## Limits for Spatial Anisotropy by Use of Nuclear-Spin–Polarized <sup>9</sup>Be<sup>+</sup> Ions

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The frequency of a nuclear spin-flip  $(|\Delta m_l| = 1)$  transition in <sup>9</sup>Be<sup>+</sup> has been compared to the frequency of a hydrogen maser transition  $(|\Delta F| = 1, \Delta m_F = 0)$  to see if the relative frequencies depend on the orientation of the <sup>9</sup>Be<sup>+</sup> ions in space. The present null result represents a decrease in the limits set by Hughes and Drever on a spatial anisotropy by a factor of about 300.

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In metric theories of gravity the influence of external gravitational fields on atomic structure is governed by the Einstein equivalence principle (EEP).<sup>1</sup> All metric theories (including general relativity) are based on the EEP which states that (i) all bodies fall in a given gravitational field with the same acceleration (weak equivalence principle), (ii) the outcome of any local nongravitational experiment is independent of the velocity and orientation of the freely falling apparatus [local Lorentz invariance, (LLI)], and (iii) the outcome of any local nongravitational experiment is independent of where and when in the universe it is performed. It follows from LLI that two different atomic clocks located at the same point in space-time will have relative rates that are independent of the velocity and orientation of the freely falling lab.

As a test of LLI the frequency of the  $2^2S_{1/2}(M_I, M_J) = (-\frac{3}{2}, +\frac{1}{2}) \rightarrow (-\frac{1}{2}, +\frac{1}{2})$  (see Fig. 1) ground-state hyperfine transition in <sup>9</sup>Be<sup>+</sup> (hereafter called the "clock" transition) has been compared to the frequency of a passive hydrogen maser<sup>2</sup> to see if a correlation between the relative rates of the two clocks and the orientation of the <sup>9</sup>Be<sup>+</sup> nuclear spin in space can be found. Such clock-comparison experiments are interpreted as the most precise tests of LLI.<sup>1,3</sup>

Various proposals for violations of LLI have been made. One of these is a coupling of a particle's spin to the gravitational field,  ${}^{4} U_{g} = U_{n} \mathbf{I} \cdot \hat{\mathbf{r}} + U_{e} \mathbf{S} \cdot \hat{\mathbf{r}}$ , where  $U_{n}$ and  $U_{e}$  are the strengths of the coupling for a nucleon



FIG. 1. Hyperfine structure (not drawn to scale) of the  ${}^{9}\text{Be}^{+} 2s \, {}^{2}S_{1/2}$  ground state as a function of magnetic field.  $\nu$  is a first-order magnetic field-independent transition at 0.819 T.

and electron spin, respectively, and  $\hat{\mathbf{r}}$  is the unit vector from the atom to the source of the field. This interaction violates parity (P) and time-reversal (T) invariance. Similarly, and LLI-violating and P-nonconserving coupling of a particle's spin to its motion with velocity V relative to some preferred frame of reference of the form  $U_v = K_n \mathbf{I} \cdot \mathbf{V} + K_e \mathbf{S} \cdot \mathbf{V}$  has been proposed.<sup>5</sup> These couplings produce a shift of the <sup>9</sup>Be<sup>+</sup> clock transition proportional to  $\cos\beta$  where  $\beta$  is the angle between the quantization axis for the ions and the direction of  $\hat{\mathbf{r}}$  or  $\mathbf{V}$ . The hydrogen maser transition  $(F=1, M_F=0) \rightarrow (F=0, M_F=0)$  is not sensitive to these perturbations in first order because, in the low operating magnetic field of the maser (  $\sim 100 \ \mu G$ ) the proton and electron spins couple together so that  $\langle \mathbf{I} \cdot \hat{\mathbf{r}} \rangle = \langle \mathbf{S} \cdot \hat{\mathbf{r}} \rangle = 0$  for any direction  $\hat{\mathbf{r}}$ , where the averaging is done in either of the two maser states.

Another LLI-violating interaction which shifts the relative rates of the 9Be+ and maser clocks was originally proposed by Cocconi and Salpeter as a model for inertial mass.<sup>6</sup> The model is motivated by Mach's principle which states that a body's inertial mass is determined by the total distribution of matter in the universe. Since matter in the nearby universe is distributed anisotropically, inertial mass could show a corresponding anisotropy. Indeed, in this model the mass of an orbiting nucleon (orbital angular momentum  $l \ge 1$ ) will depend upon the orientation of its orbit relative to the direction of the matter anisotropy. This interaction leads to a shift of the <sup>9</sup>Be<sup>+</sup> clock transition proportional to  $P_2(\cos\beta) = (3\cos^2\beta - 1)/2$ , where  $\beta$  is now the angle between the quantization axis for the <sup>9</sup>Be<sup>+</sup> ions and the direction of matter anisotropy in the nearby universe (e.g., the direction toward the galactic center or Virgo supercluster center). Searches for frequency shifts of NMR transitions in <sup>7</sup>Li which were correlated with the direction toward the galactic center were made by Hughes and co-workers<sup>7</sup> and Drever<sup>8</sup> and their null results ruled out the Cocconi-Salpeter model. It should be stated that in their model of inertial mass, Cocconi and Salpeter computed the change in kinetic energy of an orbiting nucleon induced by a spatial anistropy. Others<sup>9,10</sup> have pointed out that when anisotropic effects on both kinetic and potential

Work of the U. S. Government Not subject to U. S. copyright energies are considered, the null results are to be expected provided that the anisotropy couples in the same way to both forms of energy. These experiments can thus be regarded as a test of the universal coupling of gravity to all forms of mass-energy.<sup>1,3</sup>

Models of electrodynamics in a gravitational field which include the possibility of nonuniversal couplings are the Dicke-Peebles-Ni formalism<sup>10,11</sup> and the  $TH\epsilon\mu$  formalism.<sup>1,3,12</sup> In the  $TH\epsilon\mu$  formalism the parameters  $T_0$  and  $H_0$  describe the coupling of gravity to material particles while  $\epsilon_0$  and  $\mu_0$  describe gravity's coupling to electromagnetic fields. In this formalism the limiting speed for material particles is  $c_0$  while the speed of light is  $c_{\text{light}}$ , where  $c_0/c_{\text{light}}(T_0 \epsilon_0 \mu_0/H_0)^{1/2}$ .

If  $c_0 \neq c_{\text{light}}$  an electromagnetically bound system of charged particles experiences a coupling between its internal structure and center-of-mass motion. This produces a contribution  $\delta E = \sum \delta m_i^{jj} V^i V^j$  to the electromagnetic part of the nuclear binding energy.  $\delta m_i^{jj}$  is the anomalous inertial mass tensor<sup>1,3</sup> which depends upon the electromagnetic structure of the nucleus and **V** is the velocity of the nucleus through some preferred frame. This could be the rest frame of the sun or the mean rest frame of the universe (defined as the frame in which the cosmic 3-K background radiation is isotropic).<sup>13</sup>

In the  $TH\epsilon\mu$  formalism<sup>14</sup>

$$\delta E \simeq -\frac{1}{4} \left( 1 - \frac{T_0 \epsilon_0 \mu_0}{H_0} \right) \left\langle \frac{e^2}{c^2} \sum_{i \neq j} \frac{1}{r_{ij}^3} \left\{ (\mathbf{r}_{ij} \cdot \mathbf{V})^2 - \frac{r_{ij}^2 V^2}{3} \right\} \right\rangle_{I, M_I}, \tag{1}$$

where  $\mathbf{r}_{ij}$  is the coordinate vector from the *i*th to *j*th nuclear proton, *e* is the proton charge, *c* is the speed of light, and the summation is over the *Z* protons. Use of the Wigner-Eckart theorem gives

$$\delta E \simeq -\frac{1}{4} \left( 1 - \frac{T_0 \epsilon_0 \mu_0}{H_0} \right) \frac{3M_I^2 - I(I+1)}{I(2I-1)} \frac{e^2}{c^2} \left\langle \sum_{i \neq j} \frac{(\mathbf{r}_{ij} \times \mathbf{r}_{ij})_0^{(2)}}{r_{ij}^3} \right\rangle_{I=M_I} (\mathbf{V} \times \mathbf{V})_0^{(2)}, \tag{2}$$

where  $(\ldots)_{M}^{(J)}$  denotes the J,M spherical component of the tensor in parenthesis. The resulting variation in the <sup>9</sup>Be<sup>+</sup> clock transition frequency is

$$\delta\nu \cong \left(1 - \frac{T_0 \epsilon_0 \mu_0}{H_0}\right) \frac{(Z-1)e^2 Q}{6hR^3} \frac{V^2}{c^2} P_2(\cos\beta), \quad (3)$$

where Q and R are the electric quadrupole moment and charge radius of the <sup>9</sup>Be nucleus. We have used the following estimate for the nuclear matrix element<sup>15</sup>:

$$e^{2}\left\langle\sum_{i\neq j}\frac{(\mathbf{r}_{ij}\times\mathbf{r}_{ij})_{0}^{(2)}}{r_{ij}^{3}}\right\rangle_{I=M_{I}}\approx\frac{(Z-1)e^{2}Q}{\sqrt{6}R^{3}}.$$
 (4)

These quadrupolar perturbations do not shift the hydrogen-maser transition frequency. Finally because the  ${}^{9}\text{Be}^{+}$  nuclear spin is  $I = \frac{3}{2}$ , we are sensitive to shifts of the clock transition up to order  $P_{3}(\cos\beta)$  but no higher.

A discussion of the Be<sup>+</sup> clock is given by Bollinger et al.<sup>16</sup> A few hundred to 2000 <sup>9</sup>Be<sup>+</sup> ions are stored in a Penning trap.<sup>17</sup> The ions are cooled and optically pumped (95%) into the  $2^2 S_{1/2}(M_I, M_J) = (-\frac{3}{2}, -\frac{1}{2})$ ground state by radiation from a frequency-doubled dye laser ( $\lambda = 313$  nm, power  $\approx 20 \mu W$ ) which is the  $2^2 S_{1/2}(-\frac{3}{2},-\frac{1}{2})$ slightly below tuned  $\rightarrow 2^2 P_{3/2}(-\frac{3}{2}, -\frac{3}{2})$  transition frequency.<sup>16,17</sup> At this frequency the ions absorb radiation more strongly when their motion is toward the laser than when they move away from it. When averaged over all angles of reemission, the ion's momentum is reduced by  $h/\lambda$ per scattered photon where  $\lambda$  is the laser wavelength and h is Planck's constant. This results in a cooling of the Be<sup>+</sup> cloud to about 1 K. The observed intensity of the scattered fluorescent light from the  $2^{2}S_{1/2}(-\frac{3}{2}, -\frac{1}{2}) \rightarrow 2^{2}P_{3/2}(-\frac{3}{2}, -\frac{3}{2})$  transition is proportional to the population of the  $(-\frac{3}{2}, -\frac{1}{2})$ ground state.

At a magnetic field of about 0.8194 T [ground-state  $\left(-\frac{3}{2},-\frac{1}{2}\right) \rightarrow \left(-\frac{3}{2},\frac{1}{2}\right)$  electron spin-flip frequency of 23 914.01 MHz], the clock transition frequency,  $\nu$ , depends only quadratically on the magnetic field deviation,  $\delta B$ , as  $\delta \nu / \nu = -0.017 (\delta B / B)^2$ . The clock transition is detected by optical-microwave-rf triple resonance. Microwave radiation tuned to the electron spin-flip resonance transfers half of the ion population from the optically pumped  $\left(-\frac{3}{2},-\frac{1}{2}\right)$  state to the  $\left(-\frac{3}{2},+\frac{1}{2}\right)$  state. Some of the  $\left(-\frac{3}{2},+\frac{1}{2}\right)$  state population is transferred to the  $\left(-\frac{1}{2}, +\frac{1}{2}\right)$  state by application of rf near the 303-MHz clock transition frequency. Because of the microwave mixing this results in an additional decrease in the  $\left(-\frac{3}{2},-\frac{1}{2}\right)$  state population and therefore a decrease in the observed fluorescence.

The clock transition is driven by a synthesizer using Ramsey's separated oscillatory fields technique<sup>18</sup> with 0.5-s coherent Rabi pulses separated by a 19-s free-precession period. A computer adjusts the frequency of the synthesizer so that it remains centered on the clock transition. The time base for this synthesizer is provided by a passive hydrogen maser. Any shift of the <sup>9</sup>Be<sup>+</sup> transition frequency relative to the maser transition will appear as a variation of the synthesizer's frequency. In a 2-h measurement period the signal-

to-noise ratio was sufficient to locate the center of the 25-mHz-wide clock resonance to better than 0.5%. The results of 29 such measurements taken between 7 May and 15 June, 1984 are shown plotted against sidereal time in Fig. 2.<sup>19</sup>

Because the Earth rotates, the angle  $\beta$  changes throughout the day. That is,  $\beta = \beta(\tau)$  where  $\tau$  is the sidereal time. For example, consider the direction of motion through the mean rest frame of the universe. Our laboratory is at 40° north latitude and our magnetic field is horizontal and directed 232° from geographic north (i.e., almost southeast). Thus, at 3<sup>h</sup> sidereal time  $\beta = 158^\circ$  while at 15<sup>h</sup>,  $\beta = 34^\circ$ . Since the magnetic field direction is not changed with respect to the vertical we are insensitive to any shifts of the <sup>9</sup>Be<sup>+</sup> transition caused by the Earth's gravitational field.

We have searched for a variation in the clock transition frequency of the form

$$\nu = \nu_0 + A_k P_k(\cos\beta(\tau)) \tag{5}$$

(k = 1, 2, or 3) where  $A_k$  measures the magnitude of any LLI violation. The results of a least-squares fit of the function of Eq. (5) to the data of Fig. 2 is shown in Table I. Also shown in that table is the direction in space which determines the angle  $\beta(\tau)$ . The errors are the quadrature sum of the statistical error (one sigma) and a 27- $\mu$ Hz uncertainty in the variation of the second-order Doppler shift of the clock transition over the 29 runs. This shift arises because, to avoid light shifts of the clock transition, during the 20-s Ramsey interrogation period the ions are not laser cooled and consequently heat up from about 1 K to about 30 K. Also, magnetic field measurements at the beginning and the end of each run indicate a field deviation of  $\delta B/B \cong \pm 1.5 \times 10^{-6}$ . This leads to a peak-to-peak frequency fluctuation of the clock transition of about 12  $\mu$ Hz over the 29 runs. These were the two largest systematic errors in this measurement, although several other effects were considered.<sup>16</sup> Fluctuations



FIG. 2. Variation of the  ${}^{9}\text{Be}^{+}$  clock transition frequency referenced to a passive hydrogen maser plotted against sidereal time. Tick marks on the vertical scale are 100  $\mu$ Hz apart.

in any of the systematic errors could potentially mimic a  $P_k(\cos\beta(\tau))$  variation but the amplitude of such variations would be below our quoted uncertainty.

In addition to the directions listed in Table I we have also fitted the data of Fig. 2 for an arbitrary direction (excluding those directions within 10° of the Earth's rotational axis where our sensitivity is reduced). We find that all resulting values of  $A_k$  are consistent with zero at the 100- $\mu$ Hz level.

In the direction of the galactic center, our present limit on  $|A_2|$  (in <sup>9</sup>Be<sup>+</sup>) of 50 + 42 = 92  $\mu$ Hz is about a factor of 200 improvement over the limit of Hughes and co-workers<sup>7</sup> and Drever<sup>8</sup> on  $|A_2|$  (in <sup>7</sup>Li) for the same direction. For the direction of the Earth's motion through the mean rest frame of the universe, we take 81  $\mu$ Hz as the limit on  $|A_2|$ . Using Eq. (3) and  $V^2/c^2 \simeq 10^{-6}$  (Ref. 13) we place a limit on the *TH* $\epsilon\mu$  preferred-frame parameter<sup>1,3</sup> of  $|1 - T_0\epsilon_0\mu_0 \times H_0^{-1}| \le 10^{-18}$ . This is about 300 times smaller than the Hughes-Drever limit on this parameter if we use their limit on  $A_2$  in the direction of the galactic center and the same method for estimating the nuclear matrix element [Eq (4)]. The limits on  $A_1$  are comparable with limits set by other workers<sup>20</sup> while the limits on  $A_3$  are new. From the limit on  $A_1$  we find that  $U_n, K_n |\mathbf{V}| \leq 100 \ \mu \text{Hz}$  and because  $\langle S_z \rangle$  differs for the two levels of the clock transition by about  $2 \times 10^{-4}$ , we find  $U_e, K_e |\mathbf{V}| \leq 0.50$  Hz. Although the limits established here can be regarded as a test of LLI, we wish to emphasize that they set an upper limit on any dependence of the relative clock rates on their orientation with respect to the sun or the fixed stars, whether from a breakdown of LLI or some other cause, such as a new interaction.

It should be possible to improve the sensitivity of the present measurement by more than an order of magnitude.<sup>16</sup> A search for a  $P_1(\cos\beta)$  interaction due to the Earth's gravitational field would be facilitated by orienting the experimental magnetic field along the

TABLE I. Variation of the  ${}^{9}\text{Be}^{+}$  clock transition frequency,  $\nu$ , measured relative to a passive hydrogen maser.  $A_k$  is a measure of any LLI violation,  $\nu = \nu_0 + A_k P_k (\cos\beta(\tau))$ , where  $\beta$  is the angle between the  ${}^{9}\text{Be}^{+}$  quantization axis and the direction in space given in the table.  $\tau$  is sidereal time.

Direction	$A_k (\mu \text{Hz})$		
	$A_1$	$A_2$	$A_3$
Motion through mean rest frame of the universe	$35 \pm 40$	35 ± 46	13 ± 54
Motion through solar rest frame	36 ± 42	$30 \pm 51$	9 ± 63
Galactic center	$-49 \pm 38$	$-50 \pm 42$	$-15 \pm 46$
Virgio supercluster	$20 \pm 40$	$0 \pm 49$	$9 \pm 57$
Sun	$40 \pm 39$	$-58 \pm 43$	$16 \pm 47$

vertical. It should also be possible to make such measurements similar to the present one using nuclear magnetic resonance techniques on neutral atoms.<sup>20,21</sup> In fact, relevant data may already exist from NMR gyroscope experiments.<sup>22</sup>

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