## Relativistic shifts of bound negative-muon precession frequencies

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High-field negative-muon spin precession experiments have been performed using a backward-muon beam with substantial transverse spin polarization, facilitating high-precision measurements of the magnetogyric ratio of negative muons bound to nuclei in the ground states of muonic atoms. These results may provide a testing ground for quantum electrodynamics in very strong electromagnetic fields.

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A lepton bound in a Coulomb potential (e.g., an electron in the ground state of a hydrogenlike atom) experiences a relativistic shift of its spin precession frequency in a given magnetic field. This effect was first calculated by Breit [1] in 1928 assuming pointlike nuclei:

$$\frac{g_{\text{free}} - g}{g_{\text{free}}} = \frac{2}{3} (1 - \sqrt{1 - \alpha^2 Z^2}) \approx \frac{1}{3} \left(\frac{\overline{v}}{c}\right)^2,\tag{1}$$

where  $\alpha$  is the fine structure constant, Z is the atomic number of the nucleus, and  $\overline{v}$  is an effective mean speed of the lepton in its orbit. Breit's approximation was improved upon by Margeneau [2] and later by Ford *et al.* [3,4] in papers accompanying the first experimental measurements of such shifts for negative muons [5]. Subsequent measurements of bound  $\mu^-$  frequency shifts were made by Yamazaki *et al.* [6] in heavier muonic atoms where the finite size of the nucleus is comparable to the radius of the muon wave function, causing the relativistic shift to level off toward a constant value as a function of atomic number.

Although the muon is nominally 207 times closer to the nucleus than an equivalent electron, its relativistic shift (for a pointlike nucleus) should be the same [see Eq. (1)]; thus a comparison of  $g_{\mu}$  with recent measurements of  $g_e$  in single-electron bound states with higher-Z nuclei [7] should directly reveal differences such as finite-nuclear-size effects. The results of Yamazaki *et al.* [6] agreed with theoretical predictions, but higher experimental precision could reveal surprises.

Since then, there have been few measurements or calculations, except for some recent experiments by Mamedov *et al.* [8], in which Yamazaki's experimental result for  $Cd\mu^{-}$  atoms was called into question.

The paucity of new data is largely due to the difficulty of using higher magnetic fields to improve the resolution of relative frequency shifts. The 29.8-MeV/*c* "surface"  $\mu^+$  beams that now dominate most applications of muon spin rotation, relaxation, and resonance ( $\mu$ SR) [9–11] can be

"spin rotated" using Wien filters [12] to orient the muon spins perpendicular to their momenta, thus allowing injection of the beam into arbitrarily high magnetic fields parallel to the momenta but still perpendicular to the spin polarization. Unfortunately, there are no surface  $\mu^-$  beams, because negative pions stopping in the production target are immediately captured by positive nuclei. All polarized negative-muon beams come from "decay channels" in which the  $\pi^-$  decays in flight. Even "backward" muons are generally much higher momentum than the surface  $\mu^+$  from  $\pi^+$  decay at rest, with the result that no "spin rotators" have been built for negativemuon beams.

Recently it was discovered [13] that the backward-muon beam of the M9B superconducting muon channel at TRIUMF can be tuned to give a partially transverse muon spin polarization. The angle between the muon polarization and the beam momentum is around 30°, giving sufficient transverse polarization (nearly 50%) to allow transverse field (TF)  $\mu$ SR experiments in high magnetic fields.

We have utilized this new capability to perform TF- $\mu$ -SR experiments in a 2-T transverse field, giving significantly higher resolution for the relativistic shifts of bound negative muons.

Four identical electron detectors (plastic scintillators coupled to phototubes by plastic light guides) were arranged symmetrically around the sample in an array centered on the axis of a superconducting solenoid with a 6-in.-diam room-temperature access bore, as shown in Fig. 1. A 68-MeV/c  $\mu^{-}$  beam from backward decay of pions passed through the muon counter and a polyethylene degrader to stop in the sample. A time digitizer is started on the signal from the muon counter and stopped on a signal from any electron detector; the corresponding time bin in the corresponding histogram is incremented and the process begins again. Pileup gates reject any event in which a second muon arrives within 16  $\mu$ s of the first one, as well as events in which two electrons are detected within the data gate. We found no direct evidence for distortions in the time spectra due to inefficient pileup rejection, etc., but it was always necessary to include small background signals from muons which were captured on other elements (mostly carbon).

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FIG. 1. Schematic drawing of counter arrangement in the bore of the *Helios* superconducting magnet: incoming muons trigger the small square counter; outgoing decay electrons trigger one of the quadrants of a cylinder. The magnetic field is along the axis (parallel to the muon momentum). A typical sample is shown in dotted lines.

The magnetic field strength was limited to about 2 T in this experiment by our time resolution of  $\sim 1$  ns (which must be a small fraction of the muon precession period) and by the large radius of the cylinder formed by the electron counters (decay electrons have a radius of curvature of about 5 cm at 2 T and may not reach the detectors at higher fields). Neither of these limitations is intrinsic, but the miniaturization required to go to larger fields conflicts with the sample thickness required to stop backward muons, so it would be difficult to work at fields higher than about 5 T.

The field produced by a superconducting magnet was regulated by a temperature-corrected Hall probe to  $\sim 1$  ppm, and a succession of samples were studied without ever changing the field. Early in the experiment the beam polarity was reversed and positive muons were stopped in graphite and aluminum metal to calibrate the precession frequency with known Knight shifts; the  $\mu^+$  frequency in Al, whose shift is only +80±4 ppm [9], was used as a reference.

For each sample, four TF- $\mu$ -SR spectra were simultaneously fitted to a combination of several lifetime components (and, where appropriate, several frequencies) to obtain the best possible determination of the precession frequency in each type of muonic atom. All samples contained a natural distribution of isotopes, so that in some cases the frequency is a weighted average of the frequencies for all the spinless isotopes. (Several nuclei with spin were also studied, but the interpretation of the relativistic shifts in those cases is not obvious [14], so their values are not included here.)

The "raw" fitted frequencies are given in Table I. The largest systematic uncertainty arises from the dependence of the field on the position in the magnet combined with the finite-range straggling of the beam. This was estimated using positive muons by intentionally moving a graphite sample back and forth several cm along the beam direction, producing a frequency shift on the order of 20 ppm per cm at the point of steepest slope within the sample region. For dense samples, the muon stopping position could be controlled with confidence only to within about 1 cm, so the purely TABLE I. Measured frequencies in a constant magnetic field. Only statistical precision is exhibited in this table, to illustrate that the potential accuracy of this technique is much higher than we have achieved in this modest effort.

Sample	Frequency [MHz]
$\mu^+$ in graphite	271.69888±0.00072
$\mu^+$ in Al metal	$271.58520 \pm 0.00038$
$\mu^-$ on <sup>12</sup> C (graphite)	271.3684±0.0016
$\mu^-$ on <sup>16</sup> O (H <sub>2</sub> O)	271.258±0.010
$\mu^-$ on <sup>24</sup> Mg (metal)	$270.9259 \pm 0.0027$
$\mu^-$ on <sup>28</sup> Si	$270.6502 \pm 0.0069$
$\mu^-$ on <sup>32</sup> S (powder)	$270.406 \pm 0.008$
$\mu^-$ on <sup>40</sup> Ca (metal)	$270.164 \pm 0.069$
$\mu^-$ on Ti (metal)	$269.719 \pm 0.066$
$\mu^-$ on Zn (metal)	$268.440 \pm 0.072$
$\mu^-$ on Cd (metal)	$265.73^{+0.46}_{-0.57}$
$\mu^-$ on Pb (metal)	$264.50_{-0.62}^{+0.59}$

statistical uncertainties shown in Table I have been added in quadrature with a systematic uncertainty of about 0.006 MHz to obtain the net uncertainties shown in Table II.

Figure 2 shows the fractional frequency shift of negative muons bound in various muonic atoms, measured in a transverse magnetic field of 2 T, relative to the  $\mu^+$  precession frequency in vacuum, assuming a  $\mu^+$  Knight shift of +80±4 ppm in Al [9].

The points for  $Z \le 30$  are shown on an expanded scale in Fig. 3 and those for  $Z \le 15$  are shown further expanded in Fig. 4 to emphasize the high precision of these measurements. The error bars in Fig. 4 are roughly the size of the points except for Z=8 (oxygen in H<sub>2</sub>O molecules) where precision was limited by the small amplitude of the  $\mu^-$  pre-

TABLE II. Fractional shifts of bound  $\mu^-$  spin precession frequencies in various muonic atoms, relative to that of a free muon in vacuum. All known systematic uncertainties have been included, but no attempt has been made to correct for effects such as chemical (diamagnetic) or paramagnetic shifts of the equivalent (Z-1) impurity in the host material.

Sample	$g_{\mu}$ shift [%]
$\mu^+$ in graphite	$0.0499 \pm 0.0023$
$\mu^+$ in Al metal	$0.0080 \pm 0.0004$
$\mu^{-}$ on <sup>12</sup> C (graphite)	$-0.0718 \pm 0.0023$
$\mu^-$ on <sup>16</sup> O (H <sub>2</sub> O)	$-0.1124 \pm 0.0042$
$\mu^-$ on <sup>24</sup> Mg (metal)	$-0.2348 \pm 0.0025$
$\mu^-$ on <sup>28</sup> Si	$-0.3363 \pm 0.0034$
$\mu^-$ on <sup>32</sup> S (powder)	$-0.4262 \pm 0.0036$
$\mu^-$ on <sup>40</sup> Ca (metal)	$-0.5155 \pm 0.025$
$\mu^-$ on Ti (metal)	$-0.679 \pm 0.024$
$\mu^-$ on Zn (metal)	$-1.150 \pm 0.026$
$\mu^-$ on Cd (metal)	$-2.15_{-0.21}^{+0.17}$
$\mu^-$ on Pb (metal)	$-2.60^{+0.22}_{-0.23}$



FIG. 2. Fractional shifts of bound  $\mu^{-}$  spin precession frequencies in various muonic atoms (see Table II). As in the table, error bars include all known systematic uncertainties, but not diamagnetic or paramagnetic shifts, hyperfine anomalies, or other phenomena. The smooth curve represents the Breit formula (1). Note the strong deviation due to finite nuclear size for high Z.

cession signal, which was a factor of  $0.169\pm0.04$  smaller than that in graphite—even smaller than that reported by Evseev [15] in 1975, which was roughly 40% of the amplitude in graphite. We speculate that the difference may be related to the very high purity and careful degassing of the water sample used in this experiment. In preliminary studies of liquid nitrogen [16], Roduner found no detectable <sup>14</sup>N $\mu$ <sup>-</sup> signal in pure N<sub>2</sub>, but a large signal in N<sub>2</sub> with a small mole fraction of oxygen. This suggests a crucial role for chemical scavenging of hydrated electrons produced in the "Coulomb explosion" as the  $\mu$ <sup>-</sup> cascades down to the 1*s* state in an otherwise inert liquid. Evseev [15] also found plentiful evidence for chemical effects on  $\mu$ <sup>-</sup> depolarization.



FIG. 3. Expanded view of intermediate-Z region of Fig. 2. The deviation from the Breit curve for Zn (Z=30) is mainly due to the finite nuclear size, but for Ca (Z=20) and Ti (Z=22) the differences are probably due Knight shifts.



FIG. 4. Expanded view of low-Z region of Fig. 2.

Further work is required to convert our raw data into corrected magnetogyric ratios that can be critically compared with the predictions of theory [3,6,8]. Mamedov *et al.* [8] list seven contributions to the "anomalous" g factor of the muon in the 1s ground state of a muonic atom with zero nuclear spin:

$$g_{\mu}^{1s} = 2 \left( 1 + \sum_{i=1}^{7} a_{\mu}^{(i)} \right),$$
 (2)

where  $a_{\mu}^{(1)} = 0.001 \ 165 \ 921 \ 4(8)(3) \ (0.7 \ ppm)$  is the familiar radiative correction for the free muon [17],  $a_{\mu}^{(2)}$  is the radiative correction due to the Coulomb field of the nucleus,  $a_{\mu}^{(3)}$ is the relativistic correction,  $a_{\mu}^{(4)}$  is the nuclear polarization correction,  $a_{\mu}^{(5)}$  is "the correction due to polarization in the external magnetic field of the electron shell of the atom,"  $a_{\mu}^{(6)}$ is the diamagnetic shift due to screening by electrons, and  $a_{\mu}^{(7)}$  is the center-of-mass correction. Of all these, the relativistic correction is the largest;  $a_{\mu}^{(2)}$  is less than 2% of  $a_{\mu}^{(3)}$  even for large Z [3],  $a_{\mu}^{(7)}/a_{\mu}^{(2)} \sim m_{\mu}/M$  (where *M* is the mass of the nucleus) [3], and  $a_{\mu}^{(6)}$  is usually smaller than ±100 pm [18]. Mamedov *et al.* state that  $a_{\mu}^{(4)}$  is small compared to  $a_{\mu}^{(3)}$  but they apparently equate  $a_{\mu}^{(5)}$  with the Knight shift, since the latter is the most important correction and it is not otherwise mentioned in Eq. (2).

Knight shifts vary dramatically with the medium, as can be seen from the difference between the  $\mu^+$  frequencies in aluminum ( $K_{\mu^+}$ =+80±4 ppm) and graphite (in our sample,  $K_{\mu^+}$ =+499±23 ppm). When the polarization of core electrons is taken into account, Knight shifts on heavier atoms can be much larger and may depend strongly on many different properties of the medium.

For instance, as Mamedov *et al.* point out, the Knight shift of Cu in brass alloys varies from 0.235% to 0.07% as the Cu fraction decreases from 100% to 32% [19]. Since the muonic atom formed when a  $\mu^-$  is captured on a Zn nucleus in metallic zinc is in all chemical respects equivalent to Cu (and is sometimes written " $\mu$ Cu" for that reason), the appropriate comparison is with NMR of infinitely dilute Cu in Zn.

Unfortunately it is difficult to perform NMR at infinite dilution, so extrapolation of the trend must suffice: in zinc metal it is assumed [8] that  $K_{\mu^-} < 0.07\%$ .

A thorough analysis of these corrections is beyond the scope of this paper. We are therefore not in a position to make a definitive statement about the relativistic shift in  $Cd\mu^{-}$  where the disagreement between previous measurements [6,8] is largest. However, such corrections would have to account for more than half the net shift in cadmium to bring it into agreement with the (0.67±0.22)% value claimed by Mamedov *et al.* [8].

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We invite others to make free use of the improved precision of our measurements.

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