rb and Cs D1 radiation has been observed to be quite broad, approximately 4 GHz, thus single frequency pumping lasers are not required. There is no quenching gas such as N_2 in these cells, so excited states decay by fluorescence. But, because we plan to work with Cs vapors with densities of about 1×10^{10} cm^{-3}, we are comfortably below the radiative trapping limit. We expect that electronic spin relaxation will be dominated by binary collisions with the He buffer gas[7], making it possible to determine the electronic spin destruction rate for Cs-He collisions through "relaxation in the dark" measurements of the limiting longitudinal relaxation rate, $1/T_1$. The observed decay rate can then be related to the electronic spin destruction rate $(1/T_{sd})$, through the formula:

$$\frac{1}{T_1} = \frac{1}{1+\bar{\epsilon}} \left(\frac{1}{T_{sd}} \right)$$

where $\bar{\epsilon} \approx 21$ is the nuclear slowing down factor for ^{133}Cs in the limit of small electronic polarization[8].

We have recently refined measurements of the longitudinal relaxation time in Rb-He cells for temperatures between 100 and 180°C and established a dramatic, T^4, temperature dependence to the spin destruction process mediated by the ^{4}He buffer gas[9]. These data reinforce our expectation of extremely long spin relaxation times in lower temperature Cs-He cells. Measurement of the longitudinal spin relaxation time in a cell at 2°K provides an important benchmark towards the establishment of the long coherence times we seek for the EDM experiment, and does not need the extremely homogeneous magnetic fields required in a direct measurement of the transverse relaxation time, T_2.

Substantial progress has been made towards a measurement of T_1 through recent collaborative work between our group and Kyoto University (see Fig. 1). At Princeton, we use 1720 cylindrical glass cells, about 2.5 cm long and 2 cm in diameter. These cells are filled with about 8 atm of ^{4}He and less than a milligram of Cs. The cells are then cooled to about 2° K in a customized cryostat (an AET Engineering, Kel Tran VP). We apply a magnetic field of 5 Gauss along the z-axis. A 1 mW, circularly polarized pump beam, introduced parallel to the magnetic field, briefly illuminates the alkali metal vapor with 895nm radiation to produce polarization. At present, we use an Argon ion laser-pumped Ti:Saph laser for this purpose, although for future experiments we plan to use DBR diode lasers we have developed in collaboration with the Sarnoff Corporation and Prof. Boetez at Univ. of Wisconsin. The ensuing relaxation is then periodically monitored by a weak probe beam also introduced along the z-axis. The polarization of the probe is modulated using a photoelastic modulator (PEM). After passing through the vapor, the probe beam is detected with a low noise photodetector (for example, a Thorlabs PDA150), and monitored using a lock-in amplifier. Fluorescence signals (detected using a LN_2 cooled CCD array attached to the focal plane of a spectrometer) averaged over a one second time interval will provide adequate information on the time evolution of the density of the Cs in the cell, and serve to normalize the probe signals in the extraction of the longitudinal relaxation time.

The development of the probing technique in this measurement is central to our EDM experiment. Although the PEM, because it modulates the circular polarization of the probe beam, ensures that there is no **net** optical pumping of the Cs by the probe, it may still induce relaxation in the sample. For this reason we are interested in lowering the probe power as much as is possible. Recently Martin and Anderson[10] have used polarization probes with powers as low as 2 nW/cm^2, indicating that extremely low probe powers are technically feasible (we plan to use less than 1 $\mu W/cm^2$ for our T1 measurements). At present our probe beam is generated by a narrow-line DBR laser diode (SDL 5702-H1) which runs at 852nm and provides adequate signal-to-noise ratios for our proposed measurements. The new cryostat and probe lasers were implemented quite recently, setting the stage for measurements of the longitudinal relaxation time[11].

There are a number of measurements investigating aspects of the optical pumping which may be pursued immediately, other than the longitudinal relaxation time. For example, one might perform detailed studies of the excitation and fluorescence spectra, studies of the cluster content and evolution in the buffer gas following ablation, and spin exchange polarization of ^{3}He. Professor Yabuzaki's group is also investigating Rb vapor on the surface of the superfluid He through fluoresence induced by evanescent laser light.[1]

The Magnetic Field Environment

The more challenging process of tailoring the magnetic field environment to optimize the coherence time and minimize the magnetic field fluctuations is the next step towards a measurement of the EDM in Cs. The danger is that magnetic inhomogeneities cause Cs spins at different points in the sample volume to precess at different rates. This results in shortened coherence times. We require the inhomogenieties in the magnetic field to be smaller than roughly 0.02μG/cm (motional narrowing may be exploited to relax this figure somewhat). This places strong constraints on the design of the cryostat we utilize for these measurements because all magnetic material must be avoided in the cryostat construction. At low temperatures additional difficulties arise. Indium, often used as a sealing material for low temperature systems, is inappropriate because it becomes a superconductor at 3.4°K. The resulting Meissner effect disturbs the magnetic fields in the vicinity of the sample, noticeably degrading magnetic resonance signals. To avoid these problems, we have designed a second-generation cryostat without magnetic or superconducting materials (with one exception we discuss below).

We plan to use a combination of passive and active shielding to reduce ambient fields to below about 0.01μG. A number of such geometries have been developed, with a few of them reporting residual ambient fields at the level of 0.001μG [12] (apparatus which reduce ambient fields to about 1μG are already in operation in our laboratory). The cryostat will be enclosed in 5 nested μ-metal shields, will provide a shielding factor of roughly 10^4 to 10^5 for the residual ambient field within a large (2-3m) diameter Helmholz coil arrangement. Within the magnetic shields we will situate another set of Helmholtz coils and a set of gradient-cancelling coils. We are also considering placing a pure iron shield within the cryostat, because of the small residual fields characteristic of pure iron shields (cooling this shield may have advantages as well).

Magnetic field fluctuations, the dominant source of noise in the current EDM measurements in Cs, can be reduced up to another five orders of magnitude by introducing a simple, open-ended, cylindrical superconducting shield [13] (one to two orders of magnitude will be sufficient for the proposed work). Although the axis of this cylinder will lie along the applied external field, the presence of this shielding will induce spatial inhomogeneities in the fields within the shielded volume. For this reason, we feel it is essential to work with extremely small, homogeneous ambient fields to ensure this effect is negligible. We also plan to optimize the design of the shield to reduce the size of the inhomogenieties to a minimum. In any case, we recognize that careful experimental investigation of this geometry will be required to fully understand the implications of introducing superconducting shielding. The longitudinal holding field will be roughly 0.1 μG, facilitating fine-tuning of the magnetic field profile through measurements of free induction decay and driven spin precession under transverse optical pumping, as is discussed by Kanorsky et. al[14] for the similar case of magnetic resonance spectroscopy of Cs in solid He.

Overview of an EDM Measurement

The basic strategy is to optically pump the Cs along the z-axis (taking the z-axis to lie along a 0.1 μG external magnetic field) polarizing the Cs. A static electric field, **E** is applied along the x-axis and a circularly polarized probe laser is introduced along the y-axis. If the Cs atom has a non-zero EDM, the atom will feel a torque due to the applied electric field, causing it's spin to precess. The change in the polarization along y, ΔP_y, when the electric field is reversed is given by:

$$\Delta P_y = \frac{2P_z\ D_{Cs}\ E\ \tau_{coh}}{(2I+1)\hbar}$$

where P_y and P_z are the Cs polarization along the y and z directions, D_{Cs} is the electric dipole moment, E is the electric field strength, τ_{coh} is the coherence time, and $I = 7/2$ is the nuclear spin. The transmission of the probe beam should then be modulated in proportion to the transverse spin induced along the y-axis by this precession (see Fig. 2). In order to discuss the strengths of our approach, we introduce a figure of merit for the limit placed on an EDM by measurements of this kind (ignoring, at present, the limitations due to noise from external field fluctuations):

$$\delta D_{Cs} \propto \frac{1}{EP_z\sqrt{N\tau_{coh}T}},$$

where N is the number of probed Cs atoms and and T is the total measurement time. For the sake of discussion, we assume a pump-probe arrangement similar to Hunter's and focus on 2°K cells, in which long holding times for Cs have already been observed. We also assume a Cs atom density of about $10^{10}/\mathrm{cm}^3$, which is consistent with present experimental achievements. For the proposed measurement, we use a conservative estimate of the spin coherence time of 20 sec[6], $P_z = 0.7$, an electric field of 5 kV/cm, and a data collection period of 220h. With these numbers, comparing with the figure of merit for the published work of Hunter[3] we expect roughly a factor of 50 improvement (they used room-temperature cells, had 16 ms coherence times, $P_z \approx 0.7$, and 4.2kV/cm fields, and took 220h of data). This improvement stems almost entirely from the longer coherence times in these cells.

Almost all aspects of this experimental geometry will be determined in the process of measuring T_1 in the vapor and tailoring the magnetic fields for the EDM measurement. In particular, the probing technique should be firmly established through the measurement of T_1, where one of our particular goals is the development of extremely low power optical probes which provide minimal relaxation (and therefore reduction of the coherence time). The combination of the photoelastic modulator with very low-noise light sources and photodetectors (possibly with the detector at 2°K), should make this possible. Experimentally establishing the existence of long coherence times is the goal of the field tailoring procedure. The primary innovation we introduce with the measurement of the EDM is the application of electric fields to the volume of the cells.

The critical problem of the electric field gradient one may apply to low temperature He cells has been investigated in detail by Meats [15]. His work, in which he used brass (possibly useful for the EDM measurement) or Niobium electrodes and measured breakdown in uniform electric fields over a wide range of densities, suggests that our figure of 5 kV/cm is very conservative. Meats also indicates that Paschen's law is obeyed quite well for He[16], permitting us to evaluate a variety of experimental geometries with confidence. We have some experience with the application of electric fields to high pressure He cells, and therefore we recognize that care in the preparation of the electrode surfaces and in the conditioning process will be necessary to achieve satisfactory results.

Systematic Errors and Noise

Many of the real strengths of our technique lie in the various advantages which are inherent to the cryogenic cell arrangement. For example, the dominant source of noise in present, room temperature cell experiments are fluctuations in the magnetic fields and their gradients. These fluctuations should be many orders of magnitude smaller with the superconducting shielding in place[13]. Noise associated with the detection of the probe radiation may also be reduced in the low temperature environment. We note that any reduction of the integrated effect of the magnetic noise over a measurement interval should result in further improvements in the EDM limit one can extract.

Perhaps the greatest challenge in placing meaningful limits on the T non-invariance through measurements of the EDM is in understanding and properly dealing with systematic errors in these experiments. Hunter discusses in some detail the various experimental techniques one uses to investigate systematic errors (use of two cells to eliminate the effects of residual magnetic fields, reversal of magnetic fields and pump helicity, etc...), and all of these can be applied to the proposed measurement. One of the most powerful tools one might use to eliminate many systematic errors is to introduce both Rb and Cs into the same cell. Rb has a much smaller sensitivity to the EDM than Cs[2], so the Rb can then serve to further correct for systematic effects associated with imperfect field reversal, magnetic field variations, and other, as yet unguessed at difficulties.

The cryogenic environment provides some important fringe benefits. For example, leakage currents may be greatly reduced in the cryogenic environment due to the reduction in temperature of the materials in which these currents would flow (*e.g.* glass [17]). The average atomic velocity is at least a factor of 10 lower in cryogenic cells, reducing the magnitude of $\mathbf{v}\times\mathbf{E}$ effects as well.

Summary

We present a proposal which identifies several clear steps culminating in a measurement of the EDM in Cs. We feel that the factor of 50 improvement indicated by the figure of merit is quite conservative. The cryogenic environment and extremely long coherence times provide a number of possible advantages and avenues to improve systematic error control and greatly reduce many of the most important sources of noise in present experiments. We also feel that this measurement may have several routes open to improve our first EDM experiments. For example, there are several possible ways to increase the electric fields in the cell, including cooling the cell until the pressure is low enough to permit very large fields to be applied. It may also be possible to ablatively load cells with Tl, isotopes of Radium, or with diatomic molecules, resulting in large increases in the sensitivity of our measurements. Finally, the non-linear Faraday effect may provide an exciting avenue to large improvements in the EDM limits we obtain from measurements in superfluid cells. In any case, because of the extraordinarily long coherence times promised by the superfluid cell, it may be the natural experimental tool to pursue ever more stringent limits for T non-invariance in the future.

4 Funding

At present, an award of $5,000 directly from the physics department at Princeton remains the only source of funding specifically for the proposed research. These funds were used to purchase a compact cryostat with optical access from AET Engineering. This cryostat has now been installed and tested, and will provide the basis for the longitudinal spin relaxation measurements we are now carrying out. We have also obtained travel funds to engage in a collaboration with the group of Prof. Yabuzaki at Kyoto University through NSF grant no: INT-9726767. Support for salaries and a base level of equipment and laboratory resources is provided through a low-energy nuclear physics grant (NSF grant no: PHY9420866, which has just been renewed, but no new grant number has been issued). The proposed research activity is not, however, directly supported by this grant. Furthermore, an NSF laser grant (NSF grant no: PHY9413901) has provided reasonable laser resources to pursue this measurement, eliminating some of the major expenses associated with initiating this research. Cryogenic and glass blowing technical assistance is also available at Princeton.

The funds required for a graduate student and some of the materials, electronics and cryogenic equipment associated with this research (for example, passive magnetic shielding, a high sensitivity lock-in amplifier, flux-gate magnetometers, etc...) are not available. Without these resources, we feel an EDM measurement is impossible at Princeton. A NIST Precision Measurement Grant should provide the level of funding required to make the proposed work a reality.

Figure 1

Figure 2

References

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Budget

Equipment	Magnetic Shielding: 3 layers of μ-metal magnetic shielding For inner diameter of shielding assembly: length: 2m, diameter: 0.4m Magnetic Shielding Company (630) 766-7800	\$12,000.00
	Photoelastic Modulator: Hinds Instruments - Model II/FS42	\$ 6,090.00
Machine Shop	Shield and Cryostat Mount Miscellaneous Expenses	\$ 3,000.00
Glass Shop	Cell Construction and Miscelleous Expenses	\$ 2,000.00
Materials & Supplies	Optics, Gas Handling Equipment Miscellaneous Expenses	\$ 1,998.75
Graduate Student	$\frac{3}{4}$ Support for Graduate Student (*Full support for academic year is* \$33,215) $\frac{1}{2}$ tuition = \$11,750 Stipend = \$21,465	\$24,911.25
	Total:	\$50,000.00