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Line-shape effects in the measurement of the positronium hyperfine interval

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Recently Rich has pointed out that annihilation terms in the effective 4×4 Hamiltonian H for $n=1$ positronium cause the real parts of the Zeeman eigenvalues to be shifted by terms of order $(\lambda_s/4\pi\Delta\nu)^2\approx10^{-5}$ relative to the Breit-Rabi eigenvalues. Here λ_s is the annihilation rate of the singlet state and $\Delta\nu$ is the hyperfine interval. Rich observes that the $\Delta\nu$ measurements have not correctly dealt with decay. The Zeeman-resonance line shape is calculated here assuming that the non-Hermitian H describes the motion of the four $n=1$ levels via Schrödinger's equation. The deviations of this line shape from a Lorentzian are exhibited. The asymmetry of the line causes a shift in the line center relative to what one would obtain from a Breit-Rabi plus Lorentzian fit to a measured Zeeman-resonance curve. To take this into account, the measurement of $\Delta\nu$ by A. P. Mills, Jr. and G. H. Beaman [Phys. Rev. Lett. 34, 246 (1975)] should be increased by 2.5 ppm to $\Delta\nu$(Mills and Beaman)$=203.3875(16)$ GHz. When the Egan *et al.* measurement [P. O. Egan, V. W. Hughes, and M. H. Yam, Phys. Rev. A 15, 251 (1971)], which used a different line shape, is reinterpreted in terms of the line shape calculated here, the Egan *et al.* $\Delta\nu$ value increases by 21 ppm to $\Delta\nu$ (Egan *et al.*)$=203.3890(12)$ GHz. The weighted mean of the two corrected measurements is $\Delta\nu=203.3885(10)$ GHz.

I. INTRODUCTION

The question of whether decay terms have been correctly taken into account in precision measurements of the positronium hyperfine interval $\Delta\nu$ has been examined recently by Rich.[1,2] He points out that the real parts of the Zeeman eigenvalues are shifted by terms of order $(\lambda_s/4\pi\Delta\nu)^2\approx10^{-5}$ relative to the Breit-Rabi eigenvalues, where λ_s is the annihilation rate of the singlet ground state. Rich's conclusion that the $\Delta\nu$ measurements need to be increased to account for the shift in the eigenvalues is qualitatively correct. However, as pointed out by Rich, what is measured is not an eigenvalue but rather the line center of the positronium Zeeman resonance. It turns out that the resonance line shape is slightly asymmetric and this causes the $\Delta\nu$ measurements to be shifted. It is the purpose of this paper to examine the question of decay shifts in more detail, to display the resonance asymmetry, and to obtain accurate values for any corrections that should be applied to the experiments.

The problem of understanding the behavior of the decay-unstable positronium atom in an external magnetic field may be posed as four separate questions. First, is it a good approximation to say that the time development of the four ground-state positronium sublevels can be accurately represented by a four-level system with a 4×4 non-Hermitian Hamiltonian or mass matrix H? The answer to this question is apparently "yes" since Fontana and Lynch[3] show that the mass matrix is a good approximation as long as the decay energy $(2mc^2)$ is much greater than the energy structure of the states $(\sim h\Delta\nu)$. In that case we may go on to ask what is the correct choice of 4×4 Hamiltonian H_0 in the absence of external fields, and what additional matrix V describes the external perturbations? Finally, what is the correct way to calculate the time development so that we can understand the experimental observations? Although the Hamiltonian currently used[4–9] may be precise enough for ~1 ppm measurements of $\Delta\nu$, there are some small errors in the literature in the calculation of the time development of the four-level system,[10] and in one experiment these errors have found their way into the line shape which has been fitted to the data. In another experiment, the time development was calculated correctly, but the line shape was incorrectly assumed to be symmetric. Thus the experiments are indeed in need of correction as suspected by Rich. In the remainder of this paper, there will be first a qualitative discussion of possible magnetic field corrections to H, followed by a review of how to calculate the time development. Finally, from explicit numerically precise calculations of the Zeeman resonance, we will find corrected values for the recent $\Delta\nu$ measurements.

II. THE HAMILTONIAN

The positronium[11] ground state is split into two sublevels: a singlet $|{}^1S_0\rangle$, which annihilates into two photons (2γ's) at a rate $\lambda_s = 8\times10^9\ \mathrm{sec}^{-1}$; and a triplet $|{}^3S_1\rangle$, which annihilates into 3γ's at a rate $\lambda_t = 7\times10^6\ \mathrm{sec}^{-1}$. The difference in energy of these two levels is the hyperfine interval $\Delta\nu \sim 203$ GHz. A uniform magnetic field $\vec{\mathrm{B}}$ causes a quadratic Zeeman splitting f_{01} of the $m=0$ and ±1 triplet levels, with the Breit-Rabi formula for f_{01} being

$$f_{01} = \tfrac{1}{2}\Delta\nu[(1+4x^2)^{1/2}-1]\ , \qquad (1)$$

where $x=\mu_B g' B/h\Delta\nu$ and g' is the bound-state electron g factor to be discussed below. The Zeeman splitting is measured by applying a microwave magnetic field and observing the resonant increase in 2γ's that occurs when the conditions of Eq. (1) are fulfilled. Essentially, Eq. (1) is then solved to find a value of the hyperfine interval $\Delta\nu$.

To agree with what we mean by the hyperfine interval and the singlet and triplet decay rates, we choose the 4×4 Hamiltonian H_0 in the absence of external fields to be diagonal in the angular momentum J-diagonal basis $|{}^3S_1\, m=0\rangle$, $|{}^1S_0\, m=0\rangle$, $|{}^3S_1\, m=\pm1\rangle$:

$$H_0 = \mathrm{diag}(2\pi\Delta\nu - i\lambda_t, -2\pi\Delta\nu - i\lambda_s\ , \\ 2\pi\Delta\nu - i\lambda_t, 2\pi\Delta\nu - i\lambda_t)\hbar/2\ , \qquad (2)$$

where the zero of energy has been chosen midway between the two levels. The three independent parameters $\Delta\nu$, λ_s, and λ_t are being intensively studied both theoretically and experimentally. In Figs. 1(a) and 1(b) we see the Feynman diagrams contributing in lowest order to $\Delta\nu$. Diagram (a) is the Fermi contact interaction between the positron and electron magnetic moments and (b) is the virtual annihilation contribution to the triplet level. Diagrams (c) and (d) represent singlet and triplet annihilation. Typical higher-order diagrams are shown in Figs. 1(e)–1(h). In principle, the quantities $\Delta\nu$, λ_s, and λ_t can be found to arbitrary precision by including the contributions of sufficiently complicated diagrams.

Now we wonder what happens when an external magnetic field $\vec{\mathrm{B}}$ is turned on. The lowest-order interaction $\vec{\mu}\cdot\vec{\mathrm{B}}$ is shown in Fig. 1(i). Since the ${}^3S_1$ and ${}^1S_0$ states have opposite C parity and the photon has odd C parity, it is clear that this interaction mixes singlet and triplet and is not diagonal in our chosen representation. If we stop here and set

$$\vec{\mathrm{B}} = B_1\hat{e}_1\cos\omega t + B_3\hat{e}_3\ , \qquad (3)$$

then the interaction Hamiltonian $V=\vec{\mu}\cdot\vec{\mathrm{B}}$ has matrix elements

$$V = \begin{bmatrix} 0 & x & 0 & 0 \\ x & 0 & z & -z \\ 0 & z & 0 & 0 \\ 0 & -z & 0 & 0 \end{bmatrix} h\Delta\nu\ , \qquad (4)$$

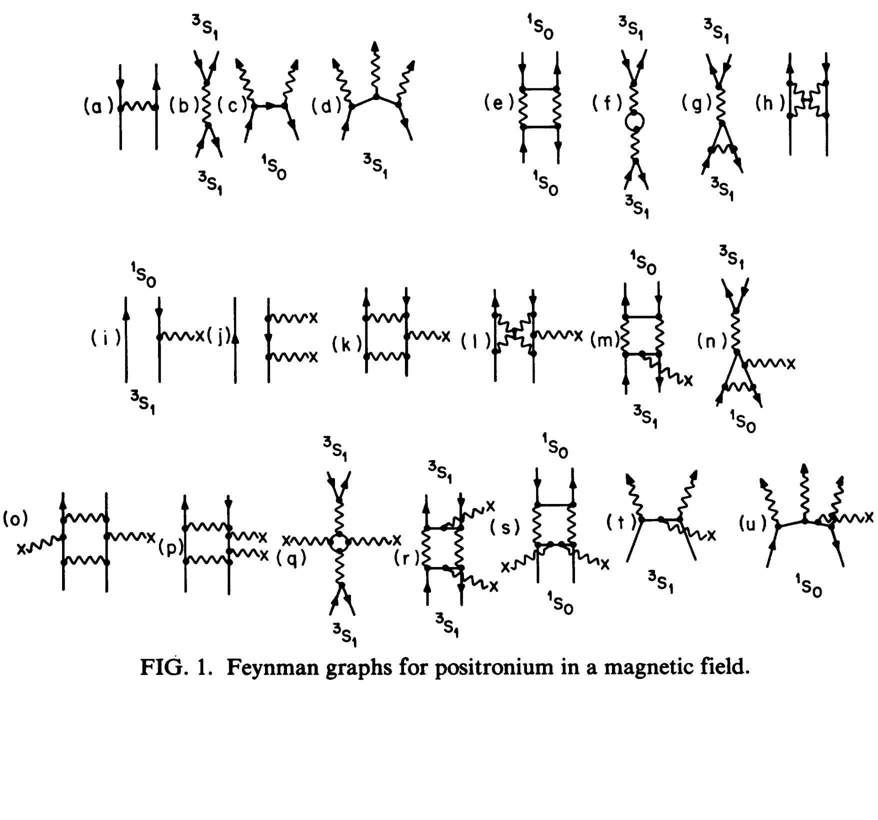

FIG. 1. Feynman graphs for positronium in a magnetic field.

where $x = B_3(g\mu_B/h\Delta\nu)$,

$$z=(\tfrac{1}{2})^{1/2}B_1(g\mu_B/h\Delta\nu)\cos\omega t \equiv 2y\cos\omega t\ ,$$

and $\frac{1}{2}g\mu_B$ is the magnetic moment of the free electron.

We must now see if there are any other diagrams with a magnetic field interaction that contribute significantly to the mass matrix. We may add extra magnetic interactions to any of the diagrams, but we must not include separable diagrams like Fig. 1(j) because these are accounted for by including the effects of V to higher orders in perturbation theory. However, nonseparable diagrams such as Figs. 1(k) and 1(l) do make off-diagonal contributions to V of order $\alpha^2\vec{\mu}\cdot\vec{B}$. Such terms give rise to a bound-state correction to the g factor which has been known for some time.[12] This effect is included in V by changing g to g' in Eq. (4) with

$$g'=g(1-\tfrac{5}{24}\alpha^2)\ . \tag{5}$$

Virtual annihilation diagrams linear in $\vec{\mu}\cdot\vec{B}$ such as Figs. 1(m) and 1(n) make a contribution to g' which is higher order than $\alpha^2\vec{\mu}\cdot\vec{B}$.[12] Thus at the 1-ppm level the part of V linear in B is apparently adequately represented by Eq. (5).

When two magnetic field interactions are considered, we get diagrams like Figs. 1(o)–1(s). The contributions to V are diagonal and proportional to B^2. A crude guess of the magnitude of such contributions would be somewhere between $\alpha^2(\mu\cdot B)^2/h\Delta\nu$ for diagrams (o) and (p) and $\alpha^2(\mu\cdot B)^2/mc^2$ for diagrams (q)–(s). This is α^2 to α^6 times the triplet $m=0$ to 1 Zeeman splitting. Thus it is not safe to ignore such terms until a better estimate is available.

Finally, there are magnetic field corrections to the annihilation rates as in Figs. 1(t) and 1(u). These corrections to the rates will also be quadratic in B because there is no interference between these amplitudes and the amplitudes for Figs. 1(c) and 1(d). The amplitude for the triplet annihilation into 2γ's is probably very small because the internal fermion line is far from the mass shell. However, one of the 3γ's in diagram (u) may be soft, thus possibly leading to a significant ${}^1S_0\rightarrow 3\gamma$ rate proportional to B^2.

These new diagonal contributions to V, which are quadratic in the applied magnetic field B, may be denoted by

$$V'=\mathrm{diag}(\beta_1,\beta_2,\beta_3,\beta_4)x^2h\Delta\nu\ , \tag{6}$$

where the β's are unknown complex constants. For a complete understanding of the Zeeman resonance experiments it is just as important to calculate the β's to first order as it is to complete the $O(\alpha^2)$ correction to $\Delta\nu$. For the present we shall calculate ignoring the possible consequences of $V'\neq 0$.

III. LINE-SHAPE CALCULATION

The time development of the positronium ground-state wave function $|\psi\rangle$ is predicted by Schrödinger's equation

$$i\hbar\partial_t|\psi\rangle = H|\psi\rangle \tag{7}$$

with H given by Eqs. (2)–(5). The eigenvalues of H with no rf ($z=0$) are complex and irrational functions of x, $\Delta\nu$, λ_s, and λ_t. Furthermore, the left and right eigenvectors of H are distinct since H is not Hermitian. It is more convenient[9] to rewrite Eq. (7) in terms of the 4×4 density matrix ρ,

$$i\hbar\dot{\rho}=H\rho-\rho H^{\dagger}\ , \tag{8}$$

which may be transformed to Liouville form

$$\dot{\vec{X}}(t)=2\pi\Delta\nu\overleftrightarrow{C}\cdot\vec{X}(t)\ , \tag{9}$$

where $\vec{X}$ is a 16-component vector and $\overleftrightarrow{C}$ a 16×16 matrix which is time dependent if $z\neq 0$. In the rotating-frame approximation[13] two of the components of $\vec{X}$ decouple from the rest for small z and we are left with a 14-component version of Eq. (9) with a constant $\overleftrightarrow{C}$. The solution is then

$$\vec{X}(t)=\exp(2\pi\Delta\nu\overleftrightarrow{C}t)\cdot\vec{X}(0)\ . \tag{10}$$

In the J-diagonal representation of Eq. (2) the components of $\vec{X}$ and $\overleftrightarrow{C}$ are as follows:

$$\begin{aligned}\vec{X}=[&\rho_{11},\rho_{22},\rho_{33},\rho_{44},(\tfrac{1}{2})^{1/2}(\rho_{12}+\rho_{21}),-i(\tfrac{1}{2})^{1/2}(\rho_{12}-\rho_{21}),(\tfrac{1}{2})^{1/2}(s\rho_{13}+s^*\rho_{31})\ ,\\ &-i(\tfrac{1}{2})^{1/2}(s\rho_{13}-s^*\rho_{31}),(\tfrac{1}{2})^{1/2}(s^*\rho_{14}+s\rho_{41}),-i(\tfrac{1}{2})^{1/2}(s^*\rho_{14}-s\rho_{41}),(\tfrac{1}{2})^{1/2}(s\rho_{23}+s^*\rho_{32})\ ,\\ &-i(\tfrac{1}{2})^{1/2}(s\rho_{23}-s^*\rho_{32}),(\tfrac{1}{2})^{1/2}(s^*\rho_{24}+s\rho_{42}),-i(\tfrac{1}{2})^{1/2}(s^*\rho_{24}-s\rho_{42})]\ ,\end{aligned} \tag{11}$$

$$
\overleftrightarrow{C}=\left(\begin{array}{cccccc|cccc|cccc}
-\lambda_t & 0 & 0 & 0 & 0 & -\sqrt{2}x & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -\lambda_s & 0 & 0 & 0 & \sqrt{2}x & 0 & 0 & 0 & 0 & 0 & -\sqrt{2}y & 0 & \sqrt{2}y \\
0 & 0 & -\lambda_t & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{2}y & 0 & 0 \\
0 & 0 & 0 & -\lambda_t & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -\sqrt{2}y \\
0 & 0 & 0 & 0 & -\bar{\lambda} & 1 & 0 & -y & 0 & y & 0 & 0 & 0 & 0 \\
\sqrt{2}x & -\sqrt{2}x & 0 & 0 & -1 & -\bar{\lambda} & y & 0 & -y & 0 & 0 & 0 & 0 & 0 \\
\hline
0 & 0 & 0 & 0 & 0 & -y & -\lambda_t & -\Omega & 0 & 0 & 0 & x & 0 & 0 \\
0 & 0 & 0 & 0 & y & 0 & \Omega & -\lambda_t & 0 & 0 & -x & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & y & 0 & 0 & -\lambda_t & \Omega & 0 & 0 & 0 & x \\
0 & 0 & 0 & 0 & -y & 0 & 0 & 0 & -\Omega & -\lambda_t & 0 & 0 & -x & 0 \\
\hline
0 & 0 & 0 & 0 & 0 & 0 & 0 & x & 0 & 0 & -\bar{\lambda} & -1-\Omega & 0 & 0 \\
0 & \sqrt{2}y & -\sqrt{2}y & 0 & 0 & 0 & -x & 0 & 0 & 0 & 1+\Omega & -\bar{\lambda} & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & x & 0 & 0 & -\bar{\lambda} & -1+\Omega \\
0 & -\sqrt{2}y & 0 & \sqrt{2}y & 0 & 0 & 0 & 0 & -x & 0 & 0 & 0 & 1-\Omega & -\bar{\lambda}
\end{array}\right) \tag{12}
$$

where $s=e^{i\omega t}$, $\Omega=\omega/2\pi\Delta\nu$, $\bar{\lambda}=\frac{1}{2}(\lambda_s+\lambda_t)$, and the λ's are now normalized by $\lambda_i \rightarrow \lambda_i/2\pi\Delta\nu$.

The signal of interest is the 2γ annihilation yield S given by

$$
\begin{aligned}
S&=(\lambda_s/2\pi\Delta\nu)\int_0^\infty \vec{Y}\cdot\vec{X}(t)dt \\
&=\lambda_s\vec{Y}\cdot(\overleftrightarrow{C})^{-1}\cdot\vec{X}(0)/2\pi\Delta\nu\ ,
\end{aligned} \tag{13}
$$

where

$$\vec{Y}=(0,1,0,0,0,\ldots)$$

and

$$\vec{X}(0)=(\tfrac{1}{4},\tfrac{1}{4},\tfrac{1}{4},\tfrac{1}{4},0,0,\ldots)$$

if the positron polarization is taken to be zero. The line shape S in the low-rf limit is thus a ratio of two polynomial expressions in ω, x, y, $\Delta\nu$, λ_s, and λ_t, fourth order in y, and tenth order in x. Because it is difficult to keep the rf power level fixed while sweeping its frequency, the experiments explore the Zeeman resonance at a fixed rf frequency ω by varying the magnetic field B, to which the parameter x is proportional. The Zeeman-resonance line shape $S(x)$ has been calculated by inverting the matrix $\overleftrightarrow{C}$ numerically using 17-decimal digit precision. The parameters were chosen to be $\omega/2\pi=3.25$ GHz, $\Delta\nu=203.385$ GHz, $\lambda_s=8.0$ GHz, $\lambda_t=(1/142.0)$ GHz. A typical resonance for rf amplitude $y=10^{-4}$ is shown in Fig. 2. The effect of the nonconstant background can be removed by subtracting from $S(x)$ a curve $S_0(x)$ obtained using $y=0$

$$R(x)=S(x)-S_0(x)\ , \tag{14}$$

as shown in the inset of Fig. 2. For $y=10^{-5}$ the peak in $R(x)$ occurs at a value of x, $x_p=0.127\,416\,1501$, which is 0.52 ppm below the value of x which satisfies the Breit-Rabi condition of Eq. (1), $x_0=0.127\,416\,2159$. If an experiment was actually measuring the peak of the resonance, then one should increase the value of $\Delta\nu$ deduced from Eq. (1) by twice this amount or 1.04 ppm. This is the line-shape correction taken into account in Ref. 14 which reported $\Delta\nu=203.3870(16)$ GHz. However, this is not the proper line-shape correction to use because the Zeeman resonance is slightly asymmetric as well as having its peak position shifted slightly. The experiments are sensitive to this asymmetry because the line center is determined mostly from data points taken where the slope of the resonance is greatest. In Ref. 14 the fitted line shape was a Lorentzian (actually the first difference of a Lorentzian). To find the proper line-shape correction we fit a Lorentzian

$$L(x)=a/[1+(x-x_p)^2/\Delta x^2]$$

to $R(x)$. Figure 3 shows $R(x)$ and the difference $R(x)-L(x)$ using the same parameters as in Fig. 2,

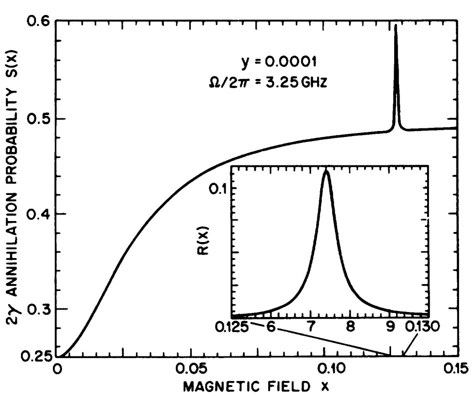

FIG. 2. Calculated 2γ annihilation probability $S(x)$ for positronium in the presence of a variable longitudinal magnetic field proportional to x, and a transverse rf magnetic field with amplitude y and fixed frequency $\Omega=2\pi(3.25)$ GHz. The inset shows the resonance line shape $R(x)$ obtained by subtracting from $S(x)$ a curve $S_0(x)$ calculated using $y=0$.

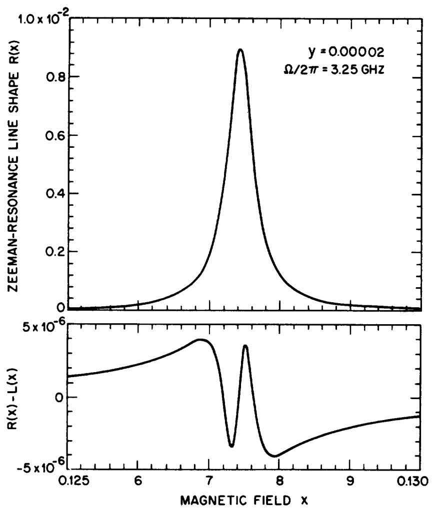

FIG. 3. Calculated Zeeman-resonance line shape $R(x)$ for $y=2\times10^{-5}$ and $\Omega=2\pi(3.25)$ GHz, and the difference between $R(x)$ and a least-squares-fitted Lorentzian curve $L(x)$.

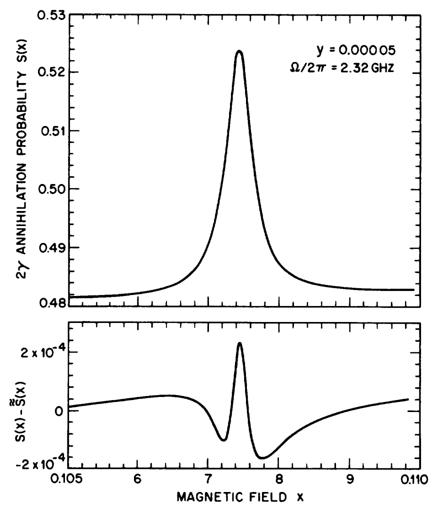

FIG. 4. Calculated 2γ annihilation probability $S(x)$ in the vicinity of the Zeeman resonance for $\Omega=2\pi(2.32)$ GHz, and the difference between $S(x)$ and a least-squares-fitted line shape $\tilde{\tilde{S}}(x)$ discussed in the text.

except $y=2\times10^{-5}$ which gives a reasonable approximation to the signal amplitude observed in Ref. 14. The fitted line center is $x_p=0.127\,415\,9551$, which is 2.05 ppm lower than x_0. When the fit is made including a sloping background (the procedure followed in Ref. 14), we get $x_p=0.127\,415\,9849$, which is 1.81 ppm below x_0. In Ref. 14, then, the line-shape correction should have been + 3.62 ppm rather than the 1 ppm actually used. The properly adjusted value of $\Delta\nu$ is $\Delta\nu=1.000\,0026\times203.3870(16)=203.3875(16)$ GHz.

We next turn our attention to the precision measurements of $\Delta\nu$ reported by Egan, Hughes, and Yam to find out if any line-shape corrections are needed. We will consider only the latest and most precise measurement of Ref. 8, where it is reported that a complete line shape $\tilde{S}(x)$ was fitted to the data:

$$\tilde{S}(x)=\frac{\lambda_{10,2}}{\lambda_{10}}\left[\tfrac{1}{4}+(\lambda_t+\lambda_{10})\left[\frac{1}{2\lambda_t}-\frac{1}{\lambda_{10}}\right]V^2/W^2\right]+\tfrac{1}{4}\,, \tag{15}$$

where

$$W^2=(\omega-\omega_{01})^2+\tfrac{1}{4}(\lambda_t+\lambda_{10})^2+2V^2(\lambda_t+\lambda_{10})^2/(\lambda_{10}\lambda_t)\,, \tag{16}$$

$$V^2=(2\pi\Delta\nu)^2x^2y^2[1+4x^2+(1+4x^2)^{1/2}]^{-1}\,, \tag{17}$$

$$\lambda_{10}=\lambda_t+(\lambda_s-\lambda_t)T(x)\,, \tag{18}$$

$$\lambda_{10,2}=\lambda_sT(x)\,, \tag{19}$$

$$T(x)=\frac{[1+2x^2-(1+4x^2)^{1/2}]}{[1+4x^2-(1+4x^2)^{1/2}]}\,, \tag{20}$$

where $\omega_{01}=2\pi f_{01}$ of Eq. (1) and where x, y, λ_s, λ_t, and ω are defined above, i.e., $x=B_3g'\mu_B/h\Delta\nu$, etc. For $y=0$ we have

$$\tilde{S}(x)=\frac{1}{4}+\frac{1}{4}\left[1+\frac{\lambda_t}{\lambda_s}\left[\frac{1}{T(x)}-1\right]\right]^{-1}. \tag{21}$$

Since this expression is irrational it is not exactly correct as explained above and in Ref. 9, and therefore the measurement of Ref. 8 will also need a correction. One in the derivation of Eq. (15) is that the eigenvalues[15] used in solving Eq. (7) did not take into account the decay shifts pointed out by Rich.[1,2] Furthermore, while there is nothing wrong with using the Breit-Rabi eigenvectors as a basis, *the off-diagonal decay matrix elements in this basis are not zero* as was tacitly assumed in Ref. 8.

To determine the line-shape correction appropriate to the measurement of Ref. 8, we first calculate $S(x)$ from Eq. (13) using $\Delta\nu=203.385$ GHz, $\omega/2\pi=2.32$ GHz, $y=5\times10^{-5}$, and λ_s and λ_t as before. We then fit the function

$$\tilde{\tilde{S}}(x)\equiv a\tilde{S}(x')+b+c(x'-x_0)$$

to $S(x)$ using as adjustable parameters $\Delta\nu'$, y', a, b, and c with $x'=(\Delta\nu/\Delta\nu')x$. $S(x)$ and the difference between $S(x)$ and the best fitting $\tilde{\tilde{S}}(x)$ are plotted in Fig. 4. The best-fit parameters are $\Delta\nu'=203.380\,92$, $y'=5.26\times10^{-5}$, $a=1.000\,01$,

$b=-4.1\times10^{-5}$, and $c=1.8\times10^{-2}$. Since $\Delta\nu'$ is 0.00408 GHz or about 21 ppm lower than $\Delta\nu$ we conclude that the final value of $\Delta\nu$ reported in Ref. 8 should be increased to $\Delta\nu=203.3849(12)+0.0041=203.3890(12)$ GHz. This is very close to the corrected $\Delta\nu$ from Ref. 14, $\Delta\nu=203.3875(16)$ GHz. The weighted mean of these two values is $\Delta\nu=203.3885(10)$ GHz.

This corrected $\Delta\nu$ must be treated with caution because the fitted line centers depend somewhat on the range over which the fit is made. It is to be hoped that the data of future measurements will be interpreted directly in terms of the more correct line shape discussed here. Finally, it is important to emphasize that a complete understanding of the positronium Zeeman resonance at the 1-ppm level needs a calculation of the α^2 corrections to $\Delta\nu$ and at least a good estimate of the quadratic magnetic field contributions to the four-level effective Hamiltonian.

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[15]See Eq. (11) of Ref. 6.