New Constraints on Time-Reversal Asymmetry from a Search for a Permanent Electric Dipole Moment of ^{199}Hg

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A search for a permanent electric dipole moment of ^{199}Hg atoms has yielded the null result $d(^{199}\mathrm{Hg})=(0.7\pm1.5)\times10^{-26}$ $e\cdot$cm, which improves by an order of magnitude the limits on several possible interactions that violate time-reversal symmetry. The experiment was performed with the use of optically pumped atomic oscillators to measure any shift in the NMR frequency of ^{199}Hg ($I=\frac{1}{2}$) produced by an external electric field.

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The search for a permanent electric dipole moment (EDM) of an elementary particle or atomic system is motivated by the long-standing evidence of time-reversal asymmetry (T nonconservation) in the CP-nonconserving decay $K_L^0\rightarrow 2\pi$.[1] A permanent EDM can exist only in the presence of a T-nonconserving force. The most accurate EDM measurements to date have yielded the null values for the neutron,[2] $|d(n)|<2.6\times10^{-25}$ $e\cdot$cm, and for the xenon atom,[3] $d(\mathrm{Xe}^{129})=(0.3\pm1.1)\times10^{-26}$ $e\cdot$cm. Because the sensitivity to T-nonconserving interactions within atoms rises rapidly with atomic number, we began a new EDM experiment with mercury atoms ($Z=80$).[4] We report here the first result, $d(^{199}\mathrm{Hg})=(0.7\pm1.5)\times10^{-26}$ $e\cdot$cm, which improves the limits on electron-nucleon T-nonconserving interactions[5–8] by an order of magnitude, and sets limits on purely hadronic T-nonconserving interactions comparable to those from the neutron experiments.[4,9–11]

The ground state of ^{199}Hg, having a $^1S_0$ electronic configuration and nuclear spin $I=\frac{1}{2}$, is completely specified by the nuclear-spin projection quantum number $m_I=\pm\frac{1}{2}$. In external electric and magnetic fields $\mathbf{E}$ and $\mathbf{B}$ the Hamiltonian is

$$H=-(d\mathbf{I}\cdot\mathbf{E}+\mu\mathbf{I}\cdot\mathbf{B})/I, \quad (1)$$

where d and μ are the electric and magnetic dipole moments.

We compare the nuclear Larmor precession frequencies in two adjacent cells, with $\mathbf{E}$ parallel to $\mathbf{B}$ in one cell and antiparallel in the other. It follows from Eq. (1) that when $\mathbf{E}$ is reversed simultaneously in both cells, the shift in Larmor frequency between the two cells is $\delta\omega_0=8dE/\hbar$, while the effect of drifts in $\mathbf{B}$ are largely canceled.

In our experiment each cell is operated as an optically pumped light-absorption oscillator[12] based on the nuclear-spin Larmor precession frequency of ^{199}Hg. The overall geometry of the experiment is shown in Fig. 1. The oscillators use the 253.7-nm resonance light from a ^{204}Hg electrodeless microwave discharge lamp, and operate as follows. The circularly polarized light propagates through the cells along $\hat{\mathbf{y}}$. At an angle of 45° is a static magnetic field $\mathbf{B}_0$, while pointing perpendicular to $\hat{\mathbf{y}}$ and $\mathbf{B}_0$ is an oscillating magnetic field B_x whose frequency, $\omega=18.0$ Hz, is close to the Larmor frequency, ω_0.

Through optical pumping the nuclear spins become polarized.[13] This polarization $\mathbf{P}$ is driven by B_x to precess at frequency ω in a cone about B_0. The projection of $\mathbf{P}$ along the light axis $\hat{\mathbf{y}}$ has a static and an oscillating component, causing the light to have a modulation at ω. For $\omega=\omega_0$, the modulation will be exactly in phase with B_x. For small deviations of ω from ω_0, the modulation will have a phase shift $\phi=T_2(\omega-\omega_0)$, where T_2 is the transverse relaxation time of the polarization. This phase shift measures the frequency shift of interest. The phase shifts from each cell are added; this added signal is integrated and used to stabilize the magnetic field B_0 by adjustment of the current to the magnetic field coil. The difference between the phase shifts of each cell then becomes the output signal that is sensitive to an EDM.

An electric field of 5–11 kV/cm is applied to each cell, and its polarity is reversed every 4 min. The EDM signal channel and other important data channels are measured by a computer every 0.5 s, averaged for 15 s, and

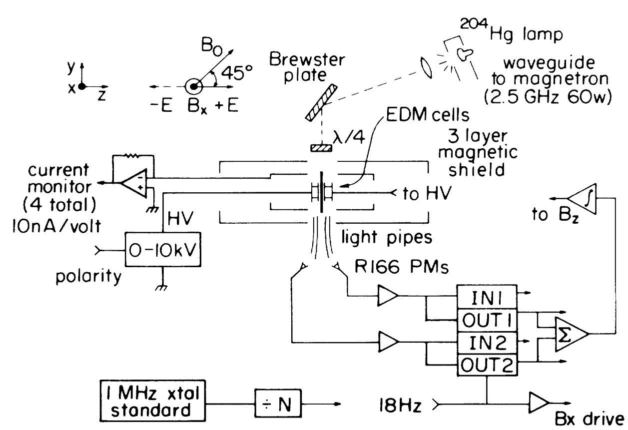

FIG. 1. Schematic of the experimental apparatus.

recorded.

The mercury EDM cells are 1-cm-long by 2.6-cm-diam fused silica (Suprasil I) cylinders with end plates made from Suprasil I disks. The inner surface of the disks are made conductive with tin oxide, according to the recipe by Coghill.[14] The cylinders are coated with Aquasil siliconizing fluid and the electrodes are coated with a thin layer of wax (dotriacontane). These coatings greatly increase the spin lifetime at room temperature. The cells are glued together with Varian Torr-Seal high-vacuum epoxy, and filled with natural mercury (16% Hg^{199}) at the 0 °C vapor pressure, along with 185 Torr of nitrogen buffer gas to prevent electrical breakdown. We have verified that the electric field appears undiminished inside the cells by measuring the excited-state Stark splitting. The Hg slowly disappears into the wax when the cell is irradiated with the ultraviolet pumping light. Each time that the wax is remelted most of the Hg returns.

The cells are held next to each other inside a sealed aluminum vessel having a slight overpressure of SF_6 or freon gas, and quartz windows for the pumping light. High voltage (HV) is applied to the outer electrode of each cell. Two separate ground planes between the cells allow the current across each cell to be monitored independently. Three coils provide the static and oscillating magnetic fields.

The cells and coils are located inside three concentric cylindrical Molypermalloy magnetic shields, whose transverse shielding factor is $\approx 10^4$. Two polished aluminum tubes serve as light pipes to collect the light transmitted through the cells and direct it to solar-blind photomultiplier (PM) tubes. The signal from the photomultiplier is amplified, filtered, and phase detected by switching-type phase-sensitive detectors using the oscillating field drive as a reference. The in-phase and quadrature reference signals, and the oscillating field drive signals, are all generated digitally from a common clock for best control of phase shifts.

T_2 for the ^{199}Hg ranges from 100 to 800 s for different cells. The pumping light shortens the ^{199}Hg lifetime to 60–100 s. The amplitude of the oscillating field was set at ≈ 19 μG so that the in-phase signals were 0.7 times their maximum values.

The sensitivity of the apparatus to the frequency difference between the two cells, $\nu_a = \nu_1 - \nu_2$, is calibrated experimentally.[15] Letting $V_i^{\rm out} = s_i \nu_i$ be the quadrature signal from cell i, and using the fact that $V_1^{\rm out} + V_2^{\rm out} = 0$ (as a result of the magnetic field servo), we find that

$$V_1^{\rm out} - V_2^{\rm out} = [4s_1s_2/(s_1+s_2)]\nu_a = \xi^{-1}\nu_a. \qquad (2)$$

To calibrate the system we apply a known magnetic field difference ($\Delta B = 140$ nG) using current through a single loop of wire concentric with the cells, and measure $\Delta(V_1^{\rm out} - V_2^{\rm out})$. Thus, $\xi = \gamma_{199}\Delta B/\Delta(V_1^{\rm out} - V_2^{\rm out})$, where γ_{199} is the gyromagnetic ratio of ^{199}Hg. Typical values for ξ are between 0.25 and 1.50 μHz/mV. The value for ξ found in this way is consistent with ξ found by measurement of s_1 and s_2 separately.

We also take into account the change in sensitivity during a run due to the change in Hg density by assuming that $s_i(t) \propto V_i^{\rm in}(t)$, the in-phase signal from cell i. This model was verified by measurement of $s(t)$ and $V_i^{\rm in}(t)$ at the beginning and end of several runs.

Our data consist of two separate sets, each set containing about fifteen runs of between 1 and 15 h duration. In data set 1 a computer records the in-phase and quadrature signal of the transmitted light from each cell, as well as the total light transmitted by each cell, the magnetic field correction signal, and the sign of the HV. The data in the first two minutes after each HV reversal are discarded to allow time for the system to equilibrate. Each datum point is an averaged value for the remaining time for a given applied voltage. Error bars for the quadrature channels are determined by the scatter during this interval.

There are drifts in ν_a of 50–200 μHz during a run, caused by changes in lamp intensity and Hg density. A multiple-regression routine was used to remove these effects by the subtraction of data correlations with the simultaneously monitored channels described in the preceeding paragraph. The fit also includes terms to remove any static phase offsets in the electronics or misalignments among $\mathbf{B}_0$, $\mathbf{B}_x$, and optical fields. A curved baseline (second order in time) is included to account for very low-frequency drifts.

Data set 2 includes several refinements. By adding 10–50 Torr of ammonia vapor to the Hg cells we were able to fabricate cells in which the Hg density decreased 3–5 times more slowly than without the ammonia. A typical run was 10–12 h in data set 2. We also began

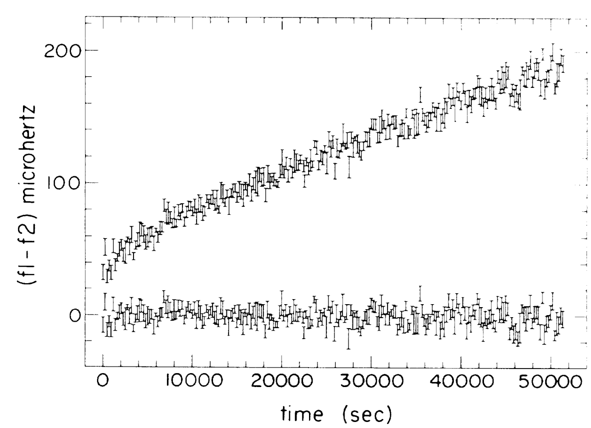

FIG. 2. Raw data for the difference in frequency between two cells (sloping curve), and residuals of regression for a typical run (run number 25). Each point corresponds to the average over one HV configuration.

taking data with 0 electric field applied in the sequence $(0,+,0,-,\ldots)$, with equal intervals (4 min) for each field configuration. In addition to the channels monitored in data set 1, we monitored the room temperature and the incident light intensity. These parameters were also included in the multiple-regression analysis.

The plot in Fig. 2 shows typical data. The plot demonstrates that most of the drift is accounted for by the regression analysis.

We look for an EDM signal in two ways. In the first, a term linear in the applied electric field (HV) is included as an additional term in the linear regression analysis. In the second technique a regression analysis with no HV term is carried out first, and then the residuals are analyzed for an HV dependence as follows. Each point in the residuals is multiplied by the sign of the HV corresponding to that point. These numbers are averaged and a scatter is determined for each run. The two techniques resulted in the same EDM values within their error bars, but the second technique gave a slightly better value for χ^2. We illustrate the results for the second technique in Fig. 3.

We consider several possible systematic effects which might mimic or mask a true EDM. There is a frequency shift[13] (light shift) due to the circularly polarized pump light of about 100 μHz. We took data with both senses of circular polarization and obtained identical results to within the statistical uncertainties. We conclude that there is no significant systematic error due to the light shift, or more generally, correlated with the sense of circular polarization. In data set 2, we analyzed the data to detect any frequency shift quadratic in $\mathbf{E}$ (arising from strains in the fused silica, for example). No E^2 shifts were detected at a level 5 times smaller than our EDM uncertainty, and the feedthrough of an E^2 shift into the EDM channel would be reduced by the excellent symmetry ($\approx 0.2\%$) between the plus and minus potentials on the HV supply. Leakage currents across the Hg cells were kept below 20 pA, which would produce frequency shifts well below our quoted uncertainty.

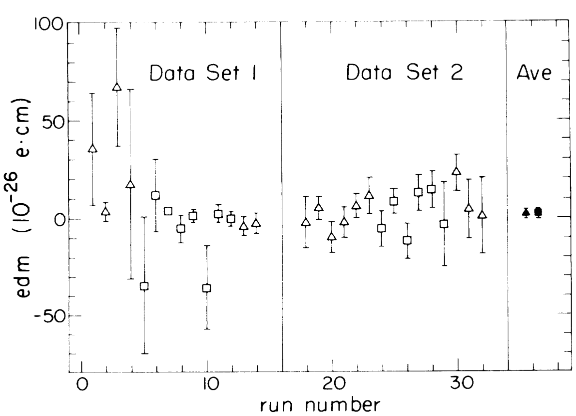

FIG. 3. Experimental results for the average EDM for each run. Triangles (squares) denote runs with right (left) circularly polarized light.

When the electric field is reversed there is a current surge to the cells of about 30 nA. The surge decays away to its steady-state value in 15 s. If all of the charging current happened to flow in a loop around the cells, it is conceivable that this could lead to a transient frequency shift with the symmetry of an EDM signal. Another possible systematic effect which could have the same signature arises from the fact that the HV is reversed with the use of a solenoid which produces a small but measurable magnetic field on the outside of the shields (≤ 1 mG). In data set 1, the solenoid is on when the HV is positive and off when the field is negative. In data set 2, the solenoid is on during the cycle $(0,+)$ and off during the cycle $(0,-)$.

In data set 2, we analyze the 0-HV periods for shifts correlated with the solenoid position or surge currents. Although no resolved shifts are seen, the statistical uncertainty is comparable to that of the EDM shifts. Therefore in data set 2 the average frequency shift correlated with 0 electric field is subtracted from the averaged shift with electric field for each run, and their statistical uncertainties are added quadratically. The final value for this data set is the weighted average of these differences. We also analyze the magnetic field correction signal for correlations with the solenoid position or surge currents, because these systematic signals should cause shifts that are primarily the same in both cells. Again no resolved shift was seen ($<4\,\mu$Hz). The ratio, R, of asymmetric (quadrature) systematic shift to common-mode (correction signal) shift is 0.05 ± 0.05.

Similarly, in data set 1, the magnetic field correction signal is analyzed for a frequency shift induced by the solenoid position or surge current. No shift was resolved. The EDM error bar for this set is the quadrature sum of the statistical uncertainty and R times the correction-signal systematic result. In summary, no evidence for any systematic error was detected. The error bar of our quoted EDM result includes the extent to which we can rule out systematic effects.

From our experimental result we obtain coupling strengths for possible T-odd electron-nucleon interactions of the form

$$H_T = iC_T(G_F/\sqrt{2})(\bar{n}\sigma^{\mu\nu}n)(\bar{e}\gamma_5\sigma_{\mu\nu}e), \tag{3}$$

$$H_S = iC_S(G_F/\sqrt{2})(\bar{n}n)(\bar{e}\gamma_5 e), \tag{4}$$

where n and e refer to nucleon and electron operators and G_F is the Fermi coupling constant. Using the results of atomic physics calculations,[7,8] we obtain

$$C_{Tn}\langle\sigma_n\rangle + C_{Tp}\langle\sigma_p\rangle = (-1.2 \pm 2.5)\times 10^{-7}, \tag{5}$$

$$C_S = (1.0 \pm 2.2)\times 10^{-5}, \tag{6}$$

where $\langle\sigma_n\rangle$ and $\langle\sigma_p\rangle$ are the expectation values for the Pauli spin operators for the neutron and proton in the

^{199}Hg nucleus. Equations (5) and (6) improve previous limits by about an order of magnitude.

The EDM of Hg is also sensitive to an intrinsic electron EDM,[7,16] d_e. We obtain $d_e = (-0.5 \pm 1.1) \times 10^{-24}$ $e \cdot$cm which is comparable to the limits set in previous atomic experiments.[17]

Hadronic T-nonconserving interactions will induce a finite Schiff moment,[10,18,19] Q_s, in an atomic nucleus. An atomic physics calculation[10] relates the EDM for ^{199}Hg to the Schiff moment of its nucleus: $d(^{199}\text{Hg}) = -4 \times 10^{-17} Q_s \; e \cdot \text{cm}$, where Q_s is in units of $e \cdot \text{fm}^3$. Our experimental result gives

$$Q_s(^{199}\text{Hg}) = (-1.8 \pm 3.8) \times 10^{-10} e \cdot \text{fm}^3, \qquad (7)$$

which constrains the strength of T-odd nucleon-nucleon interactions to the level of $10^{-2} G_F$. The bounds on various T-odd quark interactions obtainable from atomic and neutron EDM experiments have been estimated recently.[11] The above Schiff moment is expected to provide the most stringent limits to date on several classes of T-odd quark-quark interactions. Further theoretical analysis of bounds on possible quark interactions from atomic EDM limits would be helpful.

In conclusion, the comparison of the precession frequencies of two light-absorption oscillators in two different cells by optical pumping techniques has set stringent limits on the size of an EDM in ^{199}Hg, and has improved limits on possible time-reversal–asymmetric interactions in an atom by an order of magnitude. Improvements in our method would include stabilizing the ^{199}Hg density, comparing two different spin-$\frac{1}{2}$ species in the same cell, and using isotopically enriched ^{199}Hg.

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