New Limit on the Permanent Electric Dipole Moment of ^{199}Hg

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We present the first results of a new search for a permanent electric dipole moment of the ^{199}Hg atom using a UV laser. Our measurements give $d(^{199}\text{Hg}) = -(1.06 \pm 0.49 \pm 0.40) \times 10^{-28}e$ cm. We interpret the result as an upper limit $|d(^{199}\text{Hg})| < 2.1 \times 10^{-28}e$ cm (95% C.L.), which sets new constraints on $\bar{\theta}_{\text{QCD}}$, chromo-EDMs of the quarks, and CP violation in supersymmetric models.

In order for an elementary particle, atom, or molecule to have a permanent electric dipole moment (EDM) time reversal symmetry must be violated. By the CPT theorem it also implies a violation of CP symmetry. A finite EDM would give an unambiguous signal of CP violation beyond the standard model (SM), since EDMs caused by CP violation in the SM are negligible. Most extensions of the SM, such as supersymmetry, naturally produce EDMs that are comparable to or larger than present experimental limits [1]. Additional sources of CP violation are motivated by theories of baryogenesis [2].

Experimental searches for EDMs can be divided into three categories: search for the neutron EDM [3], search for the electron EDM utilizing paramagnetic atoms or molecules, the most sensitive of which is done with Tl atoms [4], and search for an EDM of diamagnetic atoms, the most sensitive of which is done with ^{199}Hg [5]. The limits set by the most sensitive experiments in each category are comparable, and they constrain different combinations of CP-violating effects [1,6].

Here we present the first results of a new search for a permanent EDM of the ^{199}Hg atom using a substantially different experimental technique, and we reduce the limit on the EDM by a factor of 4. To detect the EDM we measure the Zeeman precession frequency of ^{199}Hg nuclear spins ($I = 1/2$) in parallel electric and magnetic fields. The measurements are simultaneously performed in two cells with oppositely directed electric fields to reduce the frequency noise due to magnetic field fluctuations. A difference between the Zeeman frequencies in the two cells correlated with reversals of the direction of the electric field E is proportional to the EDM d,

$$\hbar(\omega_1 - \omega_2) = 4dE.$$

An overall schematic of the apparatus is shown in Fig. 1. Isotopically enriched ^{199}Hg vapor (92% ^{199}Hg) was contained in quartz cells with a conductive SnO coating chemically deposited on the inside surfaces to apply an electric field. The distance between the electric field plates was 11 mm. A small excess of ^{199}Hg deposited in the stem of the cells maintained the number density of ^{199}Hg atoms close to the room temperature vapor pressure. The cells also contained 450 torr of N_2 gas and 50 torr of CO gas. The walls of the cells were coated with paraffin ($C_{32}H_{66}$)

to increase the spin relaxation time. The paraffin was remelted after the cells were sealed to obtain a thin transparent coating. After such remelting the ^{199}Hg spin coherence time was typically about 300–500 sec. However, after a week of continuous UV exposure the lifetime would drop to below 100 sec. We believe this was due to damage of the paraffin coating caused by collisions with Hg atoms in the metastable 6^3P_0 state, to which they were quenched by N_2 gas. CO gas is effective in quenching ^{199}Hg atoms to the ground state. The spin coherence time could be restored by remelting the paraffin coating. The cells were placed in a sealed vessel made from carbon-filled conductive polyethylene and filled with SF_6 gas. It was located inside a three layer magnetic shield with a shielding factor of 5×10^4. A magnetic field of 15 mG was maintained inside the shields by an ultralow noise current source [7]. On a time scale of 100 sec the field was stable to 25 ppb.

Optical pumping and detection were done using a laser operating at the 253.7 nm $6^1S_0 \rightarrow 6^3P_1$ transition of Hg. To generate this wavelength we frequency quadrupled the output of a semiconductor master oscillator–power amplifier laser [8] and obtained up to 6 mW of UV light. A feedback system adjusted the current of the power amplifier to keep the light intensity constant. The intensity noise was $10^{-4}/\sqrt{\text{Hz}}$ at 10 Hz. The output of the laser was split into two beams directed perpendicular to the magnetic and electric fields. For optical pumping the light was circularly polarized and tuned to the center of the $F = 1/2$
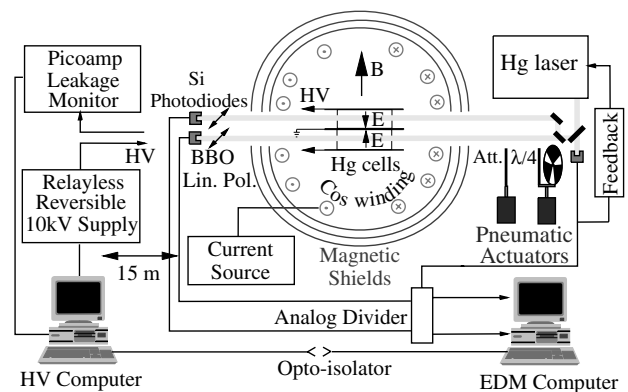

FIG. 1.   Schematic of the apparatus used to search for a permanent EDM of ^{199}Hg atoms.

hyperfine line. It was chopped at the Larmor frequency of ^{199}Hg spins with a duty cycle of 30%, building up the polarization in the rotating frame. To measure the frequency of spin precession the polarization of the light was switched to linear, the frequency detuned from resonance by 20 GHz, and the intensity attenuated to about 7 μW. Precessing ^{199}Hg spin polarization produced an optical rotation of about 60 mrad giving a 50% modulation of the intensity transmitted through BBO crystal Glan-laser polarizers oriented nearly perpendicular to the polarization of the light.

A single measurement typically consisted of a 30 sec pump phase and a 100 sec probe phase. The direction of the electric field was reversed during the pump phase. The high voltage (HV) applied to each cell was typically alternated between 10 and −10 kV. We used a solid-state relayless HV power supply located 15 m away from the magnetic shields to reduce the magnetic fields correlated with HV. We also occasionally skipped a HV reversal to guard against correlations with periodic fluctuations. The leakage currents flowing on the walls of the cells and the vessel were measured using current monitors with noise less than 0.1 pA. The vessel was designed to provide a symmetric current path for the charging and leakage currents, so the magnetic fields created by the currents were nearly orthogonal to the main magnetic field. The charging currents, which were on the order of 1 nA, did not produce an observable EDM signal even when the electric field was reversed during the probe phase. We also continuously monitored 12 other signals, including three components of the magnetic field outside of the shields, the position of the laser beam transmitted through the cell, and several laser parameters.

A typical run lasted about 24 h and consisted of several hundred individual measurements. Each of the spin precession signals was digitally filtered using a band pass fast Fourier transform filter and fit to an exponentially decaying sine wave to determine its frequency and other parameters. The scatter between successive frequency measurements was due to fluctuations of the phase and the frequency of the signal [9]. The phase noise was dominated by the signal detection noise. We verified that the whole detection system was working within 50% of fundamental shot-noise limitations [5]. In most runs the frequency noise due to magnetic field gradient fluctuations was comparable to the phase noise. The correlation between the Zeeman frequency difference and the direction of the electric field was calculated by analyzing groups of three consecutive measurements and eliminating a linear frequency drift. The statistical error for each run was determined by the actual scatter of the data.

Frequent reversals and changes were done during the experiment to check for systematic effects. We periodically reversed the data acquisition channels for the two cells and the direction of the magnetic field, which should change the sign of the EDM signal. We also frequently changed the EDM cells and their orientation in the vessel.

In addition, the paraffin in the cells was remelted and the outside surfaces cleaned each time the cells were changed, which would likely change the path of the leakage currents. Over the course of the experiment we used two different vessels and changed other components of the setup. Figure 2 shows the results of all EDM runs. The weighted average of all data gives $d(^{199}\text{Hg}) = -(1.06 \pm 0.49) \times 10^{-28}e$ cm. We do not observe any excess data scatter between runs due to changes during the experiment and the χ^2 per degree of freedom is equal to 0.95. The statistical error corresponds to a frequency difference between the two cells of 0.4 nHz, a factor of 5 smaller than in the previous experiment [5].

We looked for systematic effects by changing the operating parameters of the experiment, looking for correlations among different parameters, and exaggerating certain imperfections. The leakage currents are a potentially serious source of systematic errors because they can produce magnetic fields that are correlated with the electric field and mimic an EDM signal. It should be noted that only leakage currents flowing in a helical path around the cell will contribute to first order. Figure 3 shows a scatter plot of the EDM signal vs the leakage current in one of the cells. No statistically significant correlation was observed. The average cell leakage currents were about 0.6 pA. From the error on the correlation slope we can set a limit on the contribution of the leakage currents to the EDM signal of $0.14 \times 10^{-28}e$ cm. We estimate the error more conservatively by calculating the magnetic field created by a leakage current making one complete loop around the cell. This rather unlikely path would give an average EDM signal of $0.25 \times 10^{-28}e$ cm. A total of four vapor cells were used in the experiment in various pairs. The right panel of Fig. 4 shows that the EDM data taken with each cell are consistent. Note that if a cell had a fixed helical path for the leakage current, it would produce the same false EDM signal independent of its orientation. As can be seen in Fig. 3, the leakage currents were sometimes negative.
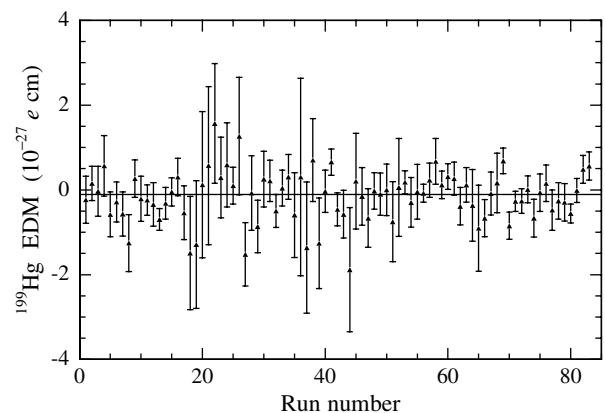

FIG. 2. ^{199}Hg EDM signal as a function of run number. The solid line shows the average of the data. Runs with larger errors were done in nonoptimal configurations.
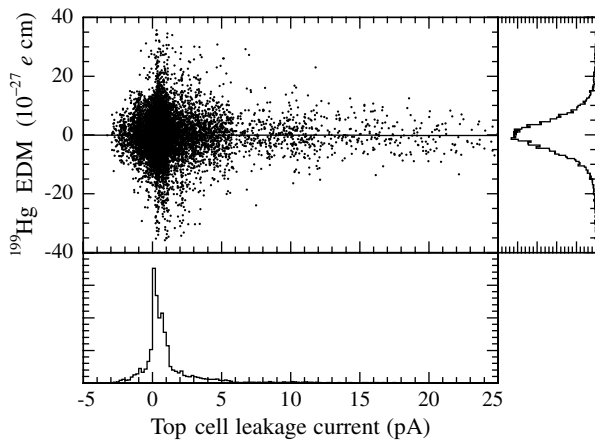
FIG. 3. Correlation between the leakage current and the EDM signal. Histograms of the leakage current and the EDM data are also shown. The solid line is a linear fit giving a correlation of $(-0.4 \pm 2.0) \times 10^{-29} e$ cm/pA.

We believe this effect was due to changes in the mutual capacitance caused by redistribution of charges on HV insulators. If the HV was not reversed for a long time, the leakage currents became positive and approached a steady state value of about 0.1 pA.

We looked for correlations with the electric field of 30 other variables, such as monitored signals and fitting parameters, and found no statistically significant correlations. Using random fluctuations of the variables we determined the cross correlation between each of them and the EDM signal $\omega_1 - \omega_2$. In this way we set upper limits on false EDM signals coming from cross correlations. All these limits are 10 to 100 times smaller than our statistical error. For the positive direction of the magnetic field the average EDM signal was $d(B+) = -(1.78 \pm 0.70) \times 10^{-28} e$ cm and for the negative direction $d(B-) = -(0.36 \pm 0.69) \times 10^{-28} e$ cm. The two results are within 1.4σ of each other. A systematic effect that does not reverse with the magnetic field would show up in the difference but cancel in the average of the two results.
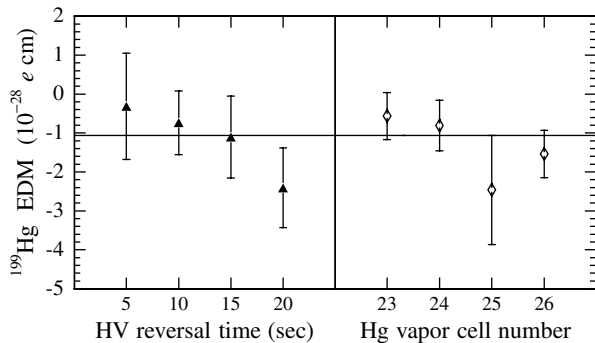

FIG. 4. The left panel shows the dependence of the EDM signal on the HV reversal time. The right panel shows the EDM signal obtained with each of the EDM cells. The solid line is an average of all data.

To study possible frequency shifts due to magnetization of the magnetic shields caused by the charging currents, we varied the high voltage reversal time from 5 to 20 sec. The dependence of the EDM signal on the HV reversal time, shown in the left panel of Fig. 4, is not statistically significant. We did not resolve any correlations of the individual Larmor frequencies ω_1 and ω_2 with the electric field outside of their error bars, which are a factor of 6 larger than the statistical error on $\omega_1 - \omega_2$.

We looked for effects proportional to E^2 in separate runs by applying the HV to only one of the two cells and alternating it between 0 and ± 10 kV. The quadratic frequency shift was less than 2 nHz. We checked that the electric field in the cells was uniform and reversible with an accuracy of 1.5% [8], which limits the effect of reversal imperfections to less than $7 \times 10^{-30} e$ cm. Although the average velocity of the atoms in the cell is equal to zero, residual $v \times E$ effects [4] can exist if the surface relaxation on the walls is asymmetric. We looked for these effects by taking data with the magnetic field intentionally misaligned by 5° from the electric field. No effects were seen at the level of $1.5 \times 10^{-28} e$ cm, which can be used to constrain this effect to less than $0.3 \times 10^{-28} e$ cm in a magnetic field aligned within 1° relative to the electric field. Among various frequency shifts caused by the probe light the most significant is due to the magnetic dipole and electric quadrupole transitions in an electric field [10]. This effect is odd in the E field and can mimic an EDM signal. It is suppressed to about $3 \times 10^{-30} e$ cm because the laser beam is directed perpendicular to the magnetic field and detuned far from resonance.

In summary, no statistically significant systematic effects that mimic an EDM signal were observed, although in several cases our systematic studies were limited by statistics. We estimate the total systematic uncertainty to be $0.40 \times 10^{-28} e$ cm by adding in quadrature the limits on systematic effects due to the leakage currents, the $v \times E$ effect, and other miscellaneous effects. Thus we obtain $d(^{199}\text{Hg}) = -(1.06 \pm 0.49 \pm 0.40) \times 10^{-28} e$ cm and interpret the result as an upper limit on the ^{199}Hg EDM $|d(^{199}\text{Hg})| < 2.10 \times 10^{-28} e$ cm (95% C.L.).

This limit can be used to place new constraints on hadronic and semileptonic CP-violating effects that are

TABLE I. Summary of limits (95% C.L.) set by the ^{199}Hg EDM and other experiments on model-independent and "naturalness" parameters.

Parameter	Limit from ^{199}Hg	Best other limit		Ref.
$\bar{\theta}_{\text{QCD}}$	1.5×10^{-10}	6×10^{-10}	n [3]	[6,11]
$\tilde{d}_d$ (cm)	7×10^{-27}	1.1×10^{-25}	n [3]	[6,12]
C_T	1×10^{-8}	5×10^{-7}	TlF [13]	[14]
C_S	3×10^{-7}	4×10^{-7}	Tl [4]	[14]
$\varepsilon_q^{\text{SUSY}}$	2×10^{-3}	1×10^{-2}	n [3]	[1]
$\varepsilon^{\text{Higgs}}$	$0.4/\tan\beta$	$0.7/\tan\beta$	Tl [4]	[1]
x^{LR}	1×10^{-3}	1×10^{-2}	n [3]	[1]

summarized in Table I. The EDM of the ^{199}Hg atom is proportional to the Schiff moment of the ^{199}Hg nucleus S, which is a measure of the difference between the distributions of the electric charge and electric dipole moment in the nucleus. Using a Hartree-Fock calculation for Hg atomic wave functions [15] and a simple nuclear shell model [14,16], the Schiff moment was calculated with an uncertainty of about 30%–50%: $d(^{199}\text{Hg}) = -3.1 \times 10^{21} S$ cm^{-2} [14]. The largest contribution to the Schiff moment comes from a CP-violating nucleon-nucleon interaction $\xi G_F (\bar{p} p)(\bar{n} i \gamma_5 n)/\sqrt{2}$. It was calculated in [16] using Woods-Saxon potentials and neglecting many-particle correlations. The result is $S = -1.8 \times 10^{-7} \xi e$ fm^3 with an uncertainty of about 50%. Possible enhancements of the Schiff moment due to collective octupole nuclear excitations have been considered recently in [17], although no definite estimates exist. As shown in [6,18], the CP-odd nucleon-nucleon interaction is dominated by π^0 exchange and is proportional to the pion-nucleon CP-odd coupling constant $\bar{g}_{\pi NN}$.

A limit on $\bar{g}_{\pi NN}$ can be used to directly constrain the CP-violating QCD vacuum angle $\bar{\theta}_{\text{QCD}}$ [19]. We obtain $|\bar{\theta}_{\text{QCD}}| < 1.5 \times 10^{-10}$, improving the limit set by the neutron EDM [3,11] by a factor of 4. We also set a limit on a linear combination of quark chromo-EDMs [6],

$$e|\tilde{d}_d - \tilde{d}_u - 0.012\tilde{d}_s| < 7 \times 10^{-27} e \text{ cm}.$$

Its limit can be compared with a constraint on a different combination of EDMs and chromo-EDMs set by the neutron EDM experiment [3,12],

$$|e(\tilde{d}_d + 0.5\tilde{d}_u) + 1.3 d_d - 0.3 d_u| < 1.1 \times 10^{-25} e \text{ cm}.$$

In most extensions of the SM, including supersymmetry, EDMs, and chromo-EDMs of the quarks have comparable size [1]. We also place new constraints on semileptonic CP-violating parameters C_S and C_T, which are significant for certain multi-Higgs models [20].

In addition to the model-independent constraints discussed above, one can set limits on specific CP-violating parameters in various extensions of the SM. For example, in the minimal supersymmetric SM the limit on the ^{199}Hg EDM can be used to set tight constraints on a linear combination of two CP-violating phases [6] and exclude a large fraction of the parameter space that would be allowed by other EDM experiments [21]. In Table I we give only general limits for naturalness parameters, as defined in [1], for supersymmetric, multi-Higgs, and left-right symmetric models. For example, in supersymmetry $\varepsilon_q^{\text{SUSY}}$ would be close to unity if the masses of the sypersymmetric particles were on the order of 100 GeV and CP-violating phases were on the order of unity.

In conclusion, we have presented the results of a new search for a permanent electric dipole moment of ^{199}Hg atoms, improving the previous limit by a factor of 4. We have set new limits on $\bar{\theta}_{\text{QCD}}$, quark chromo-EDMs, and CP violation in various extensions of the standard model. We are presently upgrading the experiment and plan to improve the statistical sensitivity by at least a factor of 2.

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