Letter of Intent

Search for an electron EDM with PbF

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The observed matter-antimatter asymmetry of the Universe is not explained by the level of CP violation that is found in *K*[1] and *B* meson decays[2,3] or that can be accommodated in the phases of the Kobayashi-Maskawa matrix [4] of the Standard Model. A compelling place to search for a new source of CP violation is in the electric dipole moment, EDM, of an elementary particle. This is particularly so because promising models for the physics beyond the Standard Model, such as SUSY models, naturally predict much enhanced moments. Thus the Standard Model prediction for the electron EDM (e-EDM) is at the 10^{-38} e-cm level [5] while SUSY predictions range from 10^{-26} to 10^{-29} e-cm [6]. The present experimental limit [7] for the e-EDM is $d_e < 1.6\times10^{-27}$ e-cm. Clearly, improving that limit by two to three orders of magnitude will either lead to discovery of dramatically new physics or drastically reduce the attractiveness of SUSY models.

The purpose of this Letter of Intent is to advise the PAC that an experiment is under study to do just that and that we intend to submit a proposal to the PAC probably in early 2007. The experiment takes advantage of the uniquely favorable characteristics of lead mono-fluoride, PbF, as first pointed out by Neil Shafer-Ray of the University of Oklahoma [8]. PbF is a relativistic, paramagnetic molecule with a single unpaired electron in the ground state. This makes the molecule reasonably calculable. PbF has a

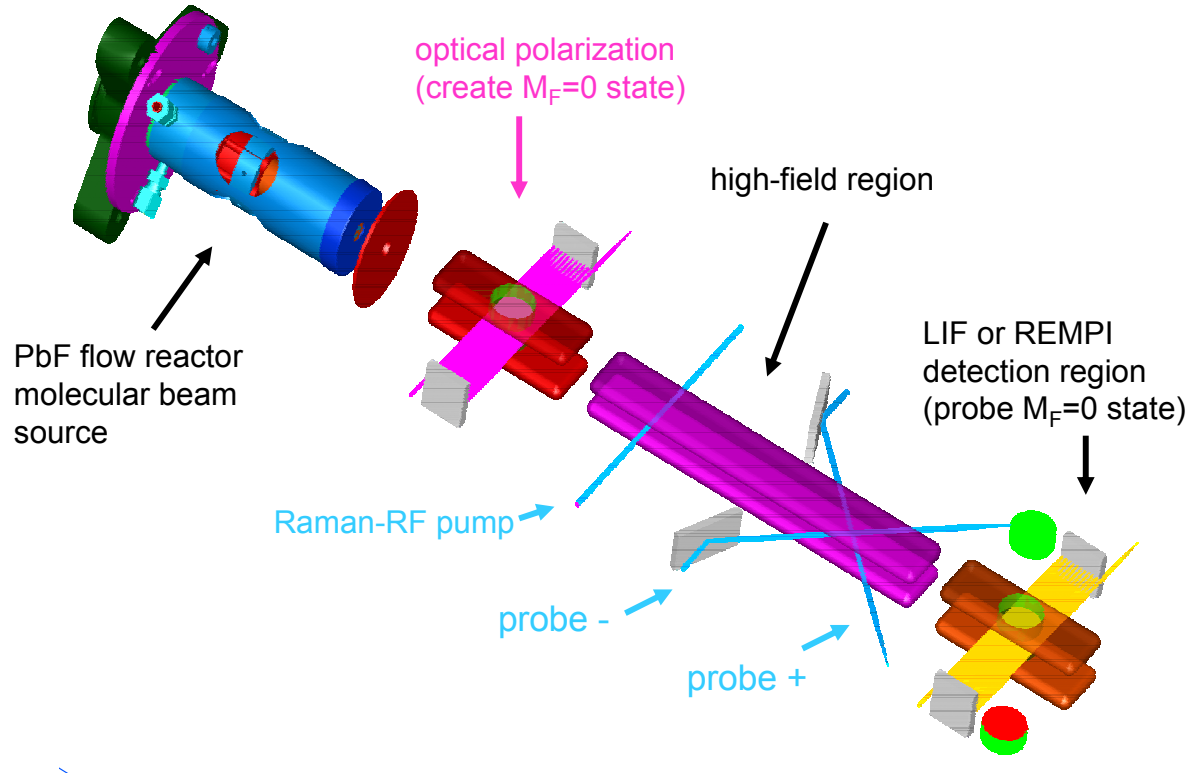

Figure 1: Schematic view of experiment. Flow reactor source is at the top left, the M_F = 0 state creation region is next, followed by the high-field region, where molecules are excited to a mixed M_F = ±1 state, evolved and then de-excited, and, finally, the detection region at the bottom right.

large molecular electric dipole that can be oriented in an external electric field to give an enhancement of the effect of an e-EDM by three orders of magnitude over that in atomic Tl, which sets the current limit. This molecular dipole also creates a large tensor shift that immunizes the molecule against one of the major systematic errors, the transverse magnetic field generated in the rest frame of the moving molecule from the Lorentz transform of the laboratory electric field. There is a rough cancellation of the magnetic moment contributions from the spin and orbital angular momentum in the ground state at zero electric field. Remarkably, the cancellation is predicted to become complete when the molecule is subjected to an electric field value near 67 kV/cm. The molecule will have a zero effective g-factor and thus no magnetic moment, limited only by the electric field regulation. Conducting the experiment near this special value will radically reduce the effect of another of the major systematic errors: time-varying stray magnetic fields. Taking advantage of the large enhancement and unique insensitivity to magnetic fields should enable a first generation experiment to improve the current limit by two orders of magnitude and ultimately may allow reaching the 10^{-32} e-cm level or lower. A comparison to other present and future EDM experiments is given in Table 1.

To measure an e-EDM, $\mathbf{d}_e$, one must detect the energy, $W = -\mathbf{d}_e \cdot \mathbf{E}$, associated with its orientation in an electric field, **E**. Since the e-EDM must be parallel to the electron's spin, this is a difference in the energy of angular momentum substates of

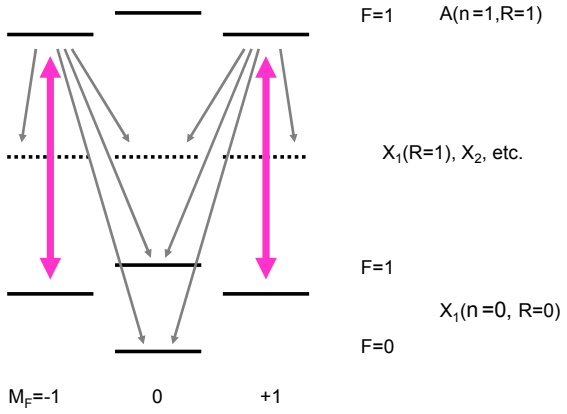

Figure 2: Schematic level diagram showing transitions to optically deplete non M_F=0 substates of the ground state. Not to scale. Magenta transitions are driven by laser at 437 nm, gray lines are decays.

opposite sign produced by an electric field along the axis of quantization. For a system with a magnetic moment, this experimental signature is also produced by a magnetic field. Maintaining the electric field value to one part in 10^5 will reduce the PbF sensitivity to magnetic field by 6 orders of magnitude relative to atomic Tl, where the current limit was set. Since the energy splitting is 3 orders of magnitude larger in PbF than Tl, the overall reduction is 9 orders of magnitude in systematic errors, assuming similar levels of background magnetic fields.

In order to determine the angular momentum substate energy splitting, W, we propose to measure the phase difference, $\Delta\Phi$, that develops over time, t, between two substates initially in phase ($W = h\,\Delta\Phi / 2\pi\, t$, where h is Planck's constant). The predicted energy splitting in PbF for an e-EDM of 10^{-29} e-cm is 0.14 mHz (in units of h) [9] and the velocity of the beam is around 200 m/s so the phase difference is 4.4×10^{-6} radians per meter of beam travel. This size phase shift can be compared to the $(-1.8\pm1.9)\times10^{-7}$ radians phase shift determination in atomic Tl that sets the current limit.

As depicted in Figure 1, the experiment would begin with a flow reactor source for the PbF, in which input fluorine gas reacts with 1100 C lead vapor. The output from this reactor is roughly 1.5×10^{17} molecules/sterad/s of PbF and 10 times that of Pb and PbF_2. (The reactor is operated in a fluorine lean mode so very little of it is emitted.) Further this flux is spread over 10^4 quantum states giving 10^{13} PbF molecules/sterad/s per quantum state.

In order to reduce the 5 orders of magnitude of undesirables, the beam passes through a state selector consisting of a skimmer and an electrostatic collector, guide and focusing element. (These are not shown in Figure 1.) Molecular states respond differently to an increasing electric field. Some states go down in energy and are thus attracted to high fields; other states go up in energy and are repelled by high fields and attracted to low field regions. The low field seeking states are the ones most readily steered (the high field seeking states end up on the electrodes.) For PbF, the most strongly low field-seeking states are among the rotational excited states with 2 units of molecular rotation, R, and it is these that will be trapped and steered. The guide terminates with focusing electrostatic optics that image the beam to the detection end of the experiment. The PbF is then optically de-excited to the no rotation (R=0) ground state with two laser photons ($2\rightarrow1\rightarrow0$), and the beam enters a region where we will de-populate the non-M_F = 0 substates of the ground state.

The ground state of PbF is referred to as the X_1 state in molecular spectroscopy notation, which means the lowest electronic excitation, X, and its lowest fine structure component, 1. The $^{208}Pb^{19}F$ state has two hyper-fine components specified by the total

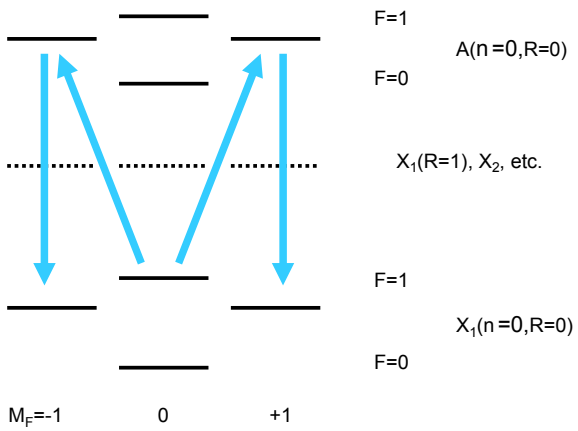

Figure 3: Schematic level diagram showing Raman transition to transfer from the M_F=0 substate of the ground state to the M_F=±1. Not to scale. Blue transitions are driven by laser at 444 nm.

angular momentum, called F in molecular physics notation. The two components are F=0 and F=1. It is the F=1 state that gives the opportunity of opposite sign substates where a phase shift can be measured and will be the focus of the experiment. At this point, the molecular beam is mainly the X_1 (R=0) state. We will optically pump the ground state with a laser directed perpendicular to the electric field and linearly polarized along it. The laser wavelength (437 nm) is chosen to drive the X_1 (n=0, R=0, F=1, M_F=±1) → A (n=1, R=1, F=1, M_F=±1), as shown in Figure 2. The A means first electronic excited state in molecular spectroscopy notation. The A state has only one fine structure component and again two hyperfine components. The n refers to the number of vibrational quanta. The A state decays to the M_F=0 levels, to other long lived levels that are not relevant and back to the M_F=±1 ones, where they are pumped again. After many cycles there is no population left in the M_F=±1 levels.

The F=1 hyperfine component of the ground state is now a pure M_F=0 state, and it next enters the high field (67 kV/cm) plates where it is nearly immune to magnetic fields. We need to prepare a coherent mixed state of the M_F=+1 and M_F=-1 substates of the F=1 component of the ground state. This defines the initial phase difference.

We do this by a two-photon Raman transition as shown in Figure 3. The laser is directed perpendicular to the electric field and linearly polarized at an angle to it so as to drive both the Δm=0 and the Δm=±1 transitions equally. The laser wavelength is chosen to be in the wings of the X_1 (R=0, F=1, M_F=±1) → A (R=0, F=1, M_F=±1) transition. The interaction time is chosen to produce a so-called π-pulse, that transfer maximum population into the mixed state. Note that there is only a single initial state so the transition amplitudes to the two substates must be treated coherently. The result is a coherent superposition of the two substates with a relative phase defined by the direction of propagation of the laser.

The prepared mixed state then evolves in time as it continues to drift in the electric field. If the two substates have identical energies, their phases will evolve identically, the phase difference remains as set. But if the substates are split by an e-EDM then an additional phase difference will develop. This means that the configuration will appear to have been generated by a laser at a different angle. This configuration can be probed by Raman pumping back to the M_F=0 substate, using two orthogonal propagation directions for the laser. Our signal is then the ratio of the M_F=0 substate intensity for the two propagation directions. Note that we are back to the M_F=0 substate so we no longer need to protect the molecule from magnetic fields by remaining at the critical electric field value. Thus we can wait to detect the M_F=0 substate until the

molecule is in a lower, more convenient magnitude field region.

Two detection methods are under consideration for the detection region. The simplest is measurement of Laser Induced Fluorescence (LIF) from excitation of the X_1 to the A state and subsequent decay to the X_2 state (700 nm). This is robust and capable of high rates but suffers from low efficiency. (A two photon excitation of the A state via the X_2 state and then detection of the blue (444 nm) de-excitation to the X_1 state might be better, albeit more complicated.) The second method is a three photon Resonant Multiple Photon Ionization (ReMPI). Again a laser drives the X_1 to A transition, a second laser pumps the A state to the B state (second electronic excitation) and a third ionizes the B state. (The second resonance step before ionization is just to reduce the chance of ionizing backgrounds in the beam.) The resulting ion and electron are accelerated to opposite field plates, depending on the direction of the electric field and counted. Attached to the electric field plates are micro-channel plate detectors that will allow coincident detection. Timing will allow discrimination against molecular dissociation and possibly isotopic selection (That will also come from the laser line width and the isotope shift of the transitions.). This is very efficient and highly selective but complex and the extension to high rates is problematic.

Of course, the angles and identical characteristics of the two probe beams cannot be guaranteed at the requisite level so the e-EDM signal will be isolated by flipping the field, which will flip its sign in each beam and leave any angle error unchanged. One can then use a four-fold ratio which cancels any probe beam variation. The phase shift expected with a 10^{-29} e-cm e-EDM for a 200 m/s beam over a 1 meter flight path is 4.4 micro-radians. A counting rate of 1 MHz will accumulate 1 sigma statistics on that size effect in less than a day. A combination of a longer flight path and beam slowing is envisioned for the ultimate limit of 10^{-32} e-cm in one day.

There is an additional systematic effect encountered in the Berkeley experiment that establishes the current limit which should be considered. When the direction of the quantization axis changes, the system will adiabatically follow, acquiring a relative phase shift even if the axis eventually returns to its original direction. This is the geometric phase effect. In the Berkeley experiment, initialization and detection of the phase were done outside the main field. As a result, the system was exposed to the non-uniformities in the fringe fields at the ends and a possible geometric phase shift was introduced. The requirement to be at the special electric field value means that we must set, evolve and probe the phase within the main field plates where the field direction is extremely well defined. Thus, the effect of the geometric phase is negligibly small.

In summary, we see that a search for an electron electric dipole moment with lead mono-fluoride presents a large and unique set of advantages:

- The EDM of the electron is more directly coupled to models of CP violation than that of the nucleon, a composite particle, with possible QCD modifications.
- PbF is a well known molecule whose spectroscopy has been studied since the 30's.
- PbF has a single unpaired electron, so the EDM enhancement can be accurately

calculated.

- PbF has a large molecular dipole, which leads to a readily oriented and very large internal field that enhances the EDM effect by 10^3 over atomic Tl.
- When subjected to an electric field near 67 kV/cm, PbF has no magnetic moment and that reduces the sensitivity to stray magnetic fields by 10^6 over Tl.
- The large molecular dipole also reduces the sensitivity to the motion-induced magnetic field by 10^6 over Tl.
- An experimental design is available which can eliminate the geometric phase effect.
- The required optical transitions are all well within current laser technology.

This effort is in a pre-proposal stage. It was originally initiated by Neil Shafer-Ray of the University of Oklahoma, seeking the infrastructure of a National Lab to stage the experiment, and a first collaboration meeting was held at BNL in February, 2005. He is working now to demonstrate experimentally the zeroing of the magnetic moment and to measure the hyperfine parameters which are only estimated from theory. This necessarily means developing the first generation source as well. The BNL-LEGS group has been simulating the experimental design in preparation for writing the final proposal. We should be in a position to submit a realistic proposal in early 2007. Present and future EDM experiments are summarized in Table 1. As can be seen, a successful proposal will inaugurate an effort with an extraordinary potential for extremely high scientific payoff.

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Table 1 – Compendium of Electric Dipole Moment Searches

System	Current Limit	Ultimate Goal	Location
Electron EDM (SUSY prediction: 10^{-26} - 10^{-29} e·cm)			
PbF	-	$<10^{-32}$	BNL
Tl	$<1.6\times10^{-27}$	-	Berkeley
PbO	-	$<10^{-29}$	Yale
YbF	$<2\times10^{-28}$	$<10^{-29}$	Sussex
HfF^{+}	-	$<5\times10^{-29}$	Colorado University
Cs, Rb	-	$<4\times10^{-30}$	Penn State
Nucleon EDM (SUSY prediction: 10^{-25} - 10^{-28} e·cm)			
^{129}Xe	-	$<10^{-30}$	Princeton
^{199}Hg	$<2.1\times10^{-28}$	$<10^{-28}$	Seattle
^{225}Ra	-	$<10^{-30}$	Argonne
n	$<6\times10^{-27}$	$<10^{-28}$	ILL Sussex
n	-	$<6\times10^{-28}$	LANL
n	-	$<10^{-28}$	SNS
p	$<5.4\times10^{-24}$	$<10^{-29}$	dEDM
D	-	$<10^{-29}$	dEDM