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Search for a Coupling of the Earth's Gravitational Field to Nuclear Spins in Atomic Mercury

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We have measured the ratio of nuclear spin-precession frequencies of ^{199}Hg and ^{201}Hg atoms for two orientations of magnetic field relative to the Earth's gravitational field. We find that the spin-dependent component of gravitational energy is less than 2.2×10^{-21} eV, a substantial improvement over previous limits. Our result provides a test of the equivalence principle for nuclear spins, and sets limits on the magnitude of possible scalar-pseudoscalar interactions which would couple to the spins.

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We report here the results of an experimental search for an interaction of the form $\boldsymbol{\sigma}\cdot\hat{\mathbf{r}}$, where $\boldsymbol{\sigma}$ is the spin operator for the nucleus of a mercury atom, and $\hat{\mathbf{r}}$ is a vector pointing towards the center of the Earth. Such an interaction violates both parity and time-reversal symmetry and could result, for example, from a breakdown of the equivalence principle for spin-polarized matter. It could also arise as a consequence of a new macroscopic-range interaction.

The equivalence between the gravitational and inertial masses of unpolarized matter has been verified to high precision. Very little is known, however, about the microscopic properties of gravity, and there remains the possibility that sensitive experiments will uncover evidence for symmetry violations or other new phenomena associated with this interaction. In particular, the notion that polarized objects may violate the equivalence principle through a coupling of intrinsic spin to gravity has been considered by a number of authors [1]. Recently, an electron spin-polarized test body was weighed in search of a $\boldsymbol{\sigma}\cdot\mathbf{g}$ force, where $\mathbf{g}$ is the local gravitational acceleration [2]. No difference in acceleration between spin-up and spin-down electrons was found at the $0.01\mathbf{g}$ level. Previously, it had been found [3] that the difference in energies between spin-up and -down deuterium atoms in the Earth's gravitational field was less than 2×10^{-19} eV. Finally, a study of the gravitational coupling of ^{9}Be$^+$ nuclear spins has recently been completed [4]. Our result provides a more stringent limit on the size of the gravitational dipole moment of a spin-polarized atomic system.

The $\boldsymbol{\sigma}\cdot\hat{\mathbf{r}}$ interaction could also arise from a new interaction coupled to something other than mass, as has been discussed extensively in connection with recent experiments to detect a fifth force [5]. The question of the spin dependence of such an interaction remains open, as most fifth-force experiments use unpolarized test bodies. In particular, a recent model [6] shows that axions (or any particle with mixed scalar-pseudoscalar couplings) may generate a monopole-dipole potential between two point objects of the form

$$V_{12}(\sigma,\mathbf{r})=\frac{\hbar}{m_1c}\frac{(g_P)_1(g_S)_2}{8\pi}\left[(\sigma_1\cdot\hat{\mathbf{r}})\left(\frac{1}{\lambda}+\frac{1}{r}\right)\right]\times\frac{\exp(-r/\lambda)}{r}, \quad (1)$$

where $(g_P)_1$ and $(g_S)_2$ are the pseudoscalar and scalar coupling constants of objects one and two, respectively, $\lambda=\hbar/m_ac$ is the interaction range (m_a is the axion mass), and $\mathbf{r}$ is the relative separation of the objects. For $m_ac^2\leq10^{-6}$ eV, the Earth may act as a source to interact with atomic or nuclear spins through Eq. (1).

Our experiment concerns two ground-state mercury isotopes contained in the same cell. Since Hg has a $^1S_0$ electronic configuration, the ground state is fully described by the nuclear-spin polarization. In the presence of a magnetic field, we compare the nuclear-spin-precession frequencies of ^{199}Hg ($I=\frac{1}{2}$) and ^{201}Hg ($I=\frac{3}{2}$) (I is the nuclear spin). A possible new spin-dependent interaction will in general couple to a different linear combination of orbital and intrinsic spin angular momenta than that which gives rise to the nuclear magnetic moment. Ignoring for the moment the quadrupole interaction of the spin-$\frac{3}{2}$ ^{201}Hg nucleus, we generalize the interaction Hamiltonian for each isotope to

$$\mathcal{H}=-g_I\mu_N\mathbf{I}\cdot\mathbf{B}+\mathcal{A}\epsilon\mathbf{I}\cdot\hat{\mathbf{r}}/|I|, \quad (2)$$

where $\mathbf{B}$ is the magnetic field, g_I is the nuclear g factor, and μ_N is the nuclear magneton. $\mathcal{A}$ is the strength of the proposed interaction, $\hat{\mathbf{r}}$ points vertically downward, and ϵ is a model-dependent projection of the relevant intrinsic spin operator [$\boldsymbol{\sigma}_1$ in Eq. (1)] assumed to lie along the nuclear angular momentum vector, $\mathbf{I}$. For $\mathcal{A} \ll \mu_N B$, the energy difference between neighboring Zeeman levels becomes

$$\nu_{199} = -g_{199}\mu_N B + \mathcal{A}\epsilon_{199}\cos\phi\,,$$
$$\nu_{201} = -g_{201}\mu_N B + \mathcal{A}\epsilon_{201}\cos\phi\,, \qquad (3)$$

where ϕ is the angle between $\hat{\mathbf{r}}$ and $\mathbf{B}$. Experimentally, we find that $g_{201}/g_{199} = -0.369\,139$. In our case the interaction of the spin-$\frac{3}{2}$ ^{201}Hg nuclei with the cell walls produces a resolved quadrupole splitting, $\nu_q = 0.050$ Hz, in the observed spectrum [7,8]. As described below, we can average the three experimentally observed frequencies to extract the quantity ν_{201} necessary for the analysis.

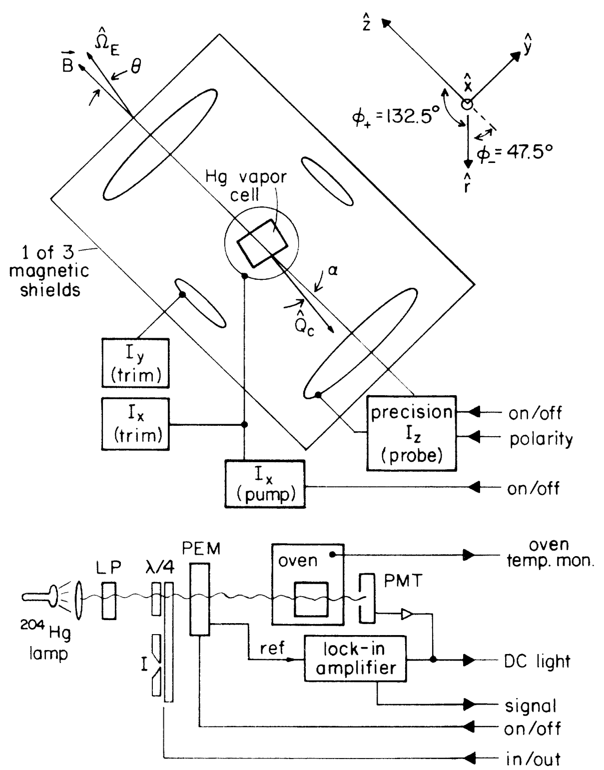

FIG. 1. Schematic of the apparatus. Abbreviations are defined as follows: LP, linear polarizer; λ/4, quarter-wave plate; I, adjustable iris; PMT, photomultiplier tube; PEM, photoelastic modulator. Arrows on the right-hand side of the figure indicate control lines from, and data lines into the computer (not shown). I_x and I_y "trim" current sources are designed to cancel residual transverse magnetic fields in the vicinity of the cell. The I_x "pump" supply is disconnected during the precession phase of the experiment, while a highly stable current supply, I_z, is turned on with a given polarity. $\cos\phi_+$ and $\cos\phi_-$ represent the projection of the unit vector $\hat{\mathbf{r}}$ along the quantization axis for these two probe field polarities. $\hat{\boldsymbol{\Omega}}_E$ refers to the axis of the Earth's rotation while $\hat{\mathbf{Q}}_C$ is the effective cell quadrupole axis.

From Eqs. (3) it is clear that as long as $\epsilon_{201}/\epsilon_{199} \neq g_{201}/g_{199}$, the ratio $\mathcal{R} = |\nu_{201}|/|\nu_{199}|$ will depend on $\mathcal{A}$, and is insensitive to changes in $\mathbf{B}$. Let $\mathcal{R}_+$ ($\mathcal{R}_-$) represent the magnitude of this ratio for $\mathbf{B}$ pointing along $+\hat{\mathbf{z}}$ ($-\hat{\mathbf{z}}$). We then define

$$\Delta \equiv \mathcal{R}_+ - \mathcal{R}_- = (2\mathcal{A}/\nu_{199})(\epsilon_{201} - \mathcal{G}\epsilon_{199})\cos\phi\,, \qquad (4)$$

where $\mathcal{G}$ is the ratio of ^{201}Hg to ^{199}Hg g factors ($\mathcal{G} = -0.369\,139$). Setting $\epsilon' = \epsilon_{201} - \mathcal{G}\epsilon_{199}$, values for the quantity $\mathcal{A}\epsilon'$ can easily be derived from the measured quantities, Δ, ν_{199}, and $\cos\phi$.

Much of the apparatus used for this measurement has been described previously [8,9]. The vapor cell containing both isotopes is located at the center of three concentric layers of magnetic shields which reduce the ambient magnetic field at the cell to less than 20 μG. Coils located within the shields allow the application of small magnetic fields (< 10 mG) for each phase of the experiment. Figure 1 is a schematic diagram of the apparatus with relevant axes and angles shown. The shields are oriented so that the axis of the concentric cylinders points along the Earth's rotation axis (making $\phi_{(+)} = 132.5°$ for our local latitude). By choosing the precession magnetic field to point in the same direction (defined as $\hat{\mathbf{z}}$) we insure that the angles θ and α are near zero, and systematic errors due to axis misalignment are quadratic in nature (see below).

Each measurement consists of a "pump" and "probe" cycle. The two Hg isotopes are optically pumped with circularly polarized light, either σ_+ or σ_-, directed along a small magnetic field B_x. Next the pump field is turned off and a probe field is switched on along $+\hat{\mathbf{z}}$ or $-\hat{\mathbf{z}}$. The incident light intensity is reduced, and a photoelastic modulator (PEM) rapidly varies the polarization of the incident light between σ_+ and σ_- during the 200-s probe interval. The absorption of light by the precessing atoms is therefore modulated at both the PEM and Larmor precession frequencies. The transmitted light is collected and demodulated at the 42-kHz PEM frequency. A typical four-frequency free-precession signal is shown in Fig. 2.

For the purpose of this search, the direction of the quantization axis for precession was switched from $+\hat{\mathbf{z}}$ to $-\hat{\mathbf{z}}$ roughly once an hour, hereby reversing the sign of $\cos\phi$. While data were taken at several larger magnetic-field values, the majority of runs had $B \approx 1.1$ mG ($\nu_{199} \approx 0.83$ Hz). Data were also taken with different incident light intensities, and with both senses of pump light polarization. Finally, the entire apparatus was rotated about the $\hat{\mathbf{z}}$ axis by 180° for a second data set ("B"), and then about the $\hat{\mathbf{x}}$ axis for a third set ("C"), to search for possible systematic errors associated with the orientation of the vapor cell relative to $\hat{\mathbf{r}}$. In all, nearly 6000 individual measurements were accumulated over a period of several months.

Individual runs were fitted in the time domain by a nonlinear least-squares function containing frequencies,

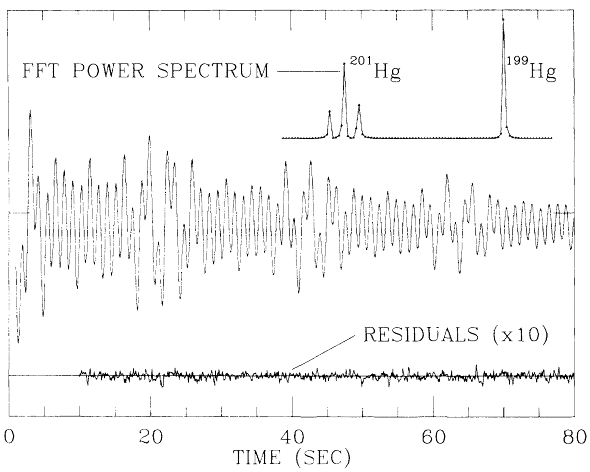

FIG. 2. Typical four-frequency free-precession signal. The sinusoidal signal is the output of the lock-in amplifier which demodulates the observed transmitted light intensity at the 42-kHz photoelastic modulator frequency. Shown below, multiplied by 10, are the residuals of a nonlinear least-squares fit of the data by a sum of decaying cosines. Typical data decayed with $1/e$ times of 60 and 80 s for ^{201}Hg and ^{199}Hg, respectively. Above the precession signal, a portion of the fast Fourier transform is shown with a solid line connecting consecutive points.

amplitudes, phases, and decay rates for the four frequencies, as well as a dc background. Typically, the uncertainty in each frequency for a single free-precession run was 10 μHz. Mean and standard deviations were computed for sets of nominally identical runs with the probe magnetic field in a given direction. Values for Δ were then computed for data sets with a given field magnitude, pump light polarization, and probe light level.

Because the apparatus is configured so that atoms precess about an axis nearly parallel (or antiparallel) to that of the Earth's rotation, the frequency ratio measured in the rotating laboratory is actually

$$\mathcal{R}^0_{\pm} = (\nu_{201} \pm \Omega_E \cos\theta)/(\nu_{199} \mp \Omega_E \cos\theta) . \qquad (5)$$

The superscript distinguishes this uncorrected ratio from $\mathcal{R}_{\pm}$ used to compute Δ in Eq. (4) above. $\Omega_E = 11.6$ μHz is the rotation rate of the Earth, and the factor of $\cos\theta$ accounts for possible residual misalignment of $\mathbf{B}$ and $\hat{\boldsymbol{\Omega}}_E$. A nonzero value for θ can arise due to mechanical misalignment of the apparatus (less than 3° here), as well as from residual magnetic fields in the x-y plane. By adding trim currents in the x and y directions, the direction of the resultant field and the magnetic shield axis were made to coincide to within 1°. We derive $\mathcal{R}_{\pm}$ from $\mathcal{R}^0_{\pm}$ assuming that $\theta = 0$; however, even if θ is as large as 3°, the resultant error in Δ is less than $4 \times 10^{-8}/\nu_{199}$(Hz). Another mechanism which could potentially affect the observed frequency ratio is the dipole light shift. The use of the PEM and the orientation of the probe magnetic field relative to the incident light made the light shifts of the Zeeman levels of both isotopes negligibly small in our case.

We now consider the effect of atom-cell surface interactions on the observed spin-precession frequencies. While such an interaction provides the dominant relaxation mechanism for ^{199}Hg, there is no evidence that it shifts the precession frequency of this spin-$\frac{1}{2}$ atom. In the case of ^{201}Hg, however, the surface-averaged electric-field gradient produces a resolved quadrupole splitting in the spectrum. The quantization axis associated with this effect is largely determined by the geometry of the cell. In our case this axis is nearly perpendicular to its broad face (nearly parallel to $\hat{\mathbf{z}}$). In lowest order, the quadrupole interaction leads to additional spin-precession frequencies equally spaced about the unperturbed ^{201}Hg Zeeman frequency. When the cell-quadrupole axis and that of the external magnetic field do not coincide, we can use second-order perturbation theory to calculate the three observed ^{201}Hg frequencies:

$$\nu_{\pm} = \nu_L \pm \nu_q \left(\tfrac{3}{2}\cos^2\alpha - \tfrac{1}{2}\right) + \frac{\nu_q^2}{\nu_L}\left(\tfrac{3}{2}\sin^2\alpha\cos^2\alpha\right) ,$$
$$\nu_0 = \nu_L + \frac{\nu_q^2}{\nu_L}\left(\tfrac{3}{16}\sin^4\alpha - \tfrac{3}{2}\sin^2\alpha\cos^2\alpha\right) . \qquad (6)$$

Here ν_L and ν_q are the unperturbed Larmor frequency and quadrupole splitting, respectively, and α is the angle between the quadrupole and magnetic-field axes. We assume (and have experimentally verified) that the surface interaction is well approximated by a single symmetric quadrupole. While the corrections approach zero for large external magnetic fields, in our experiment they can exceed 10^{-3} Hz for large values of α. Now consider a particular combination of the three observed frequencies, $\nu_a \equiv (\nu_- + 2\nu_0 + \nu_+)/4$. As can be seen from Eqs. (6), ν_a differs from ν_L by $(\nu_q^2/\nu_L)\frac{3}{32}\sin^4\alpha$. Independently, we have measured the value of α to be $\leq 3°$ [10]. Moreover, differences between values of α for $\mathbf{B}$ along $\hat{\mathbf{z}}$ and $-\hat{\mathbf{z}}$ were measured to be $< 1°$, in agreement with our earlier estimates concerning the size of stray magnetic fields. Thus, if ν_a is used for the purposes of computing $\mathcal{R}_{\pm}$ and Δ, any systematic error in Δ from residual cell-axis misalignment cannot exceed $1 \times 10^{-8}/\nu$(Hz) [11].

Using Eq. (4), for each value of Δ we computed a value for the quantity $\mathcal{A}\epsilon'$. Table I shows a summary of the data and our investigation into possible sources of systematic error. An analysis based on these results allows us to compute a final mean value and statistical uncertainty for $\mathcal{A}\epsilon'$. It also allows us to place an upper limit on the uncertainties due to systematic effects considered here. This is done by assigning a systematic uncertainty to data sets which did not include extensive reversals of a given parameter, based on any apparent systematic differences correlated with that parameter (from other data sets). We find that

$$\mathcal{A}\epsilon' = -0.12 \pm 0.14(\text{stat}) \pm 0.15(\text{syst})\ \mu\text{Hz} .$$

If we assume that the appropriate spin operator, σ_1, for the proposed interaction is the nuclear spin itself ($\epsilon_{199} = \epsilon_{201} = 1$, $\epsilon' = 1.369$), it follows that $|\mathcal{A}| < 2.2 \times 10^{-21}$

TABLE I. Summary of data and investigation of systematic errors. The first four column entries refer to the status of particular experimental parameters for a given set of data runs whose mean and statistical error are listed in the final column. Labels are as follows: (I) magnetic-field strength ("H" corresponds to $3 < |B| < 100$ mG, "L" to 1 mG); (II) pump light polarization ("+" and "−" refer to σ_+ and σ_- light, respectively); (III) probe light intensity ("H" corresponds to roughly 3 times the light intensity of "L"); (IV) cell orientation ("A," "B," and "C" refer to the apparatus orientation as described in the text). The final line contains the mean and 1σ statistical error of all data. Upper limits for systematic variation correlated with each parameter are estimated. Systematic errors are added to those data sets not including reversals or changes of some parameters. The final systematic error quoted in the text includes these contributions, as well as smaller ones from possible Earth rotation-axis and cell-axis misalignment.

I	II	III	IV	$\mathcal{A}\epsilon'$ (μHz)
H/L	+/−	H/L	A	$+0.23 \pm 0.32$
H/L	+/−	H/L	B	-0.24 ± 0.24
H/L	+/−	H/L	C	-0.17 ± 0.24
H	+/−	L	A	$+0.16 \pm 0.49$
L	+/−	L	A	$+0.28 \pm 0.47$
L	+	L	B	-0.26 ± 0.39
L	+	H	B	-0.22 ± 0.30
L	+	H/L	C	-0.36 ± 0.27
L	−	H/L	C	$+0.06 \pm 0.30$
All data				-0.12 ± 0.14

eV (95% C.L.). In the nuclear shell model, the ^{199}Hg and ^{201}Hg nuclei are characterized by valence $p_{1/2}$ and $p_{3/2}$ neutrons, respectively. The spin projections for the valence neutrons alone are $\epsilon_{201} = -\epsilon_{199} = \frac{1}{3}$, or $\epsilon' = 0.21$. In this model [12], our result implies that the energy difference between a spin-up and spin-down neutron in the Earth's gravitational field is less than 2.1×10^{-20} eV (95% C.L.). Integrating over a uniform density spherical Earth as a source for the monopole-dipole interaction of Eq. (1), we can place limits on the coupling constants for a given axion range, λ. Adopting the notation of Ref. [4], we define $D \equiv (g_p)_1 (g_S)_2 / 8\pi\hbar m_1 c$ (here m_1 is the neutron mass). For $\lambda \geq 10^6$ m, our result for $\mathcal{A}$ implies that $D(^{199}\text{Hg}, ^{201}\text{Hg}) < 3.6 \times 10^{-10}$ kg^{-1}, and, using the above nuclear model, we find that $D(\text{neutron}) < 1.7 \times 10^{-9}$ kg^{-1} (both 95% C.L.). Our results for the neutron improve by a factor of 25 the analogous interpretation of data from the ^{9}Be$^+$ stored-ion spectroscopy described in Ref. [4].

Still considering the Earth as axion source, limits placed on D by our experiment become rapidly less stringent for axion ranges less than 10^6 m ($\lambda D \approx$ const down to 1 m, D grows exponentially for $\lambda < 1$ m) [13]. Yet, ultimately, laboratory measurements such as these have the most potential significance in the short-range regime (1 mm $< \lambda <$ 10 cm) where astronomical observations have not already provided more stringent constraints [14]. A straightforward extension of this experiment to search for shorter-range forces would involve placing a small amount of very dense material in close proximity to the vapor cell. By changing the position of the small source relative to the external magnetic field and cell, a sensitive test of the above model for ranges of order 1 cm would be possible.

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[10] Another combination of the three frequencies, $(\omega_+ + \omega_-)/2 - \omega_0$, can be used to determine α independently (see Ref. [8]).

[11] If we had chosen instead to orient the precession axis vertically, along $\hat{\mathbf{r}}$ (so that $\cos\phi = 1$), the quoted uncertainties in θ and α would then have led to at least an order of magnitude larger uncertainty in Δ.

[12] We note that this crude model, which ignores multiparticle contributions to the nuclear ground state, incorrectly predicts the g-factor ratio ν_{201}/ν_{199} to be -1.0.

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