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New Limits on Spatial Anisotropy from Optically Pumped ^{201}Hg and ^{199}Hg

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The nuclear magnetic resonance frequencies of ^{201}Hg ($I=\frac{3}{2}$) and ^{199}Hg ($I=\frac{1}{2}$) have been compared by use of optically pumped atomic light-absorption oscillators to see if the relative frequencies depend on the orientation of the quantization axis in space. The null result $\delta\nu_2 < 5\times 10^{-1}$ Hz improves by over 3 orders of magnitude recent limits on any quadrupolar interactions violating spatial isotropy, and can be interpreted as the most precise test yet of local Lorentz invariance. The experiment also improves the limits on anisotropic dipole interactions.

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We report on an experiment that greatly improves the limit on how much the energy of an atomic nucleus depends upon its orientation in space.[1] Our result can be interpreted as the most precise test of Lorentz invariance to date and as setting strict limits on nonmetric theories of gravity or the size of nonuniversal coupling of gravity to different forms of mass energy.[2-4]

Early tests of nuclear-spin-coupled spatial anisotropy were carried out by Hughes and collaborators[5] and Drever,[6] who searched for a variation in quadrupole splitting among the Zeeman levels of ^{7}Li ($I=\frac{3}{2}$) as the quantization axis rotated with the Earth, and obtained an upper limit of 2×20^{-2} Hz. Last year, a measurement on trapped ^{9}Be$^+$ ions lowered the experimental limits on anisotropy to approximately 10^{-4} Hz.[7]

In our experiment we compare the nuclear-spin-procession frequencies of optically pumped ^{201}Hg ($I=\frac{3}{2}$) and ^{199}Hg ($I=\frac{1}{2}$) isotopes contained in the same cell, and look for a relative shift as the orientation of the precession axis relative to the fixed stars changes with the Earth's rotation. We have achieved a sensitivity of better than 10^{-6} Hz to Zeeman frequency shifts. Our null result, combined with the relatively large quadrupole moment of ^{201}Hg, further improves limits on anisotropic quadrupolar coupling by a factor of 2500, constraining, for example, the fractional size of possible Lorentz-noninvariant electromagnetic couplings to be $\leq 10^{-21}$.

We expand the frequency shift of a nuclear Zeeman level due to small spatially anisotropic coupling as

$$\delta\nu(m,I)=\sum_{k=1}^{2I}\nu_k(I)T_0^k(I)_{mm}P_k(\cos\beta), \qquad (1)$$

where m is the projection of the spin I along the axis of quantization, $T_0^k(I)$ are multipole spin operators[8] normalized to $\mathrm{Tr}[T_0^k,T_0^{k\dagger}]=1$ $[T_0^1(\frac{1}{2})_{mm}=\sqrt{2}m$, $T_0^1(\frac{3}{2})_{mm}=m/\sqrt{5}$, and $T_0^2(\frac{3}{2})_{mm}=(3m^2-I^2)/6]$, and $P_k(\cos\beta)$ are Legendre polynomials with β the angle of quantization relative to some assumed preferred direction in space (such as the direction of motion through the background black-body radiation, the direction toward the galactic center, etc.). The quantization axis of our apparatus rotates with the Earth in the equatorial plane.

The $\nu_k(I)$ are the frequency shifts associated with the kth multipole moment of the polarization of the Hg isotope with nuclear spin I. Our results for the dipole and quadrupole shifts from Eq. (1) are

$$\delta\nu_1\equiv[\nu_1(\tfrac{1}{2})+0.85\nu_1(\tfrac{3}{2})]\cos\theta$$
$$=1.03\pm 0.82\ \mu\text{Hz},$$
$$\delta\nu_2\equiv\nu_2(\tfrac{3}{2})\cos^2\theta=0.17\pm 0.19\ \mu\text{Hz}, \qquad (2)$$

where θ is the latitude of an assumed anisotropy axis. (In principle, our experiment is also sensitive to ν_3, but for technical reasons we have not yet calibrated our apparatus for an octupole shift. The experiment of Ref. 7 sets a bound on ν_3 as well as ν_1 and ν_2.)

Our experiment uses two optically pumped light-absorption oscillators[9] based on the nuclear-spin Larmor frequencies of the two Hg isotopes. Each oscillator operates with the geometry shown in Fig. 1. Circularly polarized resonance light propagates along the z axis. At an angle of 45° is a static magnetic field B_0 (the quantization axis) which lies in the yz plane (the Earth's equatorial plane). Along the x axis is an oscillating magnetic field whose frequency ω is close to the Larmor frequency ω_0.

Through optical pumping, the nuclear spins become polarized; the expectation value of the spin, $\langle \mathbf{I} \rangle$, precesses at frequency ω in a cone about $\mathbf{B}_0$. The projection of $\langle \mathbf{I} \rangle$ on the z axis has a static and an oscillating component. Since the absorption of the light depends on $\langle I_z \rangle$ the light will acquire a modulation at ω. For $\omega = \omega_0$ (and in the absence of additional phase shifts), the modulation will be exactly in phase with B_x. For small deviations of ω from ω_0 there will be a phase shift between B_x and the light modulation of magnitude $\phi \approx T_2 \Delta\omega$, where T_2 is the transverse spin-relaxation time.

We are interested in shifts of the nuclear Zeeman levels due to external dipole and quadrupole couplings. When we average the shift in Eq. (1) over all Zeeman levels, it is evident that the dipole sensitivity is proportional to $\langle T_0^1 \rangle$, the vector spin polarization of the atoms, while the quadrupole sensitivity is proportional to $\langle T_0^2 \rangle$, the spin alignment of the atoms. These sensitivities can be calibrated under operating conditions by the application of known dipole and quadrupole perturbations as described below. An important conceptual difference between our technique to detect external quadrupole interactions and the techniques of previous attempts is that in our case neither the Zeeman transitions nor the quadrupole shifts need to be resolved. Such sublinewidth quadrupole sensitivity arises from the presence of a nonzero spin alignment.

As seen in Fig. 1, the pumping light for both isotopes comes from a stabilized ^{204}Hg electrodeless microwave discharge lamp. The single 253.7-nm line of the ^{204}Hg overlaps the $F=\frac{1}{2}$ to $F=\frac{1}{2}$ transition in ^{199}Hg and the $F=\frac{3}{2}$ to $F=\frac{5}{2}$ transition in ^{201}Hg. The resonance light is circularly polarized to a degree of polarization greater than 95%.

The cell containing the mercury is a 2-cm-diam sphere made of high-purity fused silica (Suprasil) which was filled with mercury vapor (16% ^{199}Hg, 13% ^{201}Hg) at the 0 °C vapor pressure. To achieve long spin-relaxation times, the cell is maintained at 300 °C in a temperature-regulated oven in the center of three Helmholtz coils which provide the static and oscillating magnetic fields. The static field has a magnitude of about 20 mG, corresponding to Larmor frequencies for ^{201}Hg and ^{199}Hg of 5.5 and 15 Hz, respectively.

The cell, oven, and coils are located inside three concentric cylindrical Molypermalloy magnetic shields. A polished aluminum tube which serves as a light pipe collects the light transmitted through the cell. The intensity of the transmitted light is measured with a solar-blind photomultiplier tube. The signal from the photomultiplier tube is amplified, filtered, and phase detected against the oscillating fields. Four digitally controlled switching-type phase-sensitive detectors were built to measure the in-phase and the quadrature components of the signal from each isotope. 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. The vector spin-polarized lifetime (T_2) for the ^{199}Hg is 1120 ± 40 s and for the ^{201}Hg is 80 ± 5 s. The ^{201}Hg spin-alignment lifetime is 40 ± 5 s. With the resonance light optically pumping the

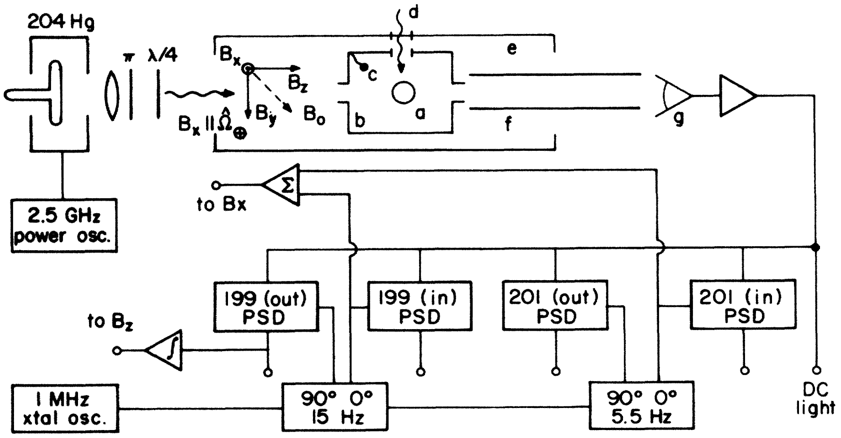

FIG. 1. Schematic of the experiment. a, Cell; b, oven; c, thermocouple; d, calibration light; e, magnetic shield; f, light pipe; and g, R-166 photomultiplier.

ensemble of atoms the ^{199}Hg lifetime is shortened to 80 s and the ^{201}Hg lifetime (vector) is shortened to 50 s. The amplitude of the oscillating field for each isotope was set so that the in-phase components were 0.7 times their respective limiting values. For the ^{201}Hg this corresponds to about 19 μG and for the ^{199}Hg about 6 μG.

The ^{199}Hg quadrature signal is used to stabilize the magnetic field by adjustment of the current in the z-axis Helmholtz coil to hold the ^{199}Hg quadrature signal at zero. The ^{201}Hg quadrature signal is then insensitive to fluctuations in either the magnetic field or the reference oscillator. Thus the ^{201}Hg quadrature signal V_{201}^{out} becomes a measure of any shift between the Zeeman energy levels of the two isotopes that does not scale as the ratio of the nuclear magnetic moments. In particular, V_{201}^{out} is sensitive to a quadrupole energy shift in ^{201}Hg since a quadrupole cannot affect the energy levels of ^{199}Hg. Because the ^{199}Hg servo also forces $\nu_1(\frac{1}{2})$ in Eq. (1) to be zero, V_{201}^{out} is sensitive to an anisotropic dipole energy shift given by $\nu_1(\frac{3}{2})-\sqrt{10}(\gamma_{201}/\gamma_{199})\nu_1(\frac{1}{2})$, where $\gamma_{201}/\gamma_{199}=-0.37$ is the ratio of the nuclear magnetic moments.

The sensitivity of the apparatus to dipole and quadrupole energy shifts is calibrated experimentally. The dipole sensitivity $\partial V_{201}^{\text{out}}/\partial\nu_1$ is measured by our changing the ^{201}Hg drive frequency by a small amount ($\Delta\nu=120$ μHz) and measuring the resulting change in V_{201}^{out}. The quadrupole sensitivity $\partial V_{201}^{\text{out}}/\partial\nu_2$ is determined by the application of a known quadrupole light shift with the use of light from a second lamp containing enriched ^{200}Hg. This isotope emits a single 253.7-nm line far enough from resonance with either pumped isotope to induce readily measurable quadrupole and dipole light shifts without seriously affecting the pump rates. The ratio of these virtual light shifts for ^{201}Hg can be calculated accurately,[8] and thus a measurement of them determines η, the ratio of quadrupole sensitivity to the measured dipole sensitivity,

$$\eta\equiv\frac{\partial V_{201}^{\text{out}}/\partial\nu_2}{\partial V_{201}^{\text{out}}/\partial\nu_1}.$$

A typical measured value of η in our apparatus is 0.7. The measured value for a given set of operating conditions agrees with an alternative calibration obtained under the same conditions by measurement of the magnitude of the precessing ^{201}Hg spin alignment and vector polarization,[10] and also falls within the range expected on the basis of a steady-state density-matrix calculation. A more detailed discussion of all aspects of this experiment will be presented elsewhere.

A computer records the in-phase and quadrature components of the transmitted light modulation for both isotopes, as well as the lamp intensity, magnetic field correction signal, oven drive current, and room temperature. The signals are averaged over a 20-min interval to obtain a datum point with an error bar determined by the scatter during this interval. We have taken six runs, each of 2.5-d continuous duration. The direction of $\mathbf{B}_0$ was changed by 90° for half of the data to discriminate against systematic effects associated with the time of day.

By multiple regression we fit the ^{201}Hg phase $V_{201}^{\text{out}}/V_{201}^{\text{in}}$ to sine and cosine components at $\omega_\oplus$ and $2\omega_\oplus$ where $\omega_\oplus$ is the Earth's rotation frequency in sidereal time. The regression analysis removes fluctuations in lamp intensity and ^{199}Hg phase, and includes terms to remove any static phase offsets due to voltage offsets in the electronics or misalignments among $\mathbf{B}_0$, $\mathbf{B}_x$, and the optical fields. A curved baseline (second order in time) is included to account for very low-frequency drifts.

The results of different analysis methods (fitting the data as six separate measurements, fitting to all data modulo one sidereal day, including correlations with all of the measurement channels) show agreement to within their estimated uncertainties. The data of Fig. 2 are typ-

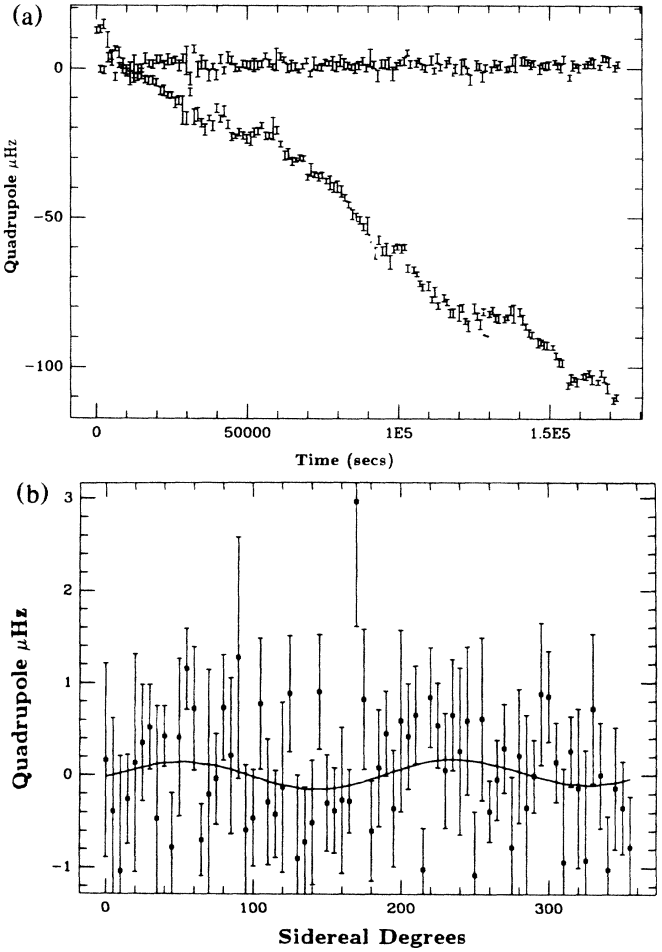

FIG. 2. (a) Raw data for V_{201}^{out} before (sloping curve) and after regression analysis for a typical 2.5-d run. (b) Entire 15 days of data averaged into 5° bins modulo one sidereal day. The solid curve is the fit to Fourier components at $\omega_\oplus$ and $2\omega_\oplus$.

ical. Before the multiple regression described above, the raw data exhibit substantial $1/f$ noise. After multiple regression we can subtract from the data correlations with the simultaneously monitored channels described in the previous paragraph. The plot of this reduced data in Fig. 2(a) demonstrates that almost all of the drift and $1/f$ noise are accounted for by these correlations. The most important source is the correlation with light intensity. Figure 2(b) shows the result of plotting all such reduced data modulo one sidereal day. The χ^2 for the fit shown in Fig. 2(b) is 1.1 which indicates that any contribution from $1/f$ noise to our reduced data is small compared to the contribution from the error bars on the 20-min data points. We estimate limits on possible systematic errors below.

Static quadrupole splittings for the ^{201}Hg nucleus occur because of a light shift and a bottle shift. Variations in the lamp intensity or dwell time of atoms on the cell walls could imitate anisotropy effects if they exhibit sizable frequency components near harmonics of $\omega_\oplus$. The measured variations in light shift were of order 1 μHz at $2\omega_\oplus$ and 4 μHz at $1\omega_\oplus$ before they were removed in the data reduction. The bottle shift was measured to be approximately 130 μHz and to vary with temperature by 2 μHz/K. Oven temperature was sufficiently stable to limit the total variation in bottle shift to less than 0.2 μHz. Another source of systematic error would be a small variation in the quadrupole sensitivity at some harmonic of $\omega_\oplus$. We were able to estimate the maximum size of any such variation from the behavior of the data when the quantization axis was changed by 90°.

The experimental errors quoted in Eq. (2) include both statistical and systematic uncertainties. At the 95% confidence level, we conclude that $|\delta\nu_1| < 2.4$ μHz and $|\delta\nu_2| < 0.48$ μHz.

One can interpret ν_2 as setting a limit of $\delta m_2 < 2\times10^{-21}$ eV, where δm_2 is any dependence of the nuclear rest mass on orientation relative to the mean rest frame of the universe.[2] A similar limit is set for most other plausible preferred frames. A nonzero δm either implies an as yet unknown cosmic background field or is a manifest violation of local Lorentz invariance (LLI) as postulated in Einstein's statement of the equivalence principle. Our limit on δm_2 can be combined with the known quadrupole moment of ^{201}Hg to place stringent limits on non-LLI field couplings, including nonuniversal couplings between gravity and other forms of mass energy. For instance, in the $TH\varepsilon\mu$ formalism,[2,3,11] photons and massive particles couple differently to gravitational fields and hence have different limiting speeds c and c_0, respectively. With the assumption that the mean rest frame of the universe is the "preferred frame," our data imply $|c-c_0| < 3\times10^{-22}c$.

The interactions $\nu_1(I)$ measure the strength of a dipolar coupling to spin that violates LLI or that arises from a background field with a preferred direction relative to the Earth. For example, a coupling of an atom's spin to an extra terrestrial field could take the form $U_1 = U_n\mathbf{I}\cdot\hat{\mathbf{r}}$,[12,13] or $U_2 = K_n\mathbf{I}\cdot\mathbf{V}$,[13,14] where U_n and K_n are the coupling strengths, $\hat{\mathbf{r}}$ is the unit vector to the source of the field, and $\mathbf{V}$ is the velocity of the atom relative to a preferred reference frame. Our results can be used to set new limits on U_n and K_n if these coupling strengths do not scale as the ratio of the nuclear magnetic moments for ^{199}Hg and ^{201}Hg.

In conclusion, the comparison of the precession frequencies of two atomic species in the same cell by optical pumping techniques has provided the most stringent limits to date on spatially anisotropic dipole and quadrupole interactions. Improvements in our method would include a rotating apparatus to reduce diurnal systematic effects, and a more stable light source, which was the dominant noise source in the present work.

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