## Search for Anomalous Spin-Dependent Forces Using Stored-Ion Spectroscopy

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Resonances in atomic ions can be used to search for new, weak, spin-dependent interactions. Upper limits on anomalous dipole-monopole and dipole-dipole couplings for the neutron and electron are determined by examining hyperfine resonances in stored <sup>9</sup>Be<sup>+</sup> ions. These experiments also place strict limits on anomalous weights of spinning gyroscopes.

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The existence of weakly interacting bosons has been suggested previously [1-10]. Laboratory experiments might detect scalar or pseudoscalar couplings of such particles to photons [11-15] or matter [6,7,16-30]. In the latter case, new spin-dependent forces would occur. Here, we report the use of stored atomic ion spectroscopy to search for anomalous potentials having a dipole-monopole or dipole-dipole character. The first is expected to include terms like [7,9,16,31]

$$V_{AB}^{A} = \hbar^{2} D \mathbf{S}_{A} \cdot \hat{\mathbf{r}} (1/\lambda_{\phi} \mathbf{r} + 1/r^{2}) \exp(-r/\lambda_{\phi}), \qquad (1)$$

where the spin  $S_A$  (in units of  $\hbar$ ) of particle A couples to particle B, r is the distance between particles,  $\lambda_{\phi}$  is the range of the force, and D is a coupling constant with units of (mass)<sup>-1</sup> [see Fig. 1(a)]. In 1968, observation of an interaction like Eq. (1) was reported [22] where particles A were proton spins and particles B comprised the Earth. Subsequent measurements [23,24] contradicted this measurement and found null results (see Table I).

A dipole-dipole interaction [Fig. 1(b)] would be expected to include terms like [7,16,31]

$$V_{AB}^{I} = (\hbar^{3}/c)T \exp(-r/\lambda_{\phi})\left[(1/\lambda_{\phi}r^{2} + 1/r^{3})\mathbf{S}_{A} \cdot \mathbf{S}_{B} - (1/\lambda_{\phi}^{2}r + 3/\lambda_{\phi}r^{2} + 3/r^{3})(\mathbf{S}_{A} \cdot \hat{\mathbf{f}})(\mathbf{S}_{B} \cdot \hat{\mathbf{f}})\right],$$
(2)



FIG. 1. Experimental configurations sensitive to (a)  $V_{AB}^{B}$  and (b)  $V_{AB}^{T}$  [refer to Eqs. (1) and (2) of the text]. In both parts, we assume the size of sample A is small compared to d, R, and l. In (a), B is assumed to be spherical with density of B particles  $\rho_{B}$ . In (b), B is assumed to be a cylinder of radius R and height l and spin density  $\rho_{B}$ . Experimentally,  $V_{AB}^{D}$  and  $V_{AB}^{T}$  are sensed as an anomalous change in energy when  $S_{A} \cdot \hat{z}$  is changed. In the experiments reported here,  $S_{A} \cdot \hat{z}$  for the electron and neutron are changed by driving a hyperfine transition in the ground state of atomic <sup>9</sup>Be<sup>+</sup> ions.

where the spin of particle A interacts with that of particle B. T has units of (mass)<sup>-2</sup> and characterizes the strength of the interaction [see Fig. 1(b)]. This type of interaction is sought in acceleration [7,16,19,20], resonance [16,22-27,29], and induced magnetism [17,18] experiments.

Such forces mediated by axions [4] have received the most attention because the axion emerges in schemes attempting to resolve the strong CP problem [4,10]. The mass and coupling of axions and related particles to matter can be severely constrained by arguments based on observed energy-loss rates of stellar objects [32]. As discussed below, these constraints on axions appear to be much stronger than those derived from current laboratory experiments. Nevertheless, the laboratory experiments are still useful because they can search for interactions outside the scope of the axion-type models.

TABLE I. Values of the coupling strength D [Eq. (1)] evaluated from laboratory experiments. Here, we assume  $\lambda_{\phi} \gg R = R_{\text{Earth.}}$ 

Experiment	Measured quantity	Result (kg <sup>-1</sup> )
Ref. [22]	D(proton)	≈0.02
Ref. [23]	D(proton)	$< 2 \times 10^{-4}$
Ref. [24]	D(deuteron)	$< 6.7 \times 10^{-8}$
Ref. [28]	D(electron)	$< 3.7 \times 10^{5}$
This work	$D(^{9}\text{Be})$	$< 9.0 \times 10^{-9}$
This work	D(electron)	$< 4.5 \times 10^{-5}$
This work	D(neutron)	$< 2.7 \times 10^{-8}$
Ref. [30]	$D(^{201}\text{Hg}) - D(^{199}\text{Hg})$	$\lesssim 3 \times 10^{-10}$

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For the assumed configurations shown in Fig. 1, either  $V_{AB}^D$  or  $V_{AB}^T$  could be observed as an anomalous dependence of energy on  $S_A \cdot \hat{z}$ . In the experiments reported here, we looked for such a dependence in the energy  $2\pi\hbar v_0$  of the atomic  ${}^9\text{Be}^+ {}^2S_{1/2}$  ground-state  $(F=1, m_F)$  $=0) \rightarrow (F=1, m_F=-1)$  hyperfine transition [33] in a magnetic field  $B_0 \simeq 0.8194$  T (transition frequency  $v_0$  $\simeq$  303 MHz). Here,  $m_F$  is the sum of the electron spin J  $(=\frac{1}{2})$  and nuclear spin I  $(=\frac{3}{2})$  projections along the magnetic-field axis, taken to be along  $\hat{z}$ . Measurements of  $v_0$  were made on about 5000  ${}^9\text{Be}^+$  ions stored in a Penning ion trap [34]. The <sup>9</sup>Be<sup>+</sup> ions are trapped and laser cooled (to reduce Doppler shifts), then optically pumped into the  $(F=1, m_F=0)$  level using a combination of laser and radio-frequency (rf) radiation. Additional rf radiation is then used to drive the ion population from the  $(F=1, m_F=0)$  to the  $(F=1, m_F=-1)$  state. Changes in the population of the two states are sensed by looking for changes in laser light scattered by the ions [34]. The frequency  $v_0$  is the frequency of radiation which drives the above transition with maximum probability. The reference for  $v_0$  is an ensemble of hydrogen maser and cesium atomic clocks. Particles A are assumed to be either the <sup>9</sup>Be nucleus or the unpaired outer electron in <sup>9</sup>Be<sup>+</sup>. For the above hyperfine transition, the changes in  $\mathbf{S}_{\mathcal{A}} \cdot \hat{\mathbf{z}}$  are given by  $|\Delta(\mathbf{S}_{\mathcal{A}} \cdot \hat{\mathbf{z}})| \equiv |\Delta S_{\mathcal{A}z}| \simeq 1$  or  $|\Delta S_{\mathcal{A}z}|$ 

 $\approx 2 \times 10^{-4}$  for the <sup>9</sup>Be nuclear spin and electron spin, respectively. (We assume that  $V_{AB}$  affects either the electron or <sup>9</sup>Be nucleus but not both at the same time.)

In the search for  $V_{AB}^{D}$ , particles B were taken to be the nucleons in the Earth. We looked for a change in  $v_0$  between the cases where  $\mathbf{B}_0$  was parallel or antiparallel to the vertical direction in the laboratory. (Since the hyperfine transition is a  $|\Delta m_F| = 1$  transition, we had to subtract the effects of the Earth's rotation.) In the search for  $V_{AB}^T$ , particles B were taken to be the electron spins in the iron pole faces of an electromagnet. We compared  $v_0$ when  $B_0$  was created by this electromagnet with  $v_0$  when  $B_0$  was created by a superconducting solenoid ( $S_B$  spins absent). The value of  $B_0$  was chosen such that  $\partial v_0 / \partial B_0$ =0.  $B_0$  was set by measuring a magnetic-field-dependent transition in <sup>9</sup>Be<sup>+</sup> [34]. (The inaccuracy in our measurement of  $B_0$  due to  $V_{AB}$  is negligible on this fielddependent transition.) For  $\partial v_0 / \partial B_0 = 0$ , we could rule out any positive results which might arise from small changes in  $B_0$  between the different configurations. The largest systematic error in the experiment was due to a background-gas pressure shift of  $v_0$  [34].

For the geometries of Figs. 1(a) and 1(b) we integrate over the volumes containing the *B* particles to find the anomalous energy  $E_A$  of a single *A* particle. Assuming  $S_A = S_{Az}\hat{z}$  and  $S_B = S_{Bz}\hat{z}$ , we find [Fig. 1(a)]

$$E_{A}^{D} = C^{D}L_{D} = C^{D}\lambda_{\phi}(R+d)^{-2} \{ [R(R+d) + \lambda_{\phi}(2R+d) + \lambda_{\phi}^{2}] \exp[-(2R+d)/\lambda_{\phi}] + [R(R+d) - \lambda_{\phi}d - \lambda_{\phi}^{2}] \exp[-d/\lambda_{\phi}) \}$$
  
= 2C<sup>D</sup>R<sup>3</sup>/3(R+d)<sup>2</sup> (for  $\lambda_{\phi} \gg R, d$ ) (3)

and [Fig. 1(b)]

$$E_{A}^{T} = C^{T} \xi_{T} = C^{T} \{ (d/r_{1}) \exp(-r_{1}/\lambda_{\phi}) - [(d+l)/r_{2}] \exp(-r_{2}/\lambda_{\phi}) - \exp(-d/\lambda_{\phi}) + \exp[-(d+l)/\lambda_{\phi}] \}$$

$$= C^{T} \{ d/r_{1} - (d+l)/r_{2} \} \quad \text{(for } \lambda_{\phi} \gg R, d, l) , \qquad (4)$$

where  $C^D = 2\pi\hbar^2 D\rho_B S_{Az}$ ,  $C^T = 2\pi(\hbar^3/c)T\rho_B S_{Az}S_{Bz}$ ,  $r_1 = (R^2 + d^2)^{1/2}$ ,  $r_2 = [R^2 + (d+l)^2]^{1/2}$ , and  $\rho_B$  is the number density of particles *B* (assumed here to be nucleons for  $V_{AB}^D$  and electron spins for  $V_{AB}^T$ ).

In our experiment, we found the shift  $\Delta v_0$  of  $v_0$  due to  $V_{AB}^D$  (where the magnetic field is upward) to be  $\Delta v_0$  $= -6.4 \pm 2.9 \pm 6.4 \ \mu$ Hz for  $d \simeq 1.5$  m [35]. The first error is the random uncertainty, the second, an estimate of the pressure shift variation between field-up and -down measurements. The estimate of the pressure shift variation was made by measuring the fluctuations in  $v_0$  over a period of many months with the magnetic field in one direction. Adding these errors in quadrature, we find  $|\Delta v_0| < 13.4 \ \mu \text{Hz} \ (< 5.5 \times 10^{-20} \text{ eV})$ . Therefore we find for the coupling constant defined in Eq. (1),  $D(^{9}Be)$ < 3.8/ $L_D$  kg<sup>-1</sup> or  $D(e^-) < 1.9 \times 10^4 / L_D$  kg<sup>-1</sup>, where  $L_D$ [Eq. (3)] is in centimeters. If we assume  $\lambda_{\phi} \gg R, d$  and  $R = R_{\text{Earth}} \approx 6.36 \times 10^8$  cm  $[\rho_B = \rho(\text{Earth}) \approx 3.33 \times 10^{24}$ nucleons/cm<sup>3</sup>] we can make the estimates of D shown in Table I. To obtain D(neutron) we argue as follows: In the simplest version of the shell model, the spin of the <sup>9</sup>Be nucleus is due to the odd neutron, which is in the  $1p_{3/2}$ shell. In this model, the expectation value of a component of the neutron spin is  $\frac{1}{3}$  of the expectation value of the corresponding component of the total nuclear spin. From this we obtain  $D(neutron) < 2.7 \times 10^{-8} \text{ kg}^{-1} \approx 4.8 \times 10^{-35} \text{ GeV}^{-1}$ . This constrains one model proposed in Ref. [9] where D might be as large as  $10^{-34} \text{ GeV}^{-1}$ .

We also found  $\Delta v_0 = -13 \pm 30 \pm 170 \ \mu \text{Hz}$  between  $v_0$ measured in an electromagnet [36] and a superconducting magnet system. The first error is the uncertainty in the reference oscillator for the time between measurements ( $\simeq$ 5 yr), the second, relatively large error is due to the uncertainty in an estimate of the pressure shift [34] in different apparatuses. Adding the errors in quadrature, we obtain  $|\Delta v_0| < 186 \ \mu \text{Hz}$  ( < 7.7×10<sup>-19</sup> eV). For the electromagnet, d = 2.7 cm, l = 33 cm, R = 15 cm ( $V_{AB}^T$  is enhanced by 2 because of the two pole faces of the magnet). We neglect the magnet yoke which will increase the effects of  $V_{AB}^{T}$  slightly. From  $\Delta v_0$  and Eq. (4), we can put an upper limit on T [Eq. (2)] for assumed values of  $\lambda_{\phi}$ . Here, for brevity, we will assume that  $\lambda_{\phi} \gg d, l, R$  in which case  $V_{AB}^{T}$  has the same form as normal magnetic coupling. In Table II, we list the ratio  $\alpha$  of  $E_A^T$  [Eq. (4)] to the normal magnetic interaction for various experi-

Experiment	$\alpha(A,B)$	Method
Ref. [25]	$\alpha(p,p) < 5 \times 10^{-5}$	Molecular spectra
Ref. [26]	$\alpha(201^{2} \text{Hg}, e^{-}) < 10^{-10} - 10^{-11}$	NMR
Ref. [26]	$\alpha(^{199}\text{Hg},e^{-}) < 10^{-10} - 10^{-11}$	NMR
Ref. [27]	$\alpha(p,e^{-}) - \alpha(d,e^{-}) < 2 \times 10^{-10}$	NMR
Ref. [20]	$\alpha(e^{-},e^{-}) \lesssim 3 \times 10^{-10}$	Acceleration
Ref. [19]	$\alpha(e^{-}, e^{-}) < 0.9 \times 10^{-11}$	Acceleration
Ref. [17]	$\alpha(e^{-}, e^{-}) < 0.005 \times 10^{-11}$	Induced magnetism
Ref. [18]	$\alpha(e^{-}, e^{-}) < 0.00085 \times 10^{-11}$	Induced magnetism
This work	$\alpha({}^{9}\text{Be}, e^{-}) < 3.8 \times 10^{-11}$	NMR
This work	$\alpha(n,e^{-}) < 2.3 \times 10^{-11}$	NMR
This work	$\alpha(e^{-},e^{-}) < 4.1 \times 10^{-11}$	NMR

TABLE II. Experimental determinations of  $\alpha(A,B)$ , the ratio of  $E_A^T$  [Eq. (4)] to normal magnetic coupling for particles A and B as indicated in Fig. 1. Here,  $e^-$  denotes electron, p denotes proton, n denotes neutron, and d denotes deuteron.

ments. From Ref. [32],  $\lambda_{\phi} \approx 10$  cm is within a "window" on the axion which has not been ruled out by stellar energy-loss rate arguments. If we take  $T = g_A g_B / 16\pi$  $\times M_A M_B$ , where  $g_A$  and  $g_B$  are coupling strengths and  $M_A$  and  $M_B$  are the masses of the elementary particles responsible for  $V_{AB}^T$ , then from Refs. [7] and [32] we estimate theoretically for axions  $\alpha(A,B) \approx 10^{-26}$  for  $\lambda_{\phi}$  $\approx 10$  cm, considerably smaller than the sensitivity of the current laboratory experiments.

So far we have assumed that  $S_A$  is due to intrinsic spin. From Eq. (1), if  $S_A$  were orbital angular momentum rather than intrinsic spin, we would expect spinning gyroscopes to have a weight dependent on  $S_A \cdot g/|g|$ , where g is the acceleration of gravity. Although an effect like this (for one sign of  $S_A \cdot g$ ) has been reported [37], more accurate subsequent experiments [38-40] found a null result (an earlier experiment on the intrinsic spin of the neutron was reported in Ref. [21]). The authors of Ref. [40] found  $\Delta m < 9 \times 10^{-8}$  kg for a 0.143 kg rotor whose angular momentum was approximately equal to 7.2  $\times 10^{32}\hbar$ . If we assume a gyroscopic weight is due to  $V_{AB}^{D}$ , and  $\lambda_{\phi} \gg R_{\text{Earth}}$ , then the force derived from Eq. (3) gives  $D < 4.0 \times 10^{-3} \text{ kg}^{-1}$  for the experiment of Ref. [40]. In our experiment, if we consider the orbital motion of the unpaired neutron in <sup>9</sup>Be to act like a gyroscope with orbital angular momentum L, we obtain  $D < 5 \times 10^{-9}$  $kg^{-1}$ , about 10<sup>6</sup> times more stringent a limit on an  $\mathbf{S}_A \cdot \mathbf{g}/|\mathbf{g}|$  effect for angular momentum [4]. From Refs. [37-40], for our experiment, we might expect to replace **S**<sub>A</sub> in Eq. (1) by  $m_{\text{neutron}}r_{\text{eq}}\omega \simeq L/r_{\text{eq}}$ , where  $r_{\text{eq}}$  is defined in Ref. [37] (taken to be  $10^{-13}$  cm here). If we assume  $\lambda_{\phi} \gg R, d$  in Eq. (3), then the resonance test reported here is approximately 10<sup>18</sup> times more sensitive than the acceleration test of Nitschke and Wilworth [40,41].

It is interesting to compare the sensitivity of resonance experiments (such as the type of experiment reported here) with acceleration tests. From Eqs. (3) and (4), we calculate the accelerations  $a_D = -(\partial E_A^D/\partial d)/m_A$  and  $a_T = -(\partial E_A^T/\partial d)/m_A$ , where  $m_A$  is the unit of mass associated with each spin in A. If we use parameters of the

current resonance and acceleration [16] experiments, the limits placed on  $C^{D}$  and  $C^{T}$  are roughly equal when  $\lambda_{\phi}$  is on the order of the size of laboratory experiments. Resonance (acceleration) tests are more sensitive for larger (smaller) values of  $\lambda_{\phi}$ .

Significant improvements in the sensitivity of the <sup>9</sup>Be<sup>+</sup> experiment could be expected. For 100% detection efficiency [34], a resonance time  $T_R$  (time taken to drive the  $v_0$  transition), N stored ions, and total measurement time  $\tau$ , the statistical uncertainty in a frequency measurement is  $\delta v = (4\pi^2 N T_R \tau)^{-1/2}$  [34]. In future experiments  $N = 10^7$ ,  $T_R = 100$  s, and  $\tau = 30$  days do not seem unreasonable, whence  $\delta v_0 \approx 3 \times 10^{-9}$  Hz. In the <sup>9</sup>Be<sup>+</sup> experiments, the pressure shift can be made negligible by using cryogenic pumping. By operating on other field-independent transitions in <sup>9</sup>Be<sup>+</sup> or other ions, we could make  $|\Delta\sigma_{S_z}| \approx 1$  for the electron and be much more sensitive to anomalous couplings to electron spin. The characteristic dimensions of the ion experiments can also be made very small and therefore can investigate relatively short ranges ( $\lambda_{\phi} < 1$  mm).

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