PHYSICAL REVIEW A VOLUME 2, NUMBER 3 SEPTEMBER 1970

Precision Redetermination of the Fine-Structure Interval of the Ground State of Positronium and a Direct Measurement of the Decay Rate of Parapositronium*

E. D. Theriot, Jr.,[†‡] R. H. Beers,[§] V. W. Hughes, and K. O. H. Ziock[||]

Gibbs Laboratory, Yale University, New Haven, Connecticut 06520
(Received 17 September 1969)

A precision redetermination of the fine-structure interval of the ground state of positronium, $\Delta\nu$, has been made in an experiment similar to earlier experiments. The frequency difference between the $M=\pm 1$ and the $M=0$ Zeeman levels of orthopositronium was measured in a static magnetic field of about 7900 G. The dependence of $\Delta\nu$ on gas pressure (argon) was observed for the first time. The result for $\Delta\nu$ is $\Delta\nu_{\text{exp}}=(2.034\,03\pm 0.000\,12)\times 10^5$ Mc/sec, where the error quoted is one standard deviation and represents an improvement in accuracy by a factor of 4 compared to earlier work. This value is in reasonable agreement with the theoretical value of $\Delta\nu_{\text{theor}}=2.034\,27\times 10^5$ Mc/sec, but a calculation of the α^4-Ry term in the theory is needed. The result for the fractional fine-structure pressure shift is $(1/\Delta\nu)(\partial\Delta\nu/\partial P)_{300^\circ\text{K}}=(-0.93\pm 0.18)\times 10^{-7}$/Torr of Ar, in which a one-standard-deviation statistical error is quoted. This value has the same sign but is 20 times larger than the fractional hfs pressure shift for hydrogen in argon. The experiment also yields the first direct measurement of the parapositronium annihilation rate $\lambda_{p\,\text{expt}}=0.799\pm 0.011\times 10^{10}$ sec^{-1}, where a one-standard-deviation error is given. This value is in excellent agreement with the theoretical value of $\lambda_{p\,\text{theor}}=0.798\times 10^{10}$ sec^{-1}.

I. INTRODUCTION

Positronium, the bound state of an electron and a positron, is an ideal system for a test of quantum electrodynamics because only a lepton and its antiparticle are present.[1–3] Furthermore, the study of positronium provides the principal test of the Bethe-Salpeter equation,[4,5] which describes the bound-state quantum-electrodynamical two-body system.

The energy separation in the ground $n=1$ state between orthopositronium ($^3S_1$ state) and parapositronium ($^1S_0$ state), which may be called the ground-state fine-structure interval, is the important quantity which has been determined for positronium. Two measurements of the fine-structure interval $\Delta\nu$ have been reported previously.[6,7] Both are in agreement with the theoretical value[8] and provide a confirmation of the theory to order α^3 Ry. The present experiment was undertaken to improve the accuracy of the measurement to provide a test of the α^4-Ry term.

Positronium annihilates into real γ rays because it is the particle-antiparticle system. For the first time a direct measurement of the two-γ-ray annihilation rate of ground-state parapositronium, λ_p, has been made in this experiment. The accuracy of this measurement provides a test of the theory of the annihilation to the order of $\alpha^6 mc^2/\hbar$.

The method of our experiment is basically similar to that of the earlier experiments and involves the measurement of an induced Zeeman transition between magnetic substates of ground-state positronium. Substantial improvement over the accuracy of the earlier measurements of $\Delta\nu$ was attained principally through the use of improved instrumentation. It was found necessary to use detection of coincident two-γ-ray annihilation rather than detection of the γ-ray energy spectrum, which was principally used in the earlier measurements. The natural linewidth of the Zeeman transition yields a value for λ_p.

Brief reports of this research have been published.[9,10]

II. THEORY OF EXPERIMENT

A. Energy Eigenvalues and Eigenstates

The theoretical value for $\Delta\nu$ is given by[8]:

$$\Delta\nu=\Delta W/h$$
$$=\tfrac{1}{2}\alpha^2 c\ \text{Ry}\left[\tfrac{7}{3}-(\alpha/\pi)(\tfrac{32}{9}+2\ln 2)-2\alpha^2\ln\alpha\right]. \quad (1a)$$

Using the following modern values[11] of the fine-structure constant α, the velocity of light c, and the Rydberg constant Ry

$$\alpha^{-1}=137.036\,02\pm 0.000\,21\ ,$$
$$c=2.997\,925\pm 0.000\,001\times 10^{10}\ \text{cm/sec},$$
$$\text{Ry}=1.097\,373\,1\pm 0.000\,000\,1\times 10^5\ \text{cm}^{-1}\ ,$$

we obtain $\Delta\nu = 2.03427 \times 10^5$ Mc/sec . (1b)

The term of order α^4 Ry has not yet been calculated.

The Zeeman-energy eigenvalues are given by[7,12]

$$W_{J,M} = W_{1,\pm1} = W_1 ,$$
$$W_{\frac{1}{2}\pm\frac{1}{2},0} = \tfrac{1}{2}(W_1 + W_0) \pm \tfrac{1}{2}\Delta W(1+x^2)^{1/2}, \tag{2a}$$

in which $W_{1(0)}$ is the energy level of orthopositronium (parapositronium) for the magnetic field $H_0 = 0$; $\Delta W = W_1 - W_0$. Furthermore,

$$x = (\mu_0 g_- - \mu_0 g_+) H_0/\Delta W, \tag{2b}$$

where[13] $g_- = -g_+ = g = 2(1 + \frac{1}{2}\alpha/\pi - 0.328\,\alpha^2/\pi^2)$

$= 2(1.0011596)$,

and μ_0 = Bohr magneton. The Zeeman energy levels are shown in Fig. 1.

The energy difference between the $M = 0$ and ±1 sublevels of orthopositronium is the quantity measured; the corresponding frequency is

$$f_{01} = \tfrac{1}{2}\ \Delta\nu[(1+x^2)^{1/2} - 1]. \tag{3a}$$

For small x, f_{01} becomes

$$f_{01} = \tfrac{1}{4}\Delta\nu x^2 = g^2\mu_0^2 H_0^2/h^2\Delta\nu. \tag{3b}$$

If f_{01} and H_0 are measured, a value for $\Delta\nu$ can be determined.

The spin eigenstates $\Psi_{J,M}$ in a magnetic field are

$$\Psi_{1,1} = \chi_{1,1} ,$$
$$\Psi_{1,-1} = \chi_{1,-1} ,$$
$$\Psi_{1,0} = (x/\{\sqrt{2}\,[1+x^2 - (1+x^2)^{1/2}]^{1/2}\}) \times\{\chi_{1,0} + [(1+x^2)^{1/2} - 1]x^{-1}\chi_{0,0}\} , \tag{4a}$$
$$\Psi_{0,0} = (x/\{\sqrt{2}\,[1+x^2 + (1+x^2)^{1/2}]^{1/2}\}) \times\{\chi_{1,0} + [(1+x^2)^{1/2} + 1]x^{-1}\chi_{0,0}\} ,$$

where the $\chi_{J,M}$'s are the zero-field spin functions. For small x, the properly normalized wave functions are

$$\Psi_{1,0} = [2/(4+x^2)^{1/2}](\chi_{1,0} + \tfrac{1}{2}x\,\chi_{0,0}),$$
$$\Psi_{0,0} = [x/(4+x^2)^{1/2}][\chi_{1,0} + (2/x)\,\chi_{0,0}] . \tag{4b}$$

B. Annihilation

Parapositronium annihilates with two-photon emission, and its annihilation rate λ_p is calculated to be[14–16]

$$\lambda_p = \tfrac{1}{2}\alpha^5(mc^2/\hbar)\,[1 - (\alpha/\pi)(5 - \tfrac{1}{4}\pi^2)]$$
$$= (0.803 - 0.005)\times10^{10}\ \mathrm{sec}^{-1}$$
$$= 0.798\times10^{10}\ \mathrm{sec}^{-1} , \tag{5}$$

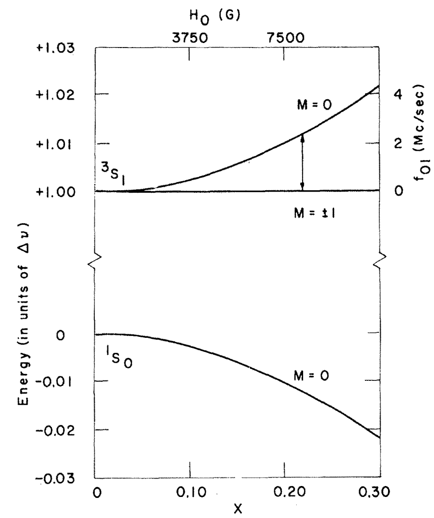

FIG. 1. Zeeman energy levels of positronium in its ground $n=1$ state. $\Delta\nu$ is the fine-structure separation between the $^3S_1$ and $^1S_0$ states of positronium at zero static magnetic field. (See Sec II A for definition of quantities.)

in which the correction term of order $\alpha^6 mc^2/\hbar$ accounts for the first-order radiative correction and the Coulomb binding effect. Orthopositronium annihilates with the emission of three photons, and the calculated[17] annihilation rate λ_0 is

$$\lambda_0 = \tfrac{1}{2}\alpha^6(mc^2/\hbar)(4/9\pi)(\pi^2 - 9) = 0.721\times10^7\ \mathrm{sec}^{-1} . \tag{6}$$

In the presence of a magnetic field H_0 the annihilation rates of the substates of positronium become

$$\lambda_{1,1} = \lambda_0 ,$$
$$\lambda_{1,-1} = \lambda_0,$$
$$\lambda_{1,0} = \left(1 - \frac{2+x^2 - 2(1+x^2)^{1/2}}{2\,[1+x^2 - (1+x^2)^{1/2}]}\right)\lambda_0 + \left(\frac{2+x^2 - 2(1+x^2)^{1/2}}{2\,[1+x^2 - (1+x^2)^{1/2}]}\right)\lambda_p = \lambda_{10,3} + \lambda_{10,2} ,$$
$$\lambda_{0,0} = \left(1 - \frac{2+x^2 + 2(1+x^2)^{1/2}}{2\,[1+x^2 + (1+x^2)^{1/2}]}\right)\lambda_0 + \left(\frac{2+x^2 + 2(1+x^2)^{1/2}}{2\,[1+x^2 + (1+x^2)^{1/2}]}\right)\lambda_p , \tag{7a}$$

or for small x

$$\lambda_{1,0} = \left(1 - \frac{x^2}{4-x^2}\right)\lambda_0 + \left(\frac{x^2}{4-x^2}\right)\lambda_p = \lambda_{10,3} + \lambda_{10,2} ,$$
$$\lambda_{0,0} = \left(1 - \frac{4}{(4-x^2)}\right)\lambda_0 + \left(\frac{4}{(4-x^2)}\right)\lambda_p . \tag{7b}$$

The annihilation rate $\lambda_{1,0}$ is of particular interest. For the magnetic field of 7900 G used, $x=0.22$ and $\lambda_{1,0} \gg \lambda_0$. Furthermore, the predominant mode of decay of $\Psi_{1,0}$ is then two-quantum annihilation. The branching ratio for the two-quantum decay rate of the state $\Psi_{1,0}$ to the three-quantum decay rate is

$$B_{1,0}=\lambda_{10,2}/\lambda_{10,3} \simeq \tfrac{1}{4}x^2\lambda_p/\lambda_0\ , \quad \text{for small } x$$
$$=13.5, \quad \text{for } x=0.22\ . \tag{8}$$

C. Theory of the Line Shape

Our experiment consists in the observation of the fraction of positronium atoms which decay by two-quantum annihilation as a function of the static magnetic field for a fixed value of the microwave frequency. From Sec. II B we see that in the magnetic field used, both $M=0$ states annihilate predominantly with two-quantum emission while the $M=\pm 1$ states annihilate with three-quantum emission. Hence, if a transition between $\Psi_{1,1}$ or $\Psi_{1,-1}$ and $\Psi_{1,0}$ is induced, the number of two-quantum annihilations will increase and the number of three-quantum annihilations will decrease.

The theory of the resonance line shape of positronium has been previously considered.[7,18] We have extended the treatment to include the three-level problem.

Consider the Hamiltonian

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}'(t)\ , \tag{9a}$$

where $\mathcal{H}_0$ is the time-independent part of the Hamiltonian, including the interaction with the static magnetic field H_0 in the z direction, which gives rise to the eigenvalues and eigenstates given in Eqs. (2) and (4). $\mathcal{H}'(t)$ is the interaction with the microwave field $\vec{H}_1 \cos\omega t$, which is linearly polarized in the x direction, and is equal to

$$\mathcal{H}'(t)=\mu_0 g_- \vec{s}^- \cdot \vec{H}_1 \cos\omega t + \mu_0 g_+ \vec{s}^+ \cdot \vec{H}_1 \cos\omega t$$
$$=\tfrac{1}{2}\mu_0 g H_1 \cos\omega t\,(\sigma_x^- - \sigma_x^+)\ , \tag{9b}$$

in which $s_x^{\pm}=\frac{1}{2}\sigma_x^{\pm}$, where σ_x is the Pauli spin matrix. $\mathcal{H}'(t)$ has the nonvanishing matrix elements

$$(1,1|\mathcal{H}'|1,0)$$
$$=[\Psi_{1,1}|\tfrac{1}{2}\mu_0 g H_1 \cos\omega t\,(\sigma_x^- - \sigma_x^+)|\Psi_{1,0}]$$
$$=(1,-1|\mathcal{H}'|1,0)$$
$$=\frac{(1+x^2)^{1/2}-1}{4[1+x^2-(1+x^2)^{1/2}]^{1/2}}\ \mu_0 g H_1\,(e^{+i\omega t}+e^{-i\omega t})$$
$$=V(e^{i\omega t}+e^{-i\omega t})\ , \tag{10}$$

where V is the time-independent part of the matrix element.

Neglecting all nonresonant terms,[12,19] the time-dependent equations for the state amplitudes of the three sublevels of orthopositronium become

$$i\hbar\dot{a}_1=a_0Ve^{+i\omega t}e^{i(W_1-W_{1,0})t/\hbar}-i\hbar(\tfrac{1}{2}\lambda_0)a_1\ ,$$
$$i\hbar\dot{a}_{-1}=a_0Ve^{+i\omega t}e^{i(W_1-W_{1,0})t/\hbar}-i\hbar(\tfrac{1}{2}\lambda_0)a_{-1}\ , \tag{11}$$
$$i\hbar\dot{a}_0=(a_1+a_{-1})V^*e^{-i\omega t}e^{i(W_{1,0}-W_1)t/\hbar}$$
$$-i\hbar(\tfrac{1}{2}\lambda_{1,0})a_0\ ,$$

where a_1, a_{-1}, and a_0 are the amplitudes of the $M=+1$, -1, and 0 states, respectively. The last terms in the equations are phenomenological terms accounting for annihilation. The principal effect of the neglected nonresonant terms is to produce a negligible shift in the energy levels.

The solution of Eq. (11) is

$$a_1+a_{-1}=A_1e^{(-\delta_1-\frac{1}{2}\lambda_0)t}+A_2e^{(-\delta_2-\frac{1}{2}\lambda_0)t}\ ,$$
$$a_0=(-\tfrac{1}{2}\hbar/iV) \tag{12a}$$
$$\times(-\delta_1A_1\exp[(-\delta_1-\tfrac{1}{2}\lambda_0)t+i(\omega_{01}-\omega)t]$$
$$-\delta_2A_2\exp[(-\delta_2-\tfrac{1}{2}\lambda_0)t+i(\omega_{01}-\omega)t])\ ,$$

where

$$\omega_{01}=(1/\hbar)\,(W_{1,0}-W_1)=2\pi f_{01}\ ,$$
$$\delta_{1,2}=\tfrac{1}{2}[i(\omega_{01}-\omega)+\tfrac{1}{2}(\lambda_{1,0}-\lambda_0)] \tag{12b}$$
$$\pm\tfrac{1}{2}\{[i(\omega_{01}-\omega)+\tfrac{1}{2}(\lambda_{1,0}-\lambda_0)]^2-8VV^*/\hbar^2\}^{1/2}\ , \tag{12c}$$

and the constants A_1 and A_2 are determined by the initial conditions.

If the positronium atom is in the state $\Psi_{1,1}$ at time $t=0$, then the initial conditions are

$$a_1=1, \qquad a_{-1}=0, \qquad a_0=0, \qquad \text{at } t=0. \tag{13}$$

$$\text{Hence,}\quad A_1+A_2=1, \qquad \delta_1A_1+\delta_2A_2=0,$$
$$\text{and}\quad A_1=\frac{\delta_2}{\delta_2-\delta_1}\ , \qquad A_2=-\frac{\delta_1}{\delta_2-\delta_1}\ . \tag{14}$$

The probability $P_{1,0}$ that the atom decays by two-quantum annihilation is given by

$$P_{1,0}=\int_0^\infty dt\,|a_0|^2\lambda_{10,2}\ . \tag{15a}$$

By integration

$$P_{1,0}=\lambda_{10,2}\frac{\hbar^2}{4|V|^2}\frac{|\delta_1|^2|\delta_2|^2}{|\delta_2-\delta_1|^2}\left(\frac{1}{\delta_1+\delta_1^*+\lambda_0}+\frac{1}{\delta_2+\delta_2^*+\lambda_0}-\frac{1}{\delta_1^*+\delta_2+\lambda_0}-\frac{1}{\delta_1+\delta_2^*+\lambda_0}\right)\ . \tag{15b}$$

Similarly, if the atom is initially in the state $\Psi_{1,-1}$, then the probability for two-quantum annihilation is

$$P_{-1,0}=P_{1,0}\ . \tag{16}$$

This equality might be expected since the $M=+1$ and -1 energy levels are degenerate.

Finally, if the atom is initially in the state $\Psi_{1,0}$, then

$$A_1 = \frac{1}{\delta_2 - \delta_1}\left(\frac{2V}{i\hbar}\right),$$
$$A_2 = -\frac{1}{\delta_2 - \delta_1}\left(\frac{2V}{i\hbar}\right). \qquad (17)$$

The probability $P_{0,0}$ for two-quantum annihilation is

$$P_{0,0} = \int_0^\infty dt\,|a_0|^2 \lambda_{10,2}$$
$$= \frac{\lambda_{10,2}}{|\delta_2 - \delta_1|^2} \times \left(\frac{|\delta_1|^2}{\delta_1 + \delta_1^* + \lambda_0} + \frac{|\delta_2|^2}{\delta_2 + \delta_2^* + \lambda_0} - \frac{\delta_1^*\delta_2}{\delta_1^* + \delta_2 + \lambda_0} - \frac{\delta_1\delta_2^*}{\delta_1 + \delta_2^* + \lambda_0}\right). \qquad (18)$$

The quantity to be compared with experiment is P_T, the change in the number of orthopositronium atoms which decay by two-quantum annihilation due to the microwave field divided by the number of orthopositronium atoms that decay by three-quantum annihilation:

$$P_T = \frac{N_1 P_{1,0} + N_{-1} P_{-1,0} + N_0(P_{0,0} - \lambda_{10,2}/\lambda_{1,0})}{N_1 + N_{-1} + (\lambda_{10,3}/\lambda_{1,0}) N_0}, \qquad (19)$$

where N_1, N_{-1}, and N_0 are the numbers of positronium atoms formed in the orthopositronium substates $M = 1, -1$, and 0, respectively, in the presence of the microwave field.

If we assume first that $\lambda_{10,3}/\lambda_{1,0} = 0$ and secondly that $N_1 = N_{-1} = N_0 = N$, then Eq. (19) simplifies to

$$P_T = \tfrac{1}{2}[P_{1,0} + P_{-1,0} + (P_{0,0} - \lambda_{10,2}/\lambda_{1,0})]. \qquad (20)$$

This is the line shape we will use to fit our data.

Some qualifying remarks should be given about the two simplifying assumptions made above. As for the first assumption, in a magnetic field of 8 kG, $\lambda_{10,3} \simeq 0.07\,\lambda_{1,0}$; hence Eq. (20) overestimates the magnitude of P_T by approximately 3.5%. There is also a magnetic field dependence in $\lambda_{10,3}/\lambda_{1,0}$ which will shift the line center, but this shift is negligible due to the small change in H_0 across the linewidth. The second assumption is true if unpolarized positrons in an unpolarized medium are used to produce positronium. However, in our experiment the positrons are obtained from a β-decay process and hence are polarized.[20] The state populations produced are summarized in Table I,[21] where $\epsilon = x/(1+x^2)^{1/2} \simeq x \simeq 0.2$ for 8 kG and p is the polarization of the positrons. Na^{22} produces positrons which are about 15% polarized,[22] so the population differences are of the order of 3%. Since the population differences only affect N_0 of Eq. (19), which already has relatively small coefficients (~ 7%), the change in magnitude of P_T due to the partial polarization of the positrons is of the order of 0.2%, either positive or negative, depending on the direction of the polarization relative to the magnetic field. The shift in the line center due to this effect is negligible, which can be seen as follows: The value of ϵ is proportional to H_0, which changes fractionally by $\Delta H_0/H_0 \simeq 3.7\times10^{-3}$ across the linewidth, and since the coefficients of N_0 are small, the shift in the line center relative to the width is negligible.

TABLE I. Relative populations of states for polarized positrons.

	Population, $H=0$	Population,[a] $H \neq 0$
$^1S_0$	N	$(1+p\epsilon)N$
$^3S_1$, $\lvert M\rvert = 1$	$2N$	$2N$
$^3S_1$, $M=0$	N	$(1-p\epsilon)N$

[a]Assuming e^+ polarized along $\vec{H}$.

1. *Line Shape in Limit of Small x and H_1*

It is useful to have an expression for the line shape in the limit of small x and small H_1. To lowest order in x, $|V|$ of Eq. (10) reduces to

$$|V| = (x/4\sqrt{2})\,\mu_0 g H_1, \qquad (21)$$

and P_T of Eq. (20) for small x and small H_1 (low microwave power) reduces to

$$P_T = \frac{x^2}{32\hbar^2}\,\frac{(\mu_0 g H_1)^2}{(f - f_{01})^2 + \frac{1}{4}\gamma^2}\,\frac{\lambda_{10,2}}{\lambda_0}, \qquad (22a)$$

where f is the microwave frequency $(=\omega/2\pi)$, and γ is the natural linewidth:

$$\gamma = \frac{1}{2\pi}\left[\left(2 - \frac{x^2}{(4-x^2)}\right)\lambda_0 + \frac{x^2}{(4-x^2)}\lambda_p\right], \qquad (22b)$$

where $\lambda_{10,2}$ is given by Eq. (7b).

In practice the applied microwave frequency f is held fixed and the magnetic field H_0 is varied through the resonance. An expression for the line shape in this case becomes, using Eqs. (22) and (3b),

$$P_T = AH_1^2/[(H_0 - H_{0r})^2 + \tfrac{1}{4}\Gamma^2], \qquad (23a)$$

where

$$A = \frac{x^2}{128\hbar^2}\,\frac{\mu_0^2 g^2 H_{0r}^2}{f_{01}^2}\,\frac{\lambda_{10,2}}{\lambda_0}, \qquad (23b)$$

$$\Gamma = \frac{H_{0r}\gamma}{2f_{01}} = \frac{H_{0r}}{4\pi f_{01}}\left[\left(2 - \frac{x^2}{4-x^2}\right)\lambda_0 + \frac{x^2}{4-x^2}\lambda_p\right], \qquad (23c)$$

$$H_{0r} = (\Delta\nu)^{1/2} h f_{01}^{1/2}/g\mu_0. \qquad (23d)$$

The resonance line in Eq. (23) is Lorentzian in shape and has a full width at half-maximum of

$$\Delta H_0 = \gamma H_{0r}/2f_{01} \qquad (24)$$

and a corresponding fractional width of

$$\frac{\Delta H_0}{H_{0r}} = \frac{\Gamma}{H_{0r}} = \frac{1}{4\pi f_{01}} \left[\left(2 - \frac{x^2}{4-x^2}\right)\lambda_0 + \left(\frac{x^2}{4-x^2}\right)\lambda_p\right]. \tag{25}$$

Power broadening of the natural resonance line will occur at sufficiently high microwave fields. The equation for the line shape can be reduced to the following form which gives the power-broadening effect:

$$P_T = AH_1^2/[(H_0 - H_{0r})^2 + B^2] , \tag{26a}$$

where $B^2 = \frac{1}{4}\Gamma^2 + aH_1^2$, (26b)

in which B is the power-broadened linewidth and a is a parameter to be determined by fitting the line shape to the data. This is a Lorentzian line that includes power broadening and can be used in a fitting program to determine H_{0r}, B, and AH_1^2. Slight deviations from this Lorentzian form arise from the inadequacy of the approximations that x and the fractional variation of H_0 over the resonance line are small.

Data on values of B versus signal height at resonance can be used to determine the natural linewidth Γ and hence λ_p. The value of the signal at resonance, S_0, is given from Eq. (26) as

$$S_0 = AH_1^2/B^2. \tag{27}$$

Use of Eqs. (27) and (26b) yields

$$1/B^2 = 4/\Gamma^2 - (4/aA\Gamma^2)S_0. \tag{28}$$

The intercept at $S_0 = 0$ of a straight-line fit to observed values of $1/B^2$ versus S_0 gives a value for Γ. Then λ_p can be determined from the following form of Eq. (23c):

$$\lambda_p = \left(\frac{4-x^2}{x^2}\right)\left[\left(\frac{4\pi f_{01}}{H_{0r}}\right)\Gamma - \left(2 - \frac{x^2}{4-x^2}\right)\lambda_0\right] . \tag{29}$$

2. *Line Shifts and Line Broadening due to Gas Collisions*

Collisions of positronium with the gas in which it is formed occur at a rate that is high compared to the annihilation rate and produce significant shifts and broadening of the resonance line. A shift in the measured positronium fine-structure interval is expected through collision mechanisms analogous to those involved in the well-known hfs pressure shifts[23] observed for hydrogen,[24] muonium,[25] and other atoms. Line broadening is expected principally due to the increase in the effective annihilation rate of orthopositronium caused by collisions. For positronium in argon the annihilation rate has been measured to be[26]

$$\lambda_{0A} = \lambda_0(1.0 + 0.0352\,\mathcal{P}), \tag{30}$$

where $\mathcal{P}$ is the argon pressure in atm.

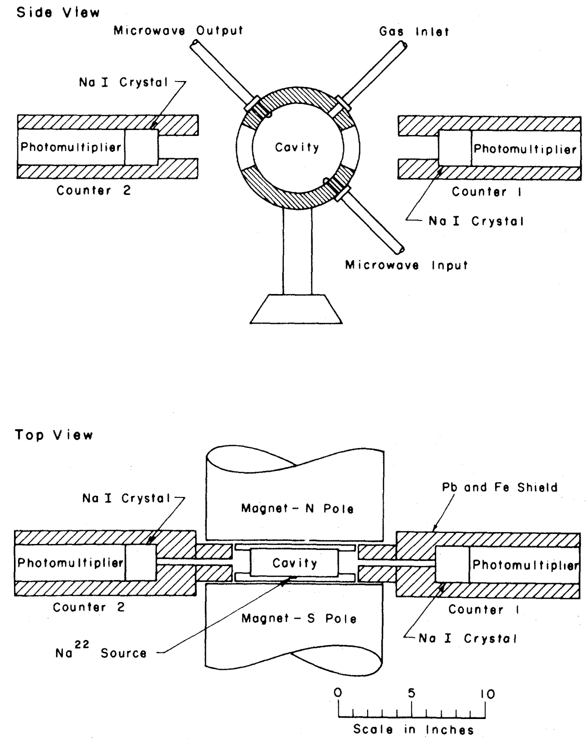

FIG. 2. Schematic diagram of experimental apparatus for positronium fine-structure experiment using γ-ray coincidence detection.

Various other collision effects and Doppler broadening appear to be negligible.[7]

III. APPARATUS

The frequency f_{01} of Eq. (3) between the $M = \pm 1$ and 0 Zeeman levels of orthopositronium is measured in a static magnetic field of 7900 G at which f_{01} is about 2383 Mc/sec. A resonance curve is taken with fixed microwave frequency by varying the magnetic field. Figure 2 is a schematic diagram of the experimental apparatus.

A. Radioactive Source

The source of the positrons was 10 mCi of Na^{22} in the form of NaCl, which was deposited in a brass cup 8 mm in diam and 3 mm high, sealed with a $\frac{1}{4}$-mil copper foil. This radioactive source was placed in a hole drilled along the axis of the microwave cavity whose dimensions were such that it was a waveguide beyond cutoff at the frequencies used, and held in place by a teflon ring. Thus the source was protected from heating by the microwaves and also did not itself disturb the Q of the cavity.

The positrons from Na^{22} have a maximum energy of 542 keV and a most probable energy of 120 keV.

These energetic positrons from the radioactive source were slowed down in argon to form positronium.[27,28]

B. Gas-Handling System

The cavity was connected to a pumping system consisting of a Leybold DO 121 oil diffusion pump and a mechanical pump. Customarily the whole system was evacuated to a pressure of about 3×10^{-6} Torr and pumped for several days. For the run the cavity was filled with gas (argon or carbon dioxide) to a pressure between 2 and 7 atm. The carbon dioxide used was Linde UP grade. Most of the data were teken in Linde UHP tank argon which had been analyzed and found to have the impurity contents oxygen (2.4 ppm), H_2O (0.2 ppm), and total hydrocarbons (< 1 ppm). For some of the lower-pressure runs this gas was passed through a barium-getter system which remained connected with the apparatus throughout the run. For most of the runs the cavity was merely sealed off after it had been filled.

C. Magnetic Field

Since we are measuring a Zeeman transition in order to determine $\Delta\nu$, the magnetic field must be determined accurately. The magnetic field was provided by a 12-in. Varian Model V4012A electromagnet with a gap of $2\frac{7}{8}$ in. The magnet was powered by a Varian Model V2100A power supply, which was current regulated and field stabilized by means of a Varian Model F-8A Nuclear Fluxmeter whose frequency was derived from a Gertsch Model FM-6 Frequency Meter. The Gertsch meter was operated in a mode which provided lock points every 10 kc/sec, derived from a crystal oscillator whose short-term stability was better than 1 part in 10^7 at 33.65 Mc/sec, which is the proton resonance frequency corresponding to about 7900 G. The NMR probe was H_2O and had a linewidth of about 0.2 G. By accurately centering a symmetrical pattern on the oscilloscope of the fluxmeter it is possible with the signal-to-noise ratio of 5 or greater, to stabilize to about $\frac{1}{20}$ of the linewidth or to 0.01 G, which corresponds to about 1 ppm at 7900 G.

The absolute value of the magnetic field was computed from the observed values of the proton NMR frequency using the proton gyromagnetic ratio for a proton in distilled water[29]:

$$\gamma_p/2\pi = (4.257\,70 \pm 0.000\,01) \times 10^3 \text{ cps/G}. \quad (31)$$

During a run the magnetic field value was monitored by measuring the proton resonance frequency with an electronic counter. The probe was located at a position within the cavity wall about 1 in. from the center of the cavity.

The homogeneity of the magnetic field in the volume of interest (a cylinder 1 in. in diam and 2 in. long at the center of the magnet) was about 10 ppm (300 cps out of 33.65 Mc/sec). It was determined by photographic studies with x-ray film that the positrons were confined by the magnetic field within a 1-in.-diam cylinder. A magnetic field plot for the center plane is shown in Fig. 3. The axial or z dependence showed the worst homogeneity and accounted for most of the variation. The homogeneity was found to be reproducible if the magnet was subjected to a set cycling procedure whenever the magnetic field was changed. The difference frequency between the probe position and the cavity center was < 10 ppm.

D. Microwave System

A block diagram of the microwave system is shown in Fig. 4. A very stable frequency source based on a crystal oscillator (Gertsch FM-6 or Hewlett Packard Model 5100A) provided a frequency of about 40 Mc/sec. This frequency was multiplied and amplified in power to produce an output signal of about 2400 Mc/sec and 500 W as input to the microwave cavity. The main amplification was provided by a Varian VA-802B klystron amplifier, which has a gain of 10^6 and a maximum output power of 1 kWcw. A small amount of power is coupled out of the cavity, rectified by a crystal detector, and used to control the amplitude of the input to the klystron. The power output of the klystron could

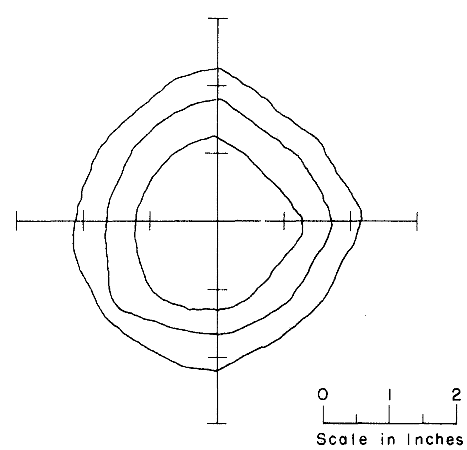

FIG. 3. Plot of magnetic field in center plane of magnet. Magnetic field at the center of the magnet was stabilized at the field value corresponding to a proton resonance frequency of 33.650 000 Mc/sec. Curves are drawn for constant magnetic field, and the field interval between adjacent curves is approximately 300 cps in units of the proton resonance frequency. At the center along the z direction the field changes by about 300 cps for a 1-in. change of position.

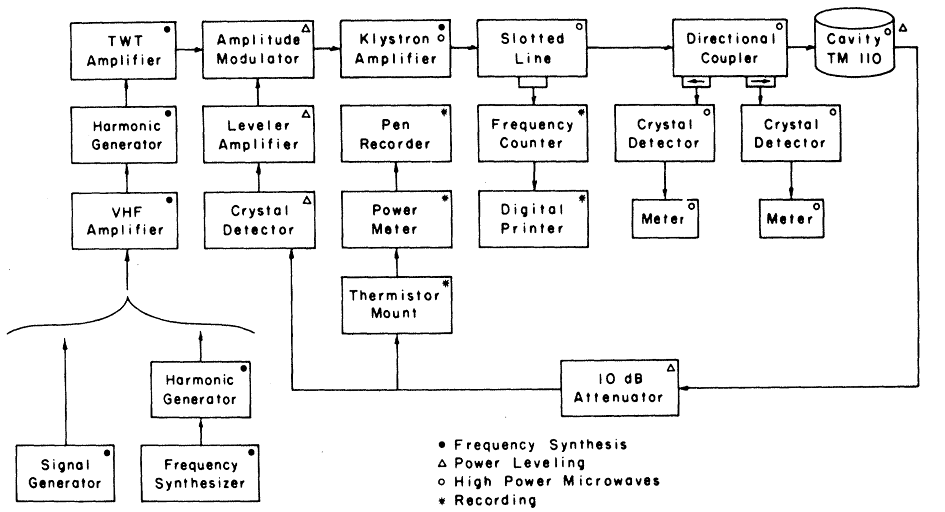

FIG. 4. Block diagram of microwave system.

be varied from 40 to 1000 W and maintained constant to within 1 or 2%. The frequency stability of the system was the same as that determined by the stable frequency of the source: 5 parts in 10^7 for the Gertsch signal generator, 1 part in 10^9 for the Hewlett Packard frequency synthesizer. Studies with a spectrum analyzer showed that the only spurious microwave signals occurring in the cavity bandpass were 300 kc/sec away from the main peak and 30 dB down in power.

The microwave cavity was a circular cylinder constructed of oxygen-free copper (OFHC) and silver plated. Its internal dimensions were 1.50 in. in length and 3.025 in. in radius. Its side walls were about 1 in. thick except where four windows 100 mils thick were cut to allow for passage of the γ rays with minimum degradation. Both end walls were removable cover plates, $\frac{5}{8}$ in. thick, with holes for the NMR probe. The cavity was water cooled. It was mounted in the magnet gap with its axis in the direction of the static magnetic field. It was operated in the TM_{110} mode; the radial dimension given corresponds to a resonant frequency of 2384 Mc/sec. This mode has the advantages of providing at the center of the cavity a strong microwave magnetic field transverse to the static magnetic field and a field configuration independent of the z (axial) coordinate. Throughout a region of $\frac{1}{2}$-in. radius from the axis of the cavity, the microwave magnetic field is a linearly polarized field with an amplitude constant to within about $2\frac{1}{2}\%$. The input loop to the cavity was designed empirically so that at resonance the cavity presented a matched load to the input lines, with a VSWR of <1.2. The measured Q_L of the cavity was 9000. A second cavity similar to the first was used in some of the early work. It was made of brass and was silver plated. Figure 4 indicates how the power level in the cavity, the microwave frequency, and the VSWR on the input line to the cavity were constantly monitored and recorded.

E. γ-Ray Detectors

Annihilation γ rays were detected with a 2-in. $\times$2-in. NaI crystal coupled to a RCA 6342A photomultiplier tube shielded from the magnetic fields and scattered γ rays by an arrangement of concentric cylinders of soft iron, lead, and netic-conetic materials (see Fig. 2). Close to the cavity there were two movable blocks of lead which provided a collimating slit, so that the γ rays from annihilations in the walls of the cavity could not be observed directly. The signals from the photomultiplier were fed through a preamplifier of conventional cathode-follower design and were analyzed according to the requirements of our two detection schemes.

IV. γ-RAY-COINCIDENCE EXPERIMENT

A. Method of Detection

The method of detection of the resonance line which proved most useful was based on the observation of the two-quantum annihilation of positronium, which yields two coincident γ rays 180° apart in direction, each with an energy of 0.511 MeV. By counting only these γ rays, the contribution of scattered background radiation from the cavity walls and detector slits was negligible.

The physical arrangement of the counters is shown in Fig. 2. The counting logic used was such that counter 2 opened a gate whenever a γ ray of energy greater than 0.400 MeV was detected. The energy of 0.4 MeV corresponded to the energy at the minimum of the Compton valley in the NaI crystal. If counter 1 also detected a γ ray in coincidence with counter 2 within a resolution time of 0.5 μsec, its output was fed through a linear gate into a 200-channel multichannel analyzer where it was stored. For analysis, only those pulses from counter 1 with an energy within the photopeak of a 0.511-MeV γ ray were used. Thus the requirement for a valid count was that two 0.511-MeV γ rays be detected in coincidence. The typical number of valid counts was 150 000 for a 10-min live time of the analyzer. The number of valid counts with the cavity evacuated was < 600 in a 10-min period, and hence this scattered background radiation produced < 0.4% of the total signal.

Equations can be written for the valid counts in the 0.511-MeV photopeak for the three following cases: (0) with a small admixture of nitric oxide in the argon gas; (1) with the microwave frequency off the resonance line; and (2) with the microwave frequency on the resonance line:

$$P^0 = N_p g_p + N_a g_p + N_o g_p ,$$
$$P^1 = N_p g_p + N_a g_p + N_o g_o , \tag{32}$$
$$P^2 = N_p g_p + N_a g_p + N_o(1 - P_T) g_o + N_o P_T g_p .$$

The notation and assumptions for these equations are the same as those in Ref. 7. The NO gas rapidly converts orthopositronium to parapositronium by spin-exchange collisions[27] so that only two-quantum annihilations of positronium occur. The quantities N_a, N_p, and N_o are the number of free positron-electron two-quantum annihilations, the number of positronium two-quantum annihilations, and the number of positronium three-quantum annihilations, respectively. A strong static magnetic field is assumed to be present for all three cases. Therefore, the number of three-quantum annihilations, N_0, arises principally from the $M = \pm 1$ substates of orthopositronium but slightly also from the $M = 0$ substate of orthopositronium. Most of the annihilations of the $M = 0$ substate of orthopositronium are two-quantum annihilations N_P. The quantity $N_o P_T$ is the number of three-quantum annihilations that are converted to two-quantum annihilations by the microwave field on resonance. The theoretical value for P_T is given in Eq. (20). The factors g_p and g_o are the efficiency factors for the detection of two-quantum and three-quantum annihilations, respectively, as valid counts as defined above. The solution for P_T is

$$P_T = (P^2 - P^1)/(P^0 - P^1). \tag{33}$$

A microwave-induced transition of an orthopositronium atom from the $M = \pm 1$ state to the $M = 0$ state is indicated by a positive value for the quantity $P^2 - P^1$.

B. Experimental Procedure

The basic experimental data are values of P^0, P^1, and P^2 as a function of magnetic field H_0 with fixed microwave frequency and power. Since the microwave cavity is necessarily a high-Q cavity and since the accurate measurement of microwave power over a frequency range is difficult, the resonance line is obtained by varying the magnetic field rather than the microwave frequency. Typically, the value of H_0 used for the off-resonance case P^1 was 200 G from the resonant value H_{0r}. The magnetic field was changed stepwise through the resonance line. For each change of the magnetic field value a cycling procedure was used to ensure the best homogeneity of the magnetic field. The line was swept back and forth many times in this manner.

Typical changes in the resonant frequency were of the order of 10^4 cps/day or 3 parts in 10^6 per day and were slowly varying. These changes, chiefly due to the change in temperature of the cavity cooling water, can be compensated for in the analysis of the data and do not constitute a significant source of error for $\Delta\nu$.

A data point consisted of counting for 10 min of live time on the analyzer, which gave about 150 000 counts. The difference in the counts for the on-resonance and off-resonance cases, $P^2 - P^1$, was typically 7500 counts, or a 5% signal. The P^0 counts were obtained at the end of a run with an admixture of about 5% NO in the argon. The data-acquisition time for a run varied from 7 to 21 days.

Data were obtained at different gas pressures. Arcing of the microwaves at the input to the cavity limited the lowest pressure at which observations could be made.

C. Analysis of Data

The raw data were in the form of γ-ray energy spectra from the multichannel analyzer. These spectra were analyzed with the Yale IBM 7090-94 computer which fitted the 0.511-MeV γ-ray photopeak with a least-squares fit of Tschebyscheff polynomials and found the center of the peak. Tschebyscheff polynomials are convenient in our case where equally spaced and exact values of the independent variable are given. Then the counts for a given condition are obtained by integrating over a region 25 channels wide about the center. An attempt was made during the data taking to hold the 0.511-MeV peak in a fixed channel of the analyzer so that these 25 channels would always correspond to the same energy region. Drifts of five channels out of 150 are not uncommon, so the energy/channel value may change by as much as 3%. The fact that the counts per channel near the limits of the integration are only 10% of the maximum counts per channel implies that slight changes in the value of energy/channel are relatively unimportant.

A serious problem is the change in counting efficiency with time caused by temperature variations. The number of peak counts for the off-resonance data points taken before and after sweeping the resonance line changed as much as 2% over the approximately 4-h period involved. These changes shift the line center and change the height of the signal. To account for this effect the data points were corrected for an assumed linear drift over the time period it took to sweep through the resonance line. One of the off-resonance magnetic field points was chosen and the values of P^1 at that point were compared for the times immediately before sweeping through the resonance and immediately afterward. Any changes in the value of P^1 were assumed to be of the form $P^1_{\text{after}} = P^1_{\text{before}}(1 + bt)$. The coefficient b was determined and all intervening data points were corrected for this time variation. Both positive and negative values of b were observed. Referring to Eq. (33), it can be seen that this correction will minimize shifts in the line center by correcting P^1 and P^2. However, P^0 must be corrected from data taken at a much later time and the efficiency correction is thus not well known. Since P^0 is independent of magnetic field, the uncertainty in the efficiency correction can change the signal size, but should not affect the determination of the line center. Data corrected in this manner showed smaller standard deviations for the values of P_T than uncorrected data.

After the data were corrected as just indicated, the values of P_T were computed from Eq. (33). For this calculation P^1 is obtained from observations taken when the magnetic field was far off resonance. Since P^1 depends on H_0 because of the magnetic quenching of the $M=0$ state of orthopositronium[30] a value for p^1 at a particular field H_0 is obtained with the assumption that P^1 varies linearly with H_0 over the small region of H_0 associated with the resonance line.

After all of the P_T values for a given run are computed, they are sorted according to magnetic field values and then subjected to a statistical criterion which checks the goodness of each data point against the average and standard deviation of the whole ensemble of data points for a particular value of H_0. Chauvenet's criterion[31] was used to reject bad data points. This criterion is that any reading of an ensemble of n readings is to be rejected when the magnitude of its deviation from the mean of the ensemble is such that the probability of occurrence of all deviations that large or larger does not exceed $1/2n$. Successive applications of Chauvenet's criterion are made until no values lie outside of the limits set by the criterion. For large groups of data points usually not more than one or two points are rejected, and the calculated standard deviations of the mean values of P_T are very close to those expected from the counting statistics alone.

The Chauvenet-criterion program converts an experimental run of from 300 to 1500 data points into 5 to 7 values of P_T corresponding to different values of magnetic field. These values of P_T with their associated standard deviations form the input for the program which actually fits a line shape to the observed data.

Two line-shape programs were used in analyzing the data: first, the simple line shape of Eq. (26) which involves the three parameters H_{0r}, B, and AH_1^2; secondly, the more complete line shape of Eq. (20) with the parameters $\Delta\nu$, H_1, and a scale factor. The primary reason for the scale factor is the neglect of the term $(\lambda_{10,3}/\lambda_{1,0})$ in the denominator of Eq. (19) in order to obtain Eq. (20). This neglect implies that Eq. (20) overestimates the magnitude of P_T by approximately 3.5%. Also neglected in Eq. (20) were variations in H_1 over the active region, inhomogeneities in H_0, and the effects of positron polarization. The fitted values were obtained by a least-squares procedure which minimized the value of χ^2:

$$\chi^2 = \sum_{\alpha=1}^{N} (P^{\alpha}_{T_{\text{exp}}} - P^{\alpha}_{T_{\text{theor}}})^2/\sigma^2_{\alpha}, \tag{34}$$

where σ_α is the standard deviation of the α data point, $P^{\alpha}_{T_{\text{expt}}}$ is the experimental value, and $P^{\alpha}_{T_{\text{theor}}}$ is the theoretical value.

The solution for the confidence interval for $\Delta\nu$ was carried out by means of the pivotal method.[32] The frequency distribution of the quantity X, defined by

$$X(\Delta\nu_0) = \sum_{\alpha=1}^{N} \frac{P^{\alpha}_{T_{\text{expt}}} - P^{\alpha}_{T_{\text{theor}}}(\Delta\nu)}{\sigma^2_\alpha} \frac{\partial P^{\alpha}_{T_{\text{theor}}}(\Delta\nu_0)}{\partial\Delta\nu_0} \times\left[\sum_{\alpha=1}^{N} \frac{1}{\sigma^2_\alpha}\left(\frac{\partial P^{\alpha}_{T_{\text{theor}}}(\Delta\nu_0)}{\partial\Delta\nu_0}\right)^2\right]^{-1/2}, \tag{35}$$

is unit normal with mean zero. The confidence intervals $\sigma_{\Delta\nu_0}$ for $\Delta\nu$ are determined by varying $\Delta\nu$ until $X=\pm1$, and at these values $\Delta\nu = \Delta\nu_0 \pm \sigma_{\Delta\nu_0}$. The effect of correlated errors in the other parameters for the determination of $\Delta\nu_0$ and $\sigma_{\Delta\nu_0}$ was found to be small.

D. Results

The results of the fits with the two different line shapes are given in Tables II and III. A typical fitted line shape is shown in Fig. 5. The experimental points are shown as dots, where the statistical counting errors are approximately the size

TABLE II. Results from complete theory [Eq. (20)] for the γ-ray-coincidence experiment.

Run	Power (W)	Pressure (lb/in.^{2} of Ar)	Source position	H_1 (G)	Scale factor	$\Delta\nu_0$ (10^5 Mc/sec)
1	565	66	S	22.38	0.957	2.033 39 ± 0.000 11
2	565	70	N	19.63	1.042	2.033 34 ± 0.000 10
3	565	75	N	20.42	0.997	2.033 25 ± 0.000 06
4	565	75	S	17.65	0.958	2.033 25 ± 0.000 09
5	113	43	S	9.04	1.020	2.033 53 ± 0.000 12
6	113	43	N	9.12	0.846	2.033 74 ± 0.000 14
7	113	41	N	7.26	0.956	2.033 66 ± 0.000 17
8	295	45	N	9.14	1.039	2.033 55 ± 0.000 05

of the dots. The solid curve is a best fit of Eq. (26).

1. *Determination of* $\Delta\nu$

In order to investigate possible systematic errors data were taken for different values of gas pressure and microwave input power and also for different source positions, as indicated in the Tables. No dependence of $\Delta\nu$ on microwave power or on source position is observed. A plot of $\Delta\nu$ versus argon pressure is shown in Fig. 6, where the dependence of $\Delta\nu$ on pressure is evident. A straight line is fitted to the experimental points, and $\Delta\nu$ for free positronium is taken as the extrapolated value at zero pressure:

$$\Delta\nu = (2.034\,03 \pm 0.000\,12) \times 10^5 \text{ Mc/sec}, \qquad (36)$$

where the quoted total error of 60 ppm is one standard deviation and includes the statistical counting error of 56 ppm and the error due to magnetic field inhomogeneity of 20 ppm. The use of a straight-line fit assumes that three-body collisions are negligible.

If the data were not corrected for the drift in detector efficiency, as discussed in Sec. IV C, the value of $\Delta\nu$ would differ from that of Eq. (36) by about 10 ppm. All other known errors associated with the microwave frequency and power, and detector stability are believed to be relatively negligible.

The value for the fractional fine-structure pressure shift determined from the slope of the fitted straight line is

$$\frac{1}{\Delta\nu}\frac{\partial\Delta\nu}{\partial P}\Big|_{300^\circ\text{K}} = (-0.93 \pm 0.18) \times 10^{-7}/(\text{Torr of Ar}), \qquad (37)$$

where the error is a one-standard-deviation statistical error.

Use of the simple line-shape theory of Eq. (26) gives values of $\Delta\nu$ approximately 20 ppm higher than that of Eq. (36). This is due to the fact that the simple line shape is symmetric in H_0 about the resonant value and does not take account of the line asymmetry associated with the H_0 dependence of the eigenstates in a magnetic field [Eq. (4)].

2. *Determination of* λ_p

From the values of the linewidth B and signal height S_0 given in Table III, the natural linewidth Γ can be determined by a fit to Eq. (28). A value for λ_p can be calculated from Γ using Eq. (29) with λ_0 replaced by λ_{0A} of Eq. (30). The value of λ_p so determined is

$$\lambda_p = (0.833 \pm 0.030) \times 10^{10} \text{ sec}^{-1}, \qquad (38)$$

TABLE III. Results from simple theory [Eq. (26)] for the γ-ray-coincidence experiment.

Run	Power (W)	Pressure (lb/in.^{2}) of Ar)	Source position	B (kc/sec)	Signal S_0 $= AH_1^2/B^2$	$\Delta\nu_0$ (10^5 Mc/sec)
1	565	66	S	108.820	0.2527	2.033 42 ± 0.000 11
2	565	70	N	99.776	0.2498	2.033 37 ± 0.000 10
3	565	75	N	102.063	0.2444	2.033 28 ± 0.000 06
4	565	75	S	93.503	0.2092	2.033 28 ± 0.000 09
5	113	43	S	71.258	0.1074	2.033 55 ± 0.000 12
6	113	43	N	71.451	0.0899	2.033 76 ± 0.000 14
7	113	41	N	67.769	0.0719	2.033 68 ± 0.000 17
8	295	45	N	71.464	0.1108	2.033 57 ± 0.000 05

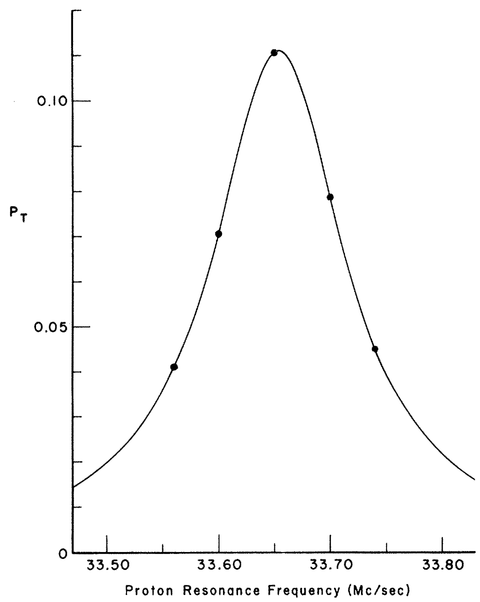

FIG. 5. A resonance line for the Zeeman transition in orthopositronium. Data were taken with argon pressure $=45$ lb/in.^{2}, microwave frequency $=2385.045$ Mc/sec, and microwave power $=100$ W. Observed points are represented by dots and the solid curve is a fitted theoretical curve.

where the error shown is one standard deviation.

3. *Scale Factor and* Q_L

The complete line-shape theory of Eq. (20) developed in Sec. II C has two parameters to determine the width and the height of the resonance line – the scale factor and the microwave magnetic field H_1.

The average value of the scale factor is 1.00 for all of the runs in Table II, in reasonable agreement with the expected value of 0.97. A slight difference was observed in the average values of scale factors obtained for the two different source positions: $f_S = 0.97 \pm 0.03$ and $f_N = 1.01 \pm 0.03$. This difference may not be significant in view of the statistical errors, but the difference has the correct sign and magnitude to be accounted for by the Na^{22} positron polarization.

For a given run the fitted value for H_1 can be used to calculate Q_L for the cavity for a TM_{110} mode:

$$Q_L = \frac{0.1622\ H_1^2(\mathrm{G}) \times f_{01}(\mathrm{Mc/sec}) \times \mathrm{Volume(cm^3)}}{20\ \mathrm{Power(W)}}\ . \tag{39}$$

The average value for the fitted Q_L for runs 1–6 was 10 000, in agreement with the value for the measured Q_L of 9000 (Sec. III D). Before runs 7 and 8, the microwave power arced badly, and the Q_L of the cavity dropped to about 5000, probably due to pitting of the silver plating.

V. γ-RAY-ENERGY EXPERIMENT

A. Method of Detection, Procedure, and Analysis

The first method of detection tried was based on the energy of the annihilation γ rays and was essentially the same as that used in earlier measurements[6,7] of $\Delta\nu$. However, we found an unexplained effect which made this method unsuitable in our experiment for a precision determination of $\Delta\nu$. Still, this method is suitable and has been used for the determination of λ_p.

The γ-ray-energy spectrum was observed with two independent NaI counters (see Fig. 7). The so-called peak region P of the spectrum extends from 0.475 to 0.550 MeV and the valley region V 0.220–0.445 MeV. The signal can be taken to be the number of counts in V, or in P, or the counts ratio V/P. The use of V/P has the advantage that it depends only on the shape of the spectrum and not on the absolute number of counts; hence small changes in detection efficiency are not important. The two counters were placed 60° apart in the cylindrical coordinate angle ϕ. The output pulses of the counters were fed into a TMC 400 channel

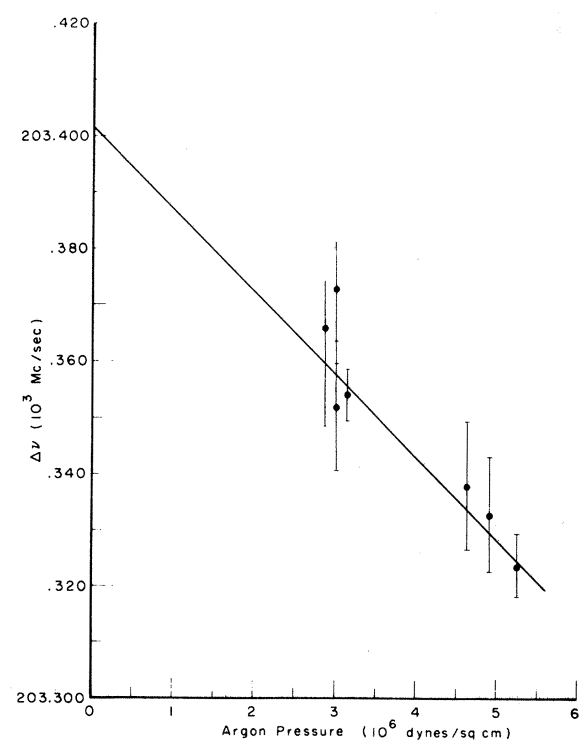

FIG. 6. Measured values of $\Delta\nu$ versus argon pressure using γ-ray-coincidence detection. Solid curve is a straight-line fit to the data.

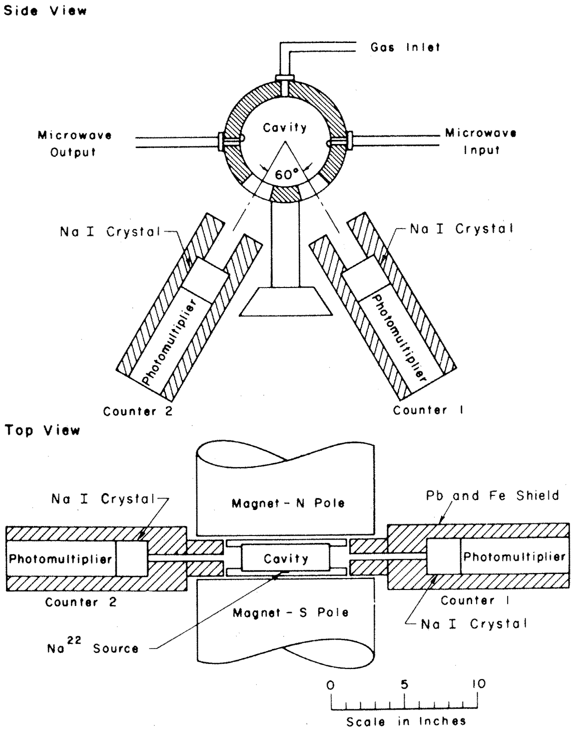

FIG. 7. Schematic diagram of experimental apparatus for positronium fine-structure experiment using γ-ray-energy detection.

analyzer and were analyzed independently in two different halves of the analyzer memory. This arrangement imposed an anticoincidence requirement on the detectors with a resolution time of 5 μsec and a dead time of 60 μsec.

A typical spectrum taken with 10 min of live time on the analyzer had about 2.5×10^6 counts in the total spectrum of which 8×10^5 were peak counts P and 1.2×10^6 were valley counts V. A typical resonance signal in V/P has an amplitude of about 15%. A large amount of scattered background radiation is present and amounts to about 50% of the total counts. If the scattered radiation is constant as the magnetic field H_0 is varied through the resonance line, the background will not distort the resonance line.

The experimental procedure was the same as that of the γ-ray-coincidence experiment. The analysis of data was also similar. The data were provided from the multichannel analyzer as γ-ray-energy spectra. The 0.511-MeV photopeak from two-quantum annihilation together with a marker peak from the 0.122-MeV γ ray of Fe^{57} were observed continuously and determined an absolute energy scale for the analyzer. A computer program fitted these peaks with Tschebyscheff polynomials and integrated the counts over the desired energy regions V and P. The quantities V, P and V/P are used in an equation similar to Eq. (32) to yield the quantity P_T similar to Eq. (33).[7]

B. Results

1. *Value of $\Delta\nu$ from the γ-Ray-Energy Experiment*

It was immediately evident from the results for the first run that the values of $\Delta\nu$ from counters 1 and 2 disagreed by some 20 standard deviations of the statistical counting error. This result was true for both the analyses with the line shapes of Eqs. (20) and (26). However, the heights and the widths of the resonance curves from counters 1 and 2 agreed.

A long search for the cause of the disagreement in $\Delta\nu$ values was undertaken. Table IV lists pos-

TABLE IV. Tests for cause of different $\Delta\nu$ values from two counters for γ-ray-energy experiment.

Test	Results on difference in $\Delta\nu$ values
1. Interchanged analyzer memory halves	no effect
2. Interchanged NaI crystal photomultiplier units	no effect
3. Rotated photomultiplier detectors by 180° inside their shields	no effect
4. Moved one detector to increase angle between detectors	no effect
5. Interchanged detector shields	no effect
6. Added iron baseplates to detector shields	no effect
7. Moved both detectors further from cavity	no effect
8. Turned cavity 180° about vertical axis	reversal of sign
9. Reversed magnetic field direction	no effect
10. Exchanged microwave input and output	no effect
11. Rotated microwave input 180° about longitudinal axis	no effect
12. Added brass over collimator slits	no effect
13. Rewrapped one counter in conetic	no effect
14. Narrowed collimator slits	no effect
15. Changed energy limits on V and P integrations	no effect
16. Placed source on opposite side of cavity	reversal of sign
17. Placed lead over magnet pole faces	no effect
18. Subtracted scattered radiation counts obtained with vacuum in cavity as a function of H_0	no effect
19. Removed anticoincidence requirement	no effect
20. Used Cu^{64} source	no effect
21. Used higher gas pressure	no effect
22. Used new cavity of OFHC copper	no effect
23. Used two Cu^{64} sources on opposite sides of cavity	agreement of $\Delta\nu$ values
24. Used CO_2 as gas	no effect

TABLE V. Values of $\Delta\nu$ for different counters and different source positions from V/P analysis for γ-ray-energy experiment.

Counter		$\Delta\nu$ (10^5 Mc/sec)
1	Source N pole	$2.033\,93 \pm 0.000\,03$
	Source S pole	$2.033\,33 \pm 0.000\,04$
	Average	$2.033\,63 \pm 0.000\,05$
2	Source N pole	$2.033\,17 \pm 0.000\,04$
	Source S pole	$2.034\,08 \pm 0.000\,04$
	Average	$2.033\,63 \pm 0.000\,06$

sible sources of systematic error which were investigated. On the basis of these tests, the only significant parameter appears to be source position.

Table V shows the values for $\Delta\nu$ obtained from the two counters for the two different source positions. We note that the average values of $\Delta\nu$ for the two counters agree.

The combined result from the two counters is

$$\Delta\nu = (2.033\,63 \pm 0.000\,04) \times 10^5 \text{ Mc/sec} , \qquad (40)$$

where a one-standard-deviation statistical error is indicated. This value of $\Delta\nu$ is obtained with an argon gas pressure of 30 lb/in.^{2} and a temperature of about 300 °K.

Table VI shows the results of the determination of $\Delta\nu$ from analysis of part of the data based on V counts, P counts, and the ratio V/P for the two counters and source positions. Again we note that the average values of $\Delta\nu$ from the two counters agree for the three methods of analysis. The values of $\Delta\nu$ based on the V analysis show much greater differences between the two source positions than the values of $\Delta\nu$ based on the P analysis, and furthermore, the P analysis shows variations of the opposite sign to that from the V or V/P analyses.

We do not understand the cause of a difference in $\Delta\nu$ values from the two counters or its dependence on source position. We believe it is probably associated with a dependence on magnetic field H_0 of the large amount of background-scattered radiation. It appears likely from the results shown in Tables V and VI that the average values of $\Delta\nu$ for the two different source positions give the correct value of $\Delta\nu$; hence the average values of $\Delta\nu$ for the two counters given in Eq. (40) are probably correct. However, because of the ambiguity involved, we do not use this result. It is interesting to note, however, that this value of $\Delta\nu$ from the γ-ray-energy experiment, when corrected for the pressure dependence of $\Delta\nu$ as given in Eq. (37), is in excellent agreement with the value of $\Delta\nu$ given in Eq. (36) from the γ-ray-coincidence experiment.

2. *Value of λ_p from the γ-Ray-Energy Experiment*

The data on the power-broadened linewidth B as a function of signal height at resonance S_0 from this γ-ray energy experiment shown in Fig. 8 have been used to determine λ_p as discussed in Sec. IIC 1. The data corresponding to the two counters and the two source positions were treated separately. Most of the data were taken with 2-atm argon. The results for the natural linewidth Γ for the case with the source on the N pole of the magnet are

$$\begin{aligned} &\text{counter 1: } \Gamma = [(3.73 \pm 0.10) \times 10^{-3}] H_{0r} , \\ &\text{counter 2: } \Gamma = [(3.68 \pm 0.07) \times 10^{-3}] H_{0r} . \end{aligned} \qquad (41)$$

The two results agree within the statistical error. The relatively small number of data with the source on the S pole of the magnet gives results in agreement with the above. Combining all these results and using Eq. (29) we find

$$\lambda_p = (0.793 \pm 0.012) \times 10^{10} \text{ sec}^{-1} , \qquad (42)$$

in which a one-standard-deviation statistical error is given.

VI. RESULTS AND DISCUSSION

Our measured value of $\Delta\nu$ is the value from Eq. (36),

$$\Delta\nu_{\text{expt}} = (2.034\,03 \pm 0.000\,12) \times 10^5 \text{ Mc/sec } (\pm 60 \text{ ppm}) \qquad (36)$$

TABLE VI. Values of $\Delta\nu$ for different counters and different source positions from analysis of V counts, P counts, and the ratio V/P for γ-ray-energy experiment.

		$\Delta\nu$ (10^5 Mc/sec)	
Counter 1	V/P analysis	V analysis	P analysis
Source N pole	$2.034\,09 \pm 0.000\,07$	$2.035\,21 \pm 0.000\,11$	$2.033\,15 \pm 0.000\,12$
Source S pole	$2.033\,27 \pm 0.000\,08$	$2.032\,15 \pm 0.000\,21$	$2.034\,28 \pm 0.000\,11$
Average	$2.033\,68 \pm 0.000\,10$	$2.033\,68 \pm 0.000\,24$	$2.033\,72 \pm 0.000\,17$
Source N pole	$2.033\,18 \pm 0.000\,10$	$2.031\,85 \pm 0.000\,08$	$2.034\,16 \pm 0.000\,10$
Source S pole	$2.034\,02 \pm 0.000\,08$	$2.035\,17 \pm 0.000\,14$	$2.033\,35 \pm 0.000\,19$
Average	$2.033\,53 \pm 0.000\,13$	$2.033\,51 \pm 0.000\,18$	$2.033\,76 \pm 0.000\,22$

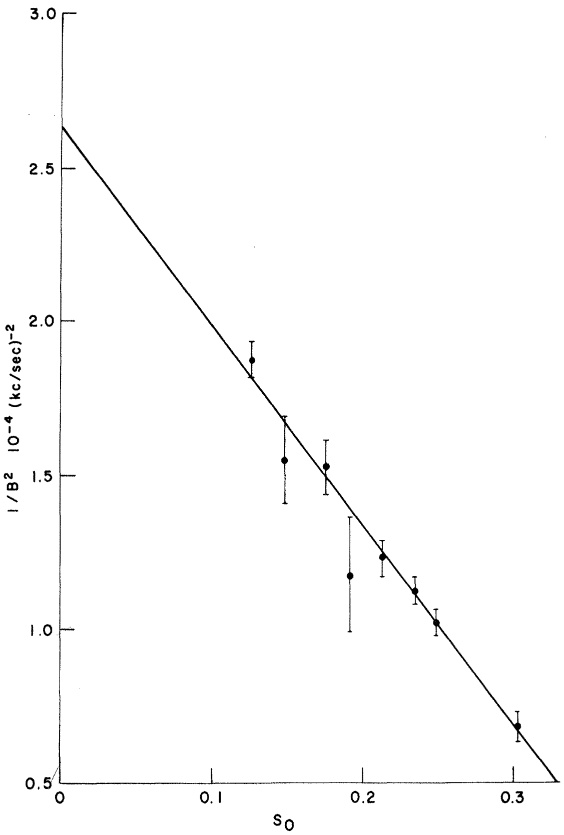

FIG. 8. Measured values of the inverse square of the linewidth $1/B^2$ versus signal height S_0. Solid curve is a straight-line fit to the data points.

in which a one-standard-deviation error is quoted. This value is based on the data taken in the γ-ray-coincidence experiment alone.

Our value for $\Delta\nu_{\text{expt}}$ is to be compared with the previous experimental values obtained by Deutsch, Brown, and Weinstein[6] of

$$\Delta\nu = (2.033\,80 \pm 0.000\,40)\times 10^5 \text{ Mc/sec} \quad ,$$

and by Hughes, Marder, and Wu[7] of

$$\Delta\nu = (2.033\,30 \pm 0.000\,40)\times 10^5 \text{ Mc/sec} \quad ,$$

in which the older values as given are in reasonable agreement with our present value. The agreement improves when our measured pressure shift is used to correct the older values. The older experiments were not sufficiently sensitive to detect the fine-structure pressure shift. Our experiment has improved the accuracy of our knowledge of $\Delta\nu_{\text{expt}}$ by a factor of 4.

The theoretical value is

$$\Delta\nu_{\text{theor}} = 2.034\,27\times 10^5 \text{ Mc/sec} \quad , \tag{1b}$$

as given in Eq. (1). The difference

$$\Delta\nu_{\text{expt}} - \Delta\nu_{\text{theor}} = (0.000\,24 \pm 0.000\,12)\times 10^5 \text{ Mc/sec} \tag{43}$$

is about two standard deviation of the experimental error. However, the uncalculated α^4-Ry term in $\Delta\nu_{\text{theor}}$ is of the order of this difference. Hence the agreement of $\Delta\nu_{\text{expt}}$ and $\Delta\nu_{\text{theor}}$ must be considered as reasonable, and a more sensitive comparison of $\Delta\nu_{\text{expt}}$ with $\Delta\nu_{\text{theor}}$ requires the calculation of the α^4-Ry term.

The result for the fractional fine-structure pressure shift for positronium in argon is

$$\frac{1}{\Delta\nu}\frac{\partial\Delta\nu}{\partial P}\bigg|_{300\text{ K}} = (-0.93 \pm 0.18)\times 10^{-7}/(\text{Torr of Ar}) \quad , \tag{37}$$

in which a one-standard-deviation error is given (see Sec. IV D 1). This is the first experiment in which a fine-structure pressure shift for positronium in a gas has been observed.

The value of the fractional pressure shift given in Eq. (37) has the same sign but is 20 times the magnitude of the fractional hfs pressure shift for hydrogen[24] or muonium[25] in argon. The Born-Oppenheimer approximation is not valid for the treatment of positronium collisions as it is for hydrogen collisions,[33] and hence a new theoretical treatment is required for the positronium fine-structure pressure shift. The polarizability is an important parameter in the theory of pressure shifts[23] and, since the polarizability of positronium is some eight times larger than that of hydrogen, it can be expected that the pressure shift for positronium will be much larger than that for hydrogen.

The measured values of λ_p are

$$\lambda_p = (0.793 \pm 0.012)\times 10^{10}\text{ sec}^{-1} \tag{42}$$

from the γ-ray-energy experiment (Sec. V B 2) and

$$\lambda_p = (0.833 \pm 0.030)\times 10^{10}\text{ sec}^{-1} \tag{38}$$

from the γ-ray-coincidence experiment (Sec. IV D 2). These are combined to give

$$\lambda_{p_{\text{expt}}} = (0.799 \pm 0.011)\times 10^{10}\text{ sec}^{-1} \quad , \tag{44}$$

in which a one-standard-deviation statistical error is quoted.

The previous experimental value for λ_p was obtained from a measured value for λ_p/λ_o[30]:

$$\lambda_p/\lambda_o = 1302 \pm 15\%$$

and for λ_o[34–36]:

$$\lambda_o = (0.7262 \pm 0.0015)\times 10^7\text{ sec}^{-1} \quad ,$$

which give

$$\lambda_{p_{\text{expt}}}(\text{old}) = (0.945 \pm 0.141)\times 10^{10}\text{ sec}^{-1} \quad .$$

Our present determination is about 13 times more accurate than the old value.

The theoretical value for λ_p is

$$\lambda_{p_{\mathrm{theor}}} = 0.798 \times 10^{10}\ \mathrm{sec}^{-1} \quad . \tag{5}$$

The difference between $\lambda_{p_{\mathrm{expt}}}$ and $\lambda_{p_{\mathrm{theor}}}$ is

$$\lambda_{p_{\mathrm{expt}}} - \lambda_{p_{\mathrm{theor}}} = 0.001 \pm 0.011 \ , \tag{45}$$

which represents excellent agreement within the experimental error. This result provides a good confirmation of the leading term in the theory of λ_p [see Eq. (5)], but is not sufficiently accurate to test the correction term of order $\alpha^6 mc^2\hbar$.

ACKNOWLEDGMENTS

We would like to acknowledge the participation of Dr. H. G. Robinson in the early stages of the experiment and the help of E. R. Carlson in taking the data.

---

*Research supported in part by the National Science Foundation.

†National Science Foundation Predoctoral Fellow.

‡Present address: National Accelerator Laboratory, Batavia, Ill.

§Present address: EG & G, Inc., Los Alamos, N. M.

‖Present address: Physics Department, University of Virginia, Charlottesville, Va.

[1]G. Källén, in *Encyclopedia of Physics*, edited by S. Flügge (Springer-Verlag, Berlin, 1958) Vol. V, No. 1.

[2]H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Academic, New York, 1957).

[3]*Atomic Physics*, edited by V.W. Hughes, B. Bederson, V. W. Cohen, and F. J. M. Pichanick (Plenum, New York, 1969).

[4]E. E. Salpeter and H. A. Bethe, Phys. Rev. 84, 1232 (1951).

[5]M. Gell-Mann and F. Low, Phys. Rev. 84, 350 (1951).

[6]M. Deutsch and S. C. Brown, Phys. Rev. 85, 1047 (1952); R. Weinstein, M. Deutsch and S. Brown, *ibid.* 94, 758 (1954); 98, 223 (1955).

[7]V. W. Hughes, S. Marder, and C. S. Wu, Phys. Rev. 106, 934 (1957).

[8]R. Karplus and A. Klein, Phys. Rev. 87, 848 (1952); T. Fulton, D. A. Owen, and W. W. Repko, Phys. Rev. Letters 24, 1035 (1970).

[9]E. D. Theriot, Jr., R. H. Beers, and V. W. Hughes, Bull. Am. Phys. Soc. 12, 74 (1967).

[10]E. D. Theriot, Jr., R. H. Beers, and V. W. Hughes, Phys. Rev. Letters 18, 767 (1967).

[11]B. N. Taylor, W. H. Parker, and D. N. Langenberg, Rev. Mod. Phys. 41, 375 (1969).

[12]P. Kusch and V. W. Hughes, in *Encyclopedia of Physics*, edited by S. Flügge (Springer-Verlag, Berlin, 1959), Vol. XXXVII, No. 1.

[13]C. M. Sommerfield, Ann. Phys. (N. Y.) 5, 26 (1958). The theoretical electron g value has recently been calculated to higher accuracy, but the higher accuracy is not needed for the present paper. See J. Aldins, T. Kinoshita, S. J. Brodsky, and A. J. Dufner, Phys. Rev. Letters 23, 441 (1969).

[14]P. A. M. Dirac, Proc. Cambridge Phil. Soc. 26, 361 (1930).

[15]I. Harris and L. M. Brown, Phys. Rev. 105, 1656 (1957).

[16]This equation corrects a misprint in Eq. (7) of Ref. 10.

[17]A. Ore and J. L. Powell, Phys. Rev. 75, 1696 (1949).

[18]O. Halpern, Phys. Rev. 94, 904 (1954).

[19]F. Bloch and A. Siegert, Phys. Rev. 57, 522 (1940).

[20]C. S. Wu, E. Ambler, R. W. Hayward, D. D. Hoppes, and R. P. Hudson, Phys. Rev. 105, 1413 (1957).

[21]L. Dick, L. Feuvrais, L. Madansky, and V. L. Telegdi, Phys. Letters 3, 326 (1963).

[22]L. A. Page and M. Heinberg, Phys. Rev. 106, 1220 (1957).

[23]G. A. Clarke, J. Chem. Phys. 36, 2211 (1962).

[24]F. M. Pipkin and R. H. Lambert, Phys. Rev. 127, 787 (1962).

[25]W. E. Cleland, J. M. Bailey, M. Eckhause, V. W. Hughes, R. M. Mobley, R. Prepost, and J. E. Rothberg, Phys. Rev. Letters 13, 202 (1964).

[26]F. F. Heymann, P. E. Osmon, J. J. Viet, and W. F. Williams, Proc. Phys. Soc. (London) 78, 1038 (1961).

[27]M. Deutsch, Progr. Nucl. Phys. 3, 131 (1953).

[28]V. W. Hughes, J. Appl. Phys. 28, 16 (1957).

[29]E. R. Cohen and J. W. M. DuMond, Rev. Mod. Phys. 37, 537 (1965).

[30]S. Marder, V. W. Hughes, and C. S. Wu, Phys. Rev. 98, 1840 (1955).

[31]A. G. Worthing and J. Geffner, *Treatment of Experimental Data* (Wiley, New York, 1943), p. 171.

[32]K. A. Brownlee, *Statistical Theory and Methodology in Science and Engineering* (Wiley, New York, 1965), p. 127.

[33]N. F. Mott and H. S. W. Massey, *Theory of Atomic Collisions* (Oxford U. P., London, England, 1965).

[34]R. H. Beers and V. W. Hughes, Bull. Am. Phys. Soc. 13, 633 (1968).

[35]V. W. Hughes, in *Atomic Physics*, edited by V. W. Hughes, B. Bederson, V. W. Cohen, and F. J. M. Pichanick (Plenum, New York, 1969), p. 33.

[36]B. G. Duff and F. F. Heymann, Proc. Roy. Soc. (London) A270, 517 (1962).