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Precision Measurement of the Hyperfine Interval of Muonium by a Novel Technique: Ramsey Resonance in Zero Field*

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A novel method for measuring the hyperfine interval $\Delta\nu$ of muonium in "zero" magnetic field is described. It consists in applying two successive coherent microwave pulses (lengths τ, separation T) to muonium, and observing the change in muon polarization at $t > T + 2\tau$. By extrapolating two low-pressure runs in Kr we obtain $\Delta\nu(0) = 4463.3013(40)$ MHz, while a joint fit with earlier Chicago Kr data gives $\Delta\nu(0) = 4463.3012(23)$ (0.5 ppm). With $f_\mu/f_p = 3.183\,338(13)$ this implies (a) $\alpha^{-1} - 137 = 0.036\,19(30)$ (2.2 ppm) and (b) a proton polarizability $\delta_N^{(2)} = -(4\pm 4)$ ppm.

The hyperfine interval $\Delta\nu$ of muonium can be determined with high precision by two microwave resonance methods, viz. (a) directly, by observing $F=1 \to F=0$ transitions in "zero" external field,[1] and (b) from the frequencies of suitable Zeeman transitions in a strong external field.[2] Notwithstanding recent advances[3] in the latter technique, the direct method has several potential advantages. These are, however, largely compensated by the fact that a straightforward zero-field resonance[1] yields, for a given signal height, a much larger linewidth Γ than the high-field resonance (e.g., 1400 vs 520 kHz for 90% saturation), while the statistical uncertainty in locating the center of a Lorentzian goes as Γ/h. This large linewidth is connected, as we shall see later, with the three-level nature of the zero-field transitions.

We present here a novel technique which, while yielding linewidths narrower than the high-field resonance (in fact, narrower than the "natural" linewidth $\Gamma_0 = 140$ kHz!), preserves all the advantages of the zero-field method. Our technique is, in virtue of this reduction in linewidth and of several lesser factors, statistically about 25 times more powerful than the (optimized) straightforward resonance in zero field. The latter bears roughly the same relation to the novel method as the Rabi resonance (single oscillating field) for atomic beams does to its modification by Ramsey (two separated oscillating fields).[4] In fact, this work was strongly inspired by that of Ramsey and his associates.

Although the exact treatment of the quantum mechanics underlying our method is extremely simple, let us first describe it qualitatively: Assume that muons, with initial polarization $P_z(0) = 1$, form (in "zero" field) muonium in the $F = 1$, $M = 1$ state. A microwave field B_1 oscillating, say, in the x direction (with frequency $\omega \sim \omega_0 = 2\pi\Delta\nu$) will induce transitions to the $F = 0$, $M = 0$ ground state, and from it back to both $|M| = 1$ states. $P_z(t)$ will vary, reaching -1 when only the $M = -1$ state is populated, and returning to $+1$ later.

In the straightforward method, one detects the resonant depolarization implied by this oscillation of P_z. Notice that $P_z(t)$ goes through zero at a time τ when the populations of the $M = \pm 1$ states become equal. If B_1 is switched off at τ, *then* $\vec{P}$ *will keep oscillating in the (x, y) plane along* $\overleftrightarrow{P}(\tau)$ *with frequency* ω_0 (because one has prepared a coherent superposition of $F = 1$ and $F = 0$ states). If the microwave field is switched on again at a time T later, the $\vec{P}$ is rotated to $P_z = -1$ only if $\vec{B}_1$ and $\vec{P}$ are in the same phase relation to each other at $T + \tau$ as they were at τ. This is clearly a resonance condition for $\Delta = \omega_0 - \omega$. The preceding considerations are illustrated in Fig. 1(a); Fig. 1(b) shows for analogy the case of the Ramsey resonance, where $\vec{P}$ precesses about an external $\vec{B}_0$.

Exact description.—Using the interaction representation and an obvious notation, the equations of motion of the amplitudes are

$$i\dot{a}_{\pm 1} = \pm \Omega e^{i\Delta t} a_{00}, \quad i\dot{a}_{00} = \Omega e^{-i\Delta t}(a_1 - a_{-1}), \tag{1}$$

where the matrix element Ω is proportional to B_1. Evidently $\dot{a}_1 + \dot{a}_{-1} = 0$, and one is dealing with an equivalent two-level problem. In fact, putting

$$C_p \equiv (a_1 - a_{-1})/\sqrt{2}, \quad C_q = a_{00}, \quad b = \sqrt{2}\Omega,$$

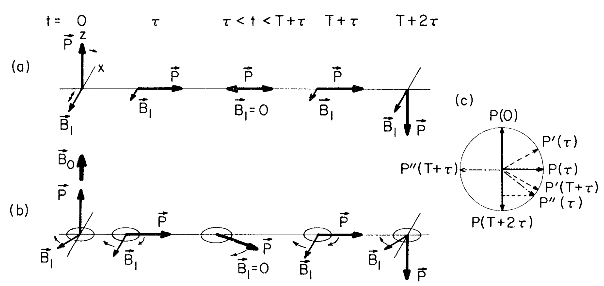

FIG. 1. Motion of polarization $\vec{P}$ in double-pulse experiments: (a) this experiment (without phase shift); (b) Ramsey resonance; and (c) illustration of polarization loss in present experiment. P shows the evolution of the polarization at an "ideal" point in the cavity, P' at a point where B_1 is the weaker, P'' at a point where B_1 is stronger.

(1) is transformed into

$$i\dot{C}_p = bC_q e^{i\Delta t}, \quad i\dot{C}_q = bC_p e^{-i\Delta t}, \tag{1'}$$

the classic Rabi equations. We solve (1) with the initial conditions $a_n = \delta_{n1}$, $P_z(0) = 1$; since the experiments detect $\Delta P_z(t) = P_z(0) - P_z(t)$, the quantity of main interest is $P_z(t) = |a_1|^2 - |a_{-1}|^2 = \sqrt{2}\,\mathrm{Re}C_p$.

For a succession of two pulses of frequency ω and width τ, separated by an interval T, one finds

$$P_z(2\tau + T) = \mathrm{Re}[F^2(\tau)e^{+i\Delta\tau} + 2G^2(\tau)e^{+i\Delta(T+\tau)}] \tag{2}$$

with

$$F(\tau) = \cos(a\tau/2) - i(\Delta/a)\sin(a\tau/2) \to 0,$$

$$G(\tau) = -i(b\sqrt{2}/a)\sin(a\tau/2) \to -i/\sqrt{2},$$

$$a \equiv (\Delta^2 + 4b^2)^{1/2},$$

where the limiting values are for 90° pulses ($a\tau = \pi$) and near resonance ($\Delta \ll b$). Thus, for that situation $P_z(2\tau + T) = -1\cos\Delta(T + \tau)$, again as anticipated in the qualitative argument.[5]

In practice, one could in the usual manner[1] form a signal

$$S(\Delta) = (N_{\mathrm{off}} - N_{\mathrm{on}})/(N_{\mathrm{off}} + N_{\mathrm{on}}) \approx A[P_z(0) - P_z(2\tau + T)]/2P, \tag{3}$$

where the A is the *effective* muon decay asymmetry for the experimental setup, and the N's are the decay positron rates (for $t > 2T + \tau$) with rf on and off, respectively. Note that the peak signal is, because of the complete reversal of $\vec{P}$, *twice* as large as in the straightforward approach. Instead of counting with rf on/off, it is, however, preferable to shift the phase of the second pulse alternately by $\pm\pi/2$, following Ramsey and Silsbee.[4] One then forms the signal

$$S_\delta(\Delta) = [N(\pi/2) - N(-\pi/2)]/[N(\pi/2) + N(-\pi/2)] \approx AG^2 \sin\Delta(T + \tau). \tag{4}$$

The advantages of this signal are (a) it crosses zero at resonance ($\Delta = 0$); (b) it has a full "linewidth" Γ given by $\Gamma(T + \tau) = \pi/3$, i.e., $\Gamma \simeq 50$ kHz for $T + \tau \simeq 3.4$ μsec; and (c) *all* the time is spent with rf on, i.e., collecting real data. Having emphasized the advantages of our approach, we have to mention two factors which slightly decrease its power: (a) By starting to count *after* the second pulse, a fraction $\exp[-(2\tau + T)]$ of the available decays is lost; (b) the complete polarization reversal implied above is possible only for a fixed amplitude B_1. Since any rf field is necessarily inhomogeneous, the desired 90° flip (at $t = \tau$) will occur only in the mean. In some points of the

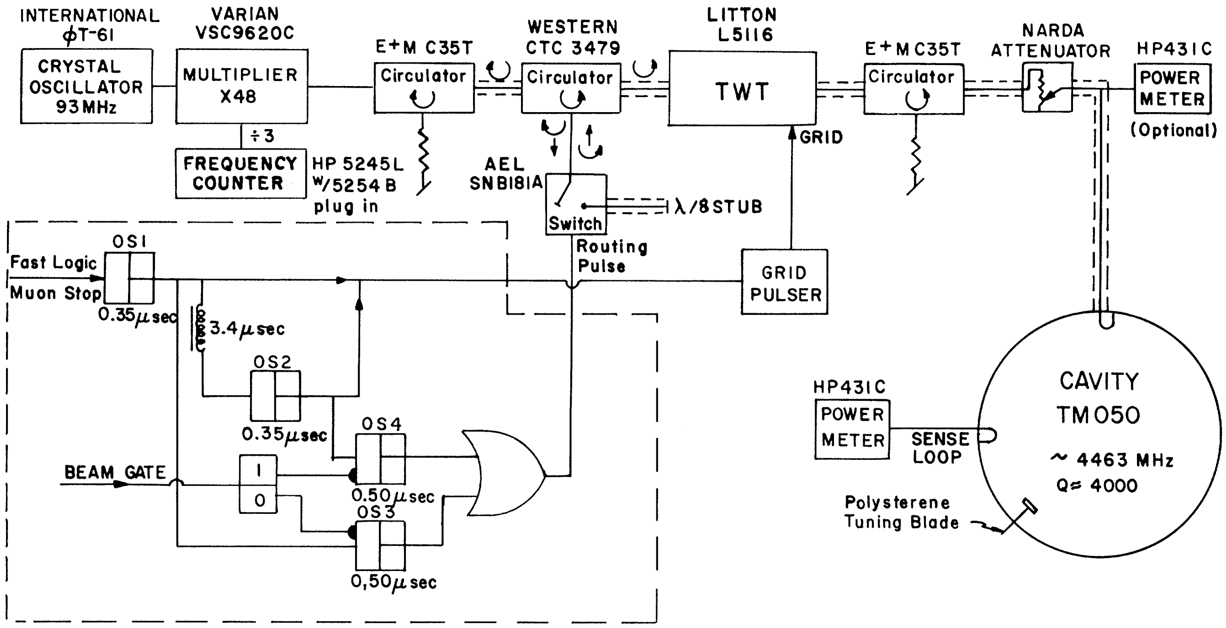

FIG. 2. Microwave logic network. First pulse (of a pair) is fired by one shot OS 1 and shifted by OS 3; second pulse is fired by OS 2, shifted by OS 4. The 3.4-μsec delay between OS 1 and OS 2 is between leading edges, thus setting $T+\tau$ as defined in text.

cavity $\vec{P}$ will be flipped more, in some less; the second pulse will merely double the flip angles, and $P_z(2\tau+T)$ will not attain -1. This situation is illustrated in Fig. 1(c).

Experimental arrangement.—The muonium target and the associated electronics were essentially as described earlier,[3] except that we used an Al cavity, excited in the TM_{050} mode with $Q\approx 4000$. An axial magnetic field of $\lesssim 10$ mG was achieved by three concentric cylindrical shields, similar to Ref. 1. The microwave part of the apparatus, its sole new feature, is illustrated in Fig. 2. The requisite large (1.2-kW) rf pulses with $\nu \simeq 4500$ MHz were generated with a grid-controlled traveling-wave-tube amplifier; each stopping muon, as defined by a fast signature, generated its pair of square grid pulses of fixed width τ_g and separation T. A relative phase shift $\delta = \pm\pi/2$ was introduced into the second rf pulse of each pair by means of a $\lambda/8$ stub. The (stochastic) muon beam consisted of bursts of 0.8 msec length; the sign of δ was alternated with each beam gate, i.e., burst, by the logic shown in Fig. 2.

Data collection.—The time distributions of backward and forward decay positrons for both signs of δ (or rf on/off) were stored over an interval of 10 μsec in four 100-channel banks. First, the grid pulse length τ_g and power level required for a single 90° flip were established empirically by measuring the resultant asymmetry. The signal $S_\delta(\omega)$ was then measured with $\tau_g = 0.3$ μsec and $T+\tau = 3.4$ μsec; the latter value was chosen on the basis of the accidental level, which amounted to $\leqslant 10\%$ for an observation interval of 4.3 μsec past $2\tau+T$.

Data were collected with Ar at 7150 Torr and with Kr at 2742 and 6469 Torr.[6] Typical superstop[3] rates for these runs were 200–600 per sec. The pressure and temperature were monitored continuously by means of a quartz Bourdon gauge and a thermistor, yielding an accuracy in density of better than 0.1%.

Results.—The function $A\sin\Delta(T+\tau)$, with A and Δ as free parameters, was fitted to the signal points, as shown in Fig. 3 for the low-pressure Kr data. This gave

$$\Delta\nu(6469\text{ Torr, Kr}) = 4\,462\,999.78(2.96)\text{ kHz}, \quad (5)$$

$$\Delta\nu(2742\text{ Torr, Kr}) = 4\,463\,173.26(1.90)\text{ kHz}, \quad (6)$$

$$\Delta\nu(7150\text{ Torr, Ar}) = 4\,463\,152.17(2.37)\text{ kHz}. \quad (7)$$

The errors given above are standard deviations based on statistics alone. No effects that would systematically shift the frequency at which the signal crosses zero by an amount significant at

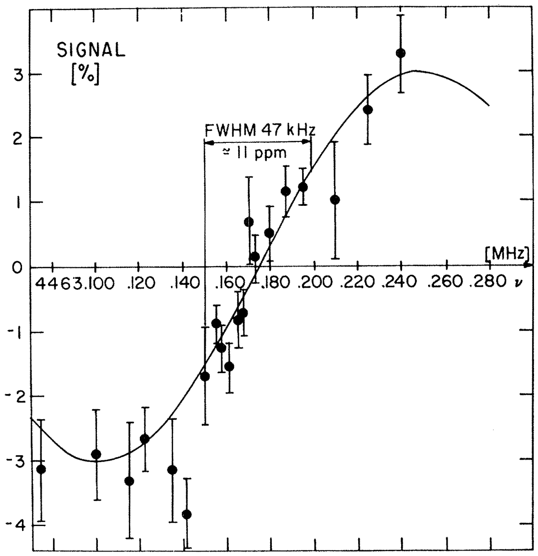

FIG. 3. Data points obtained in Kr at 2742 Torr. Curve is $A\sin\Delta(T+\tau)$ fitted to data points ($T+\tau=3.4$ μsec); $\chi^2/N=1.12$.

this level could be identified. Among the effects considered were (a) unequal amplitudes of the two pulses, (b) departure of δ from $\pm 90°$, (c) difference between the cavity and the oscillator frequencies, and (d) effects of static fields.

Extrapolating (5) and (6) to zero density, one obtains

$$\Delta\nu(0\text{ Torr, Kr}) = 4\,463\,301.33(3.95)\text{ kHz (0.88 ppm)} \tag{8}$$

and a coefficient of fractional (linear) pressure shift (FPS)

$$C_{\rm FPS}(\text{Kr}) = -10.47(0.21)\times 10^{-9}\text{ Torr}^{-1}. \tag{9}$$

Note that both (8) and (9) are in excellent agreement with earlier, less accurate results both from this laboratory[3b] and from Yale[7]; the FPS furthermore agrees remarkably well with the value $-10.4(2)\times 10^{-9}$ Torr^{-1} reported[8] for atomic H in Kr. It appears hence appropriate to fit (5), (6), and the data of Ref. 3b jointly; this yields

$$\Delta\nu(0\text{ Torr, Kr}) = 4\,463\,301.17(2.3)\text{ kHz (0.5 ppm)} \tag{10}$$

and

$$C_{\rm FPS}(\text{Kr}) = -10.37(0.7)\times 10^{-9}\text{ Torr}^{-1}. \tag{11}$$

A quadratic pressure shift of the magnitude suggested in Ref. 7 for Kr (see following Letter[9]) would not affect (10) by more than the quoted uncertainty.

Assuming that the FPS for muonium in Ar also has the "atomic" value $-4.78(3)\times 10^{-9}$,[10] we extrapolate (7) linearly, obtaining

$$\Delta\nu(0\text{ Torr, Ar}) = 4\,463\,304.71(2.56)\text{ kHz (0.6 ppm)}, \tag{12}$$

in accord both with (10) and earlier work.[3,7] Nevertheless, we shall use only (10) for the following analysis.

To extract α from $\Delta\nu(0)$ one needs a value for the muon magnetic moment. Adopting $f_\mu/f_p = 3.183\,347(9)$ as reported by Hague *et al.*,[11] one obtains

$$1/\alpha - 137 = 0.036\,38(19)\ (1.7\text{ ppm}), \tag{13}$$

whereas our result[3b] $f_\mu/f_p = 3.183\,338(13)$ yields

$$1/\alpha - 137 = 0.036\,19(30)\ (2.2\text{ ppm}). \tag{14}$$

While these two results are in good agreement, it must be remembered that (14) contains no allowance for the g_j-shift uncertainties discussed in Ref. 3b. The current value without quantum electrodynamics is 0.036 08(26).[12]

From the ratio $\Delta\nu(\mu e)/\Delta\nu(pe)$ one can compute either f_μ/f_p or the proton polarizability $\delta_N^{(2)}$:

$$f_\mu/f_p = 3.183\,326(13)\ (\text{with } \delta_N^{(2)}=0), \tag{15}$$

$$\delta_N^{(2)} = -(4\pm 4)\text{ ppm} \quad [\text{with } f_\mu/f_p = 3.183\,338(13)]. \tag{16}$$

The smallness of $\delta_N^{(2)}$ may be taken as evidence for the recently computed recoil corrections[13] which contribute 5.6 ppm to $\Delta\nu(\mu e)$.

The novel technique described here will allow one to push the accuracy in $\Delta\nu(0)$ much further yet. A corresponding improvement in the knowledge of the muon moment will, however, be needed to make that accuracy useful.

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Quadratic hfs Pressure Shifts of Muonium in Argon and Krypton*

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The very precise, low-pressure measurements of the hfs interval $\Delta\nu(p)$ of muonium in Ar and Kr described in the preceding Letter are combined with the high-pressure measurements from Yale to determine the magnitude of quadratic pressure shifts. Fitting by the form $\Delta\nu=\Delta\nu(0)[1+ap+bp^2]$, we find $b(\mathrm{Ar})=7.1(2.3)$ and $b(\mathrm{Kr})=8.7(3.1)$ (in units of 10^{-15} Torr^{-2}). Older evidence for these quadratic shifts from similar fits involving earlier Chicago data is reviewed.

In the preceding Letter[1] we have described a novel method for measuring the hyperfine interval $\Delta\nu$ of muonium, and presented the most accurate determinations (<0.6 ppm) of $\Delta\nu(p)$ obtained to date in gases at very low pressure (<10 atm).

In this note we wish to discuss the impact of these accurate determinations on the question of a quadratic term in the dependence of $\Delta\nu$ on p. We shall also comment on a recent Letter of the Yale group[2] on the same topic.

The effort of the Chicago group[1,3-5] has in the past been directed towards *avoiding* quadratic pressure shifts, rather than measuring their magnitude. To this purpose, we have consistently worked at gas densities sufficiently low that such effects do not invalidate a *linear* extrapolation to $\Delta\nu(0)$ beyond the quoted (statistical) error, and have developed the special techniques required with currently available muon beams for achieving high accuracy at such densities. For this reason, our own data can provide only precise measurements of the coefficient[6] a in the usual expression $\Delta\nu(p)=\Delta\nu(0)[1+ap+bp^2]$; to obtain b, the high-pressure data of the Yale group[2,7] —who worked mostly at high densities—have to be fitted jointly with ours. Such fits yield accurate results for a and b for Kr as well as for Ar; these results are summarized in the top two rows of Table I. They show (1) good agreement