# Hyperfine Structure of Positronium in Its Ground State*†

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The energy difference between orthopositronium and parapositronium in the ground $n=1$ state, which is called the hyperfine structure separation $\Delta\nu$, has been determined in an experiment similar to that of Deutsch, Brown, and Weinstein. The frequency difference between the $M=\pm1$ and the $M=0$ Zeeman levels of orthopositronium was measured in a static magnetic field of 8000 gauss at which it is about 2460 Mc/sec. A resonance curve was taken with fixed frequency by varying the magnetic field, and an observed fractional line width, $\Delta H_0/H_{0r}$, of about $4.5\times10^{-3}$ was obtained. The principal part of the line width ($3.6\times10^{-3}$) is due to the annihilation lifetime of positronium; microwave power broadening and magnetic field inhomogeneity account for the remainder. No Doppler broadening is present despite the high velocity of the positronium atoms, because the mean free path of a positronium atom in the gas is small compared to the wavelength of the microwave radiation. The value of $\Delta\nu$ was computed from the observed values of the frequency and magnetic field at resonance by use of the Breit-Rabi formula to give the result: $\Delta\nu=(2.0333\pm0.0004)\times10^5$ Mc/sec. The error quoted is 5 times the statistical error and is believed to represent an upper limit to possible systematic errors. The value agrees with the values of Deutsch *et al.* of $(2.0338\pm0.0004)\times10^5$ Mc/sec and $(2.0335\pm0.0005)\times10^5$ Mc/sec, obtained with fields of 9500 and 9000 gauss, respectively, and hence confirms the use of the Breit-Rabi formula. The experimental values agree with the theoretical value for $\Delta\nu$ of 2.0337 $\times10^5$ Mc/sec computed to order $\alpha^3$ ry by Karplus and Klein.

## 1. INTRODUCTION

THE most critical tests of the validity of quantum electrodynamics to date, and, in particular, of the contributions of virtual radiative processes, have been measurements of the energy levels of a single electron in an external Coulomb field[1] (Lamb shift) and in an external magnetic field[2] (anomalous magnetic moment of the electron). At present there exists a small discrepancy of 0.65 Mc/sec between the observed value of the Lamb shift in hydrogen (1057.77±0.10 Mc/sec) and the theoretical value (1057.12±0.16 Mc/sec). A similar discrepancy exists for deuterium. For singly ionized helium the observed value of the Lamb shift (14 043±13 Mc/sec) is in agreement with the theoretical value (14 043±3 Mc/sec). The observed value of the spin magnetic moment of the electron, $g_s/g_l$, is derived from two measurements. One is the measurement of the electronic $g$ value in the ground state of hydrogen relative to the proton $g$ value, $g_J/g_p$, and the other is the measurement of the orbital $g$ value of the free electron relative to the proton $g$ value, $g_l/g_p$. The quantity $g_J/g_p$ has been measured to an accuracy of about 1 part per million (ppm), but the quantity $g_l/g_p$ has been measured to an accuracy of only 12 ppm. If the average of the two measured values of $g_l/g_p$ is used, the experimentally determined value of $g_s/g_l$ is 2(1.001156) with an error of about ±10 ppm. This value agrees with the theoretical value $g_s/g_l=2(1.0011454)$, but the experimental error is sufficiently large so that a convincing test of the fourth-order ($\alpha^2$) radiative correction is not provided.

Both for the Lamb shift and the anomalous magnetic moment experiments the electron is in a hydrogen atom or a hydrogen-like atom. For a pure test of quantum electrodynamics, of course, the presence of the proton in hydrogen introduces certain complications. Even though the field of the proton can be treated as a classical Coulomb field to a very good approximation, small but appreciable effects arise which require a more adequate theory for the proton.

Positronium, the bound state of an electron and a positron, is an ideal system for a test of quantum electrodynamics because no particles foreign to the theory are present. Furthermore, the bound-state two-body system requires a certain extension of the theory, which is described by the Bethe-Salpeter equation,[3] as compared to the single electron in a classical external field. The study of positronium provides the principal test of the Bethe-Salpeter equation. A two-body equation for the electron and the proton is used to calculate a relativistic reduced mass correction to the Lamb shift in hydrogen, but the correction is of the order of the present discrepancy between the theoretical and experimental values of the Lamb shift, and hence this application does not provide any confirmation for the two-body equation.

The energy separation in the ground $n=1$ state between orthopositronium ($^3S_1$ state) and parapositronium ($^1S_0$ state), which is called the hyperfine structure separation, is the important quantity which has been

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1 Triebwasser, Dayhoff, and Lamb, Phys. Rev. **89**, 98 (1953); Novick, Lipworth, and Yergin, Phys. Rev. **100**, 1153 (1955); E. E. Salpeter, Phys. Rev. **89**, 92 (1953); earlier references will be found in these three papers.

2 Koenig, Prodell, and Kusch, Phys. Rev. **88**, 191 (1952); R. Beringer and M. A. Heald, Phys. Rev. **95**, 1474 (1954); J. H. Gardner, Phys. Rev. **83**, 996 (1951); P. Franken and S. Liebes, Jr., Phys. Rev. **104**, 1197 (1956); R. Karplus and N. Kroll, Phys. Rev. **77**, 536 (1950).

3 E. E. Salpeter and H. A. Bethe, Phys. Rev. **84**, 1232 (1951); M. Gell-Mann and F. Low, Phys. Rev. **84**, 350 (1951).

determined for positronium. The theoretical value of the hyperfine structure splitting has been computed to order $\alpha^3$ ry ($\alpha$=fine structure constant) on the basis of the Bethe-Salpeter equation.[4] A measurement of the hyperfine structure splitting has been reported by Deutsch, Brown, and Weinstein,[5] which is in agreement with the theoretical value and provides a confirmation of the theory to order $\alpha^3$ ry. The experiment reported in the present paper was begun at about the same time as the original experiment of Deutsch and Brown. It was decided to complete our experiment despite the publication of a value by Deutsch *et al.*, because it was felt that an independent redetermination of this important quantity would be valuable. This paper contains a complete report of the hyperfine structure measurement, which has so far been discussed only in abstract and letter form.§

## 2. THEORY OF THE EXPERIMENT

### 2.1. Energy Levels

The energy separation between orthopositronium and parapositronium is of the order $\alpha^2$ ry. Part of the splitting is due to a magnetic spin-spin interaction term analogous to that which gives rise to the hyperfine structure splitting in the $s$ states of hydrogen. For positronium this term is of order $\alpha^2$ ry rather than of order $(m/M)\alpha^2$ ry ($m$=electron mass, $M$=proton mass) as in hydrogen, because the positron has a magnetic moment of 1 Bohr magneton whereas the proton has a magnetic moment of only 2.8 nuclear magnetons. A second interaction term is present in positronium. It is the virtual annihilation interaction characteristic of a particle-antiparticle system,[6] and it contributes about one half of the hyperfine structure splitting in positronium. The contribution to the positronium hyperfine structure splitting of order $\alpha^3$ ry arises from processes involving two photons and from the vacuum polarization corrections to one photon exchange and virtual annihilation. The theoretical value for the hyperfine structure splitting in the ground state of positronium has been computed to the order $\alpha^3$ ry[7,8] and is given by

$$\Delta W/h=\Delta\nu=\alpha^2(\mathrm{ry}_\infty/h)\left[\frac{7}{6}-\left(\frac{16}{9}+\ln 2\right)\frac{\alpha}{\pi}\right]$$

$$=2.0337\times10^5\ \mathrm{Mc/sec}. \quad (1)$$

[4] R. Karplus and A. Klein, Phys. Rev. **87**, 848 (1952).

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

§ A preliminary report of the experiment described in this paper has appeared. Marder, Hughes, and Wu, Bull. Am. Phys. Soc. Ser. II, **2**, 38 (1957).

[6] H. J. Bhabha, Proc. Roy. Soc. (London) **A154**, 195 (1936).

[7] See reference 4; references to earlier work in which the $\alpha^2$ ry term was calculated are given in this paper.

[8] S. DeBenedetti and H. C. Corben, *Annual Review of Nuclear Science* (Annual Reviews, Inc., Stanford, 1954), Vol. 4, p. 191.

A direct transition between the $^1S_0$ and $^3S_1$ states of positronium would occur at a wavelength of about 1.5 mm. Since positronium is short-lived and since the transition is a magnetic dipole transition, a microwave magnetic field of the order of a gauss would be required. The attainment of such a large field at a 1.5-mm wavelength is probably possible, but it would present a difficult problem and would probably require pulsed operation. A transition at a lower frequency at which adequate cw power is available and from which the hyperfine structure splitting $\Delta\nu$ can be determined is obtained by utilizing the Zeeman effect of positronium. The ratio of the natural radiative width to the resonant frequency is the same for the Zeeman transition as for the direct $\Delta\nu$ transition, and hence there is no fundamental advantage gained by studying the direct $\Delta\nu$ transition. (See Sec. 2.3.)

The Hamiltonian for positronium in a static magnetic field $H_0$ can be taken as

$$\mathcal{H}_s=\mathcal{H}_0+\mu_0 g_- \mathbf{s}_-\cdot\mathbf{H}_0+\mu_0 g_+ \mathbf{s}_+\cdot\mathbf{H}_0. \quad (2)$$

$\mathcal{H}_0$ is the Hamiltonian term which is independent of magnetic field and which leads to the zero field energy level scheme for positronium; $\mu_0$ is the Bohr magneton; $g_-$ and $g_+$ are the electron and positron gyromagnetic ratios, respectively, including radiative corrections[9]

$$g_-=-g_+=g=2(1+\alpha/2\pi-2.973\alpha^2/\pi^2)=2(1.0011454);$$

$\mathbf{s}_-$ and $\mathbf{s}_+$ are the electron and positron spin operators, respectively. Diagonalization of this Hamiltonian yields the Breit-Rabi formula for the energy levels.[8,10]

$$W_{F,M}=W_{\frac{1}{2}\pm\frac{1}{2},0}=\tfrac{1}{2}(W_1^{(0)}+W_0^{(0)})\pm\tfrac{1}{2}(\Delta W)(1+x^2)^{\frac{1}{2}},$$
$$W_{1,\pm1}=W_1^{(0)}. \quad (3)$$

$W_1^{(0)}$ and $W_0^{(0)}$ are the energy values at zero magnetic field of orthopositronium and parapositronium, respectively, $\Delta W=W_1^{(0)}-W_0^{(0)}$, and $x=2g\mu_0 H/\Delta W$. It is seen that the $M=\pm1$ states of orthopositronium are unperturbed by the magnetic field. Figure 1 shows the energy levels as a function of magnetic field. The energy difference between the $M=0$ and the $M=\pm1$ substates of orthopositronium is the quantity accessible to measurement, and the corresponding Bohr frequency is given by

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

$$f_{01}\simeq(\Delta\nu/4)x^2,\ \text{for small values of } x. \quad (4b)$$

For a magnetic field of 8000 gauss $x\simeq0.22$, and thus $f_{01}\simeq2400$ Mc/sec. Microwave magnetic fields that are of sufficient intensity to effect an observable transition are readily available at this frequency.

[9] R. Karplus and N. Kroll, Phys. Rev. **77**, 536 (1950).

[10] N. F. Ramsey, *Molecular Beams* (Oxford University Press, London, 1956), p. 83.

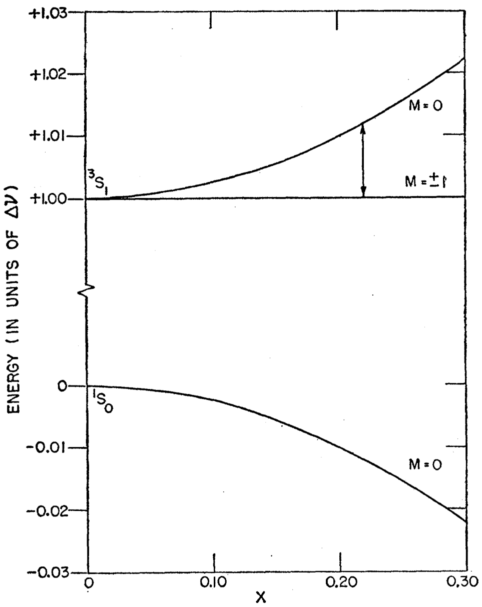


FIG. 1. Zeeman energy levels of positronium in its ground $n=1$ state. $\Delta\nu$ is the hyperfine structure separation between the $^3S_1$ and $^1S_0$ states of positronium at zero static magnetic field. The $M$ values designate the magnetic substates. $x=2g\mu_0H_0/(h\Delta\nu)$. (See Sec. 2.1 for definition of quantities.)

### 2.2. Decay Modes

The observation of a transition between the $M=\pm1$ and $M=0$ substates of orthopositronium depends on the different modes of annihilation of the states in a static magnetic field.[11] A pure singlet state annihilates with the emission of two gamma rays each having an energy of $mc^2$. A pure triplet state annihilates with the emission of three gamma rays whose energies sum to $2mc^2$ and whose energy distribution is continuous up to the maximum energy of $mc^2$.[12] In the presence of a magnetic field the $M=\pm1$ states of orthopositronium are pure triplet states and hence will decay by three quantum annihilation. On the other hand, the $M=0$ state of orthopositronium is perturbed by the magnetic field to include a mixture of singlet state. By the use of first order perturbation theory the wave function for the $M=0$ state of orthopositronium in a magnetic field is found to be

$$\psi_{1,0}=[1+(\tfrac{1}{2}x)^2]^{-\frac{1}{2}}(u_{1,0}+\tfrac{1}{2}xu_{0,0}), \qquad (5)$$

in which $u_{1,0}$ is the zero field eigenstate of $M=0$ orthopositronium and $u_{0,0}$ is the zero field eigenstate of parapositronium. The total annihilation rate of the state $\psi_{1,0}$ is given approximately for small $x$ by

$$\gamma_0=\gamma_t+(\tfrac{1}{2}x)^2\gamma_s, \qquad (6)$$

in which $\gamma_t(=7.21\times10^6\ \text{sec}^{-1})$ is the annihilation rate of a pure triplet state and $\gamma_s(=8.03\times10^9\ \text{sec}^{-1})$ is the annihilation rate of a pure singlet state. The branching ratio, $B$, of the number of two-quantum annihilations to the number of three-quantum annihilations is

$$B=(\tfrac{1}{2}x)^2(\gamma_s/\gamma_t). \qquad (7)$$

For a field of 8000 gauss Eqs. (6) and (7) give $\gamma_0$ and $B$ to an accuracy of about 1%, which is adequate for the applications to the present experiment. The branching ratio is 13.5 for $H_0=8000$ gauss, and hence 0.93 of the decays will be two quantum annihilations. Thus, if a positronium atom in the magnetic field undergoes a transition from either of the $M=\pm1$ states of orthopositronium to the $M=0$ state of orthopositronium, the decay will change from a three-quantum annihilation to a predominantly two-quantum annihilation. The reverse transition from the $M=0$ state to either of the $M=\pm1$ states is also observable, of course, but, as will be discussed in the following section, this transition is much less probable.

### 2.3. Natural Line Shape: Simple Theory

The 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. The principal factor determining the resonance line shape is the lifetime of the states due to the annihilation process. Other factors which must be considered are microwave power broadening, Doppler broadening, collision broadening, and broadening due to the inhomogeneity of the magnetic field.

The resonance line shape due to the annihilation lifetime of positronium will be discussed first. The derivation of the results to be stated is given in Appendix I. The Hamiltonian for positronium in a steady magnetic field $\mathbf{H}_0$ in the $z$ direction and a microwave magnetic field $\mathbf{H}_1\cos\omega t$ linearly polarized in the $y$ direction is

$$\mathcal{H}=\mathcal{H}_0+\mu_0g_-\mathbf{s}_-\cdot\mathbf{H}_0+\mu_0g_+\mathbf{s}_+\cdot\mathbf{H}_0+\mathcal{H}'(t), \qquad (8)$$

in which

$$\mathcal{H}'(t)=\mu_0g_-\mathbf{s}_-\cdot\mathbf{H}_1\cos\omega t+\mu_0g_+\mathbf{s}_+\cdot\mathbf{H}_1\cos\omega t. \qquad (8a)$$

For low microwave fields and near resonance, the decay rate of the $M=+1$ state of orthopositronium is given by

$$\gamma_1^{(\text{rf})}=\gamma_t+\frac{\gamma_0|V|^2}{\hbar^2[(\omega_{01}-\omega)^2+(\tfrac{1}{2}\gamma_0)^2]}=\gamma_t+\gamma', \qquad (9)$$

where $\omega_{01}=2\pi f_{01}$, and $V$ is the amplitude of the matrix element of $\mathcal{H}'(t)$ between the $M=+1$ and the $M=0$ states of orthopositronium. The matrix element $V$ is given by

$$V=+[ix/(4\sqrt{2})]\mu_0gH_1, \qquad (9a)$$

where terms of higher order in $x$ have been neglected. The expression for $\gamma'$ neglects terms of relative order

[11] Hughes, Marder, and Wu, Phys. Rev. 98, 1840 (1955); M. Deutsch, Progr. Nuclear Phys. 3, 131 (1953).

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

$\gamma_t/\gamma_0(\simeq 0.07)$. The decay rate, $\gamma_0$, of the $M=0$ state is $(\frac{1}{2}x)^2\gamma_s$ in this approximation. In the absence of a magnetic field the $M=+1$ state decays with the pure triplet state decay rate $\gamma_t$. The second term $\gamma'$ represents the additional decay rate due to the microwave magnetic field and corresponds predominantly to two-quantum annihilations. Provided the microwave power level is sufficiently low so that $\gamma' \ll \gamma_t$, the fraction, $P_1$, of the decays that are two-quantum annihilations is given by

$$P_1=\gamma'/\gamma_t. \tag{10}$$

The microwave magnetic field also induces transitions from the $M=-1$ state to the $M=0$ state of orthopositronium. The same expressions (9) and (10) apply. The fraction of the $M=0$ state decays that are converted to three quantum decays by the microwave field is less than that given by Eq. (10) by a factor of order $\gamma_t/\gamma_0$, and hence can be neglected in the approximation considered. The fraction $P_1$ as a function of applied frequency $\omega$ is the resonance line shape due to the annihilation lifetime. The line shape is Lorentzian with a width $\Delta f$ between half-intensity points of $(\frac{1}{2}x)^2(\gamma_s/2\pi)$ cps or a corresponding fractional half-width $\Delta f/f_{01}$ of $\gamma_s/(2\pi\Delta\nu)(=6.3\times10^{-3})$, obtained by using Eq. (4b) for $f_{01}$.

In practice the applied frequency $\omega$ is held fixed and the magnetic field $H_0$ is varied through the resonance. A useful expression for the line shape in this case is obtained from Eq. (10) by use of Eq. (4a), which relates the resonant frequency to the magnetic field value. Since the fractional variation of $H_0$ over the resonance line is small and since $x$ is small, the line shape is given to a good approximation by

$$P_1=\frac{A}{(H_0-H_{0r})^2+\frac{1}{4}\Gamma^2}, \tag{11}$$

where

$$A=\frac{\gamma_s H_{0r}^2(\mu_0 g H_1)^2}{128\gamma_t \hbar^2(\pi\Delta\nu)^2}, \quad \Gamma=\frac{\gamma_s H_{0r}}{4\pi\Delta\nu}.$$

$H_{0r}$ is the resonance value of the static field corresponding to the microwave frequency $\omega$ as given by Eq. (4a). This expression was derived by retaining only the lowest nonvanishing terms in $x$ and $\Delta x$. The resonance line given by Eq. (11) is Lorentzian in shape and has a total width $\Delta H_0$ between half-maximum points of $\gamma_s H_{0r}/(4\pi\Delta\nu)$ in gauss or a corresponding fractional half-width $\Delta H_0/H_{0r}$ of $\gamma_s/(4\pi\Delta\nu)(=3.1\times10^{-3})$. Slight deviations of the line shape from this Lorentzian form arise because of the inadequacy of the approximations that $x$ and the fractional variation of $H_0$ over the resonance line are small. These effects will be discussed in Sec. 2.4.

### 2.4. Line Shape: Higher Order Effects

There are many factors which modify the simple natural line shape given in Eqs. (9), (10), and (11) of the last section. We discuss first the modifications obtained by a more accurate treatment of the effect of the annihilation lifetime. Corrections of relative order $\gamma_t/\gamma_0$ are present to Eqs. (9) and (10) for the fraction of $M=+1$ or of $M=-1$ positronium atoms decaying by two photon annihilation. In addition, in the computation to this higher accuracy, the effect of the microwave field in modifying the decay of the $M=0$ state of orthopositronium must be considered. The total change, $P_T$, in the number of orthopositronium atoms which decay by two photon annihilations due to the microwave field divided by the number of $M=+1$ and $M=-1$ orthopositronium atoms formed is the quantity observed experimentally. The expression for $P_T$ is derived in Appendix I for the limit of low microwave fields $[|V|/\hbar \ll \frac{1}{2}(\gamma_0-\gamma_t)]$ and is given by

$$\begin{aligned} P_T=&\gamma_{02}\epsilon\left\{\frac{1}{\gamma_t}+\frac{1}{\gamma_0}-\frac{(\gamma_0+\gamma_t)}{\{\frac{1}{2}(\gamma_0+\gamma_t)\}^2+(\omega_{01}-\omega)^2}\right\} \\ &+\frac{\gamma_{02}}{\gamma_0}\epsilon\left\{\frac{\gamma_0-\gamma_t}{\gamma_0}+\frac{2[\{\frac{1}{2}(\gamma_0-\gamma_t)\}^2-(\omega_{01}-\omega)^2]}{[\{\frac{1}{2}(\gamma_0-\gamma_t)\}^2+(\omega_{01}-\omega)^2]}\right\} \\ &-\gamma_{02}\epsilon\frac{\left[\left(\frac{\gamma_0-\gamma_t}{2}\right)^2(\gamma_0+\gamma_t)+(\omega_{01}-\omega)^2(\gamma_0-3\gamma_t)\right]}{\left[\left(\frac{\gamma_0-\gamma_t}{2}\right)^2+(\omega_{01}-\omega)^2\right]\left[\left(\frac{\gamma_0+\gamma_t}{2}\right)^2+(\omega_{01}-\omega)^2\right]^2}, \end{aligned} \tag{12}$$

in which

$$\epsilon=\frac{(|V|/\hbar)^2}{(\omega_{01}-\omega)^2+[\frac{1}{2}(\gamma_0-\gamma_t)]^2}, \quad \gamma_{02}=(\tfrac{1}{2}x)^2\gamma_s$$

$V_{02}$ is the two-quantum decay rate of the $M=0$ state of orthopositronium. The first and the last two terms refer to the $M=\pm1$ and the $M=0$ states, respectively. The contribution to $P_T$ from the neglected terms of higher order in the ratio $(|V|/\hbar)/\frac{1}{2}(\gamma_0-\gamma_t)$ will amount to about 4% at the microwave power level used. $P_T$ is symmetrical with respect to frequency variations on either side of the resonance frequency $\omega_{01}$, but the line shape is not Lorentzian. The half-width is about 1.15 times the half-width of the line given by Eq. (10) or $\Delta f/f_{01}=7.2\times10^{-3}$.

A line shape can be derived from Eq. (12) with the assumptions used to derive Eq. (11) from Eq. (10), which is applicable to the actual experiment in which the magnetic field $H_0$ is varied and the microwave frequency $\omega$ is fixed. This line shape is symmetrical in field about the resonant value but is not Lorentz shaped. Its half-width is 1.15 times the half-width of the line given by Eq. (11) or $\Delta H_0/H_{0r}$ is $3.6\times10^{-3}$. This half-width will be the natural width of the resonance line due to the annihilation lifetime of positronium. The inaccuracy introduced in the derivation from Eq. (12) of the line shape as a function of $H_0$ by the neglect of higher order terms in $x$ and $\Delta x$ alters the line width by only about 1%. Furthermore, numerical machine calculations disdussed in Sec. 5.2 have established that the center of the resonance line given in Eq. (11) agrees with the resonance value of the field given by Eq. (10) to within an accuracy of 1 part in 20 000, which is sufficient for the applications in this paper. A similar agreement can be expected between the resonance field given by Eq. (12) and the center of the resonance line derived from Eq. (12) with the assumptions that $x$ and $\Delta x$ are small.

Power broadening of the natural resonance line will be present at sufficiently high microwave fields. The effect of power broadening to the lowest order in $|V|/\hbar$ can be derived from Eq. (9). The expression for $P_1$ given in Eq. (10) neglects the effect of power broadening because its derivation used the condition $\gamma' \ll \gamma_t$. If this condition is not valid, $P_1$ is given by

$$P_1 = \gamma'/(\gamma_t + \gamma'). \tag{13}$$

This expression includes the effect of power broadening on the simple natural line shape.

The velocity of the positronium atoms in the gas will determine the Doppler width of the resonance line. Positronium is formed by the capture of an electron from a gas atom by a positron, and it is expected that most positronium atoms will be formed with a kinetic energy of several electron volts.[13] If a geometrical cross section is used for the collision of positronium with an argon atom and if a pressure of one atmosphere of argon is assumed, a positronium atom formed in the $M = \pm 1$ states of orthopositronium will undergo some $10^5$ elastic collisions during its lifetime of $10^{-7}$ sec. Because positronium has so small a mass, however, it may be expected to retain a kinetic energy of about 1 ev during its lifetime. Measurements[14] of the angular correlation between annihilation quanta from positronium decays in argon at pressures of 17 and 28 atmospheres confirm that singlet positronium, which has a lifetime of $10^{-10}$ sec, decays while having a kinetic energy of 1 or 2 ev. Furthermore, these angular correlation measurements made in argon at 28 atmospheres in a static magnetic field of 10 kilogauss indicate that the $M=0$ state of orthopositronium retains about 0.3 ev during its lifetime of $5 \times 10^{-9}$ sec. These observations confirm the expectation that under our conditions with a pressure of 1 or 2 atmos positronium will retain a kinetic energy of about 1 ev. At this kinetic energy positronium has a velocity of $4 \times 10^7$ cm/sec, and a fractional Doppler broadening of $2 \times 10^{-3}$ might be expected. However, collisions of the positronium atom with gas atoms occur with a mean free path $L$ which is small compared to the wavelength $\lambda$ of the resonant radiation involved. In this case only a reduced Doppler width is observed.[15] The fractional reduced Doppler width is given by: $(2.8L/\lambda) \times$ (normal Doppler width). Under our conditions $L \simeq 10^{-4}$ cm and $\lambda \simeq 12$ cm, so the reduced Doppler width is negligible.

Collision line broadening or line shifts are not expected to be important because the energy levels involved are not easily perturbed by electric forces and magnetic forces are small. In Appendix II some theoretical considerations on collisions are presented, but it should be emphasized that reliable theoretical estimates of collision effects are difficult to make. Experimentally, measurements of the positronium hyperfine structure separation in various gases and at various pressures have indicated no significant effect of gas or pressure on the position of the center or the width of the resonance line.

[13] H. S. W. Massey and C. B. O. Mohr, Proc. Phys. Soc. (London) A67, 695 (1954).

[14] M. Heinberg and L. A. Page, Bull. Am. Phys. Soc. Ser. II, 1, 168 (1956).

[15] R. H. Dicke, Phys. Rev. 89, 472 (1953); J. P. Wittke and R. H. Dicke, Phys. Rev. 103, 620 (1956).

### 2.5. Gamma-Ray Annihilation Spectra

An analysis of the gamma-ray energy spectra can be made to determine the fraction $P_1$ of $M = \pm 1$ orthopositronium atoms which decay by two quantum annihilation due to the microwave field. Equations similar to Eq. (6) of reference 11 can be written for the peak and valley counts for the three cases: (0) with a small admixture of NO in the principal gas, (1) with a microwave frequency off the resonance line, and (2) with a microwave frequency on the resonance line.

$$\begin{aligned}
P^0 &= N_p g_p + N_a g_p + N_0 g_p, \\
V^0 &= N_p h_p + N_a h_p + N_0 h_p, \\
P^1 &= N_p g_p + N_a g_p + N_0 g_0, \\
V^1 &= N_p h_p + N_a h_p + N_0 h_0, \\
P^2 &= N_p g_p + N_a g_p + N_0(1-P_1) g_0 + N_0 P_1 g_p, \\
V^2 &= N_p h_p + N_a h_p + N_0(1-P_1) h_0 + N_0 P_1 h_p.
\end{aligned} \tag{14}$$

The notation and underlying assumptions for these equations are similar to those of reference 11 and will only be briefly summarized here. The quantities $N_a$, $N_p$, and $N_0$ 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. Hence the number of three-quantum annihilations, $N_0$, is due principally to the $M = \pm 1$ substates of orthopositronium, and the majority of the annihilations of the $M=0$ substate of orthopositronium are included with the two-quantum annihilations $N_p$. $P_1$ is the fraction of the $M = \pm 1$ orthopositronium three quantum annihilations that are converted to two quantum annihilations by the microwave field. The small fraction of $M=0$ orthopositronium decays that are converted to three quantum decays by the microwave field is neglected in Eq. (14). The factors $g_p$, $g_0$, $h_p$, and $h_0$ are the efficiency factors for the detection of two-quantum and three-quantum annihilations in the peak and in the valley regions, respectively.

Solution of Eqs. (14) for $P_1$ yields, without approximation,

$$P_1=y/(1+y), \qquad (15)$$

where

$$y=\rho(R^1-R^2)/(R^2-R^0),$$

in which $R^0=V^0/P^0$, $R^1=V^1/P^1$, $R^2=V^2/P^2$, and $\rho=P^1/P^0$. If the microwave frequency in case (2) is actually off the resonance curve, $y=0$ and $P_1=0$ as it must. If the change in valley to peak ratio within the resonance curve $(R^1-R^2)$ is small compared to the change $(R^1-R^0)$, which corresponds to complete conversion of three-quantum annihilations to two-quantum annihilations, then $y$ is small and the series expansion is valid:

$$P_1=y-y^2+\cdots \qquad (16a)$$

or

$$P_1\simeq\rho(R^1-R^2)/(R^2-R^0). \qquad (16b)$$

Thus it is seen from the leading term that $P_1$ is proportional to the change $(R^1-R^2)$ in the valley to peak ratio. The departure from proportionality is of the order $y$ and in practice amounts to about 10%. (See Sec. 5.1.)

Several further comments can be made about the approximations implied by the use of Eq. (14). The values of $N_a$, $N_p$, and $N_0$ may be different for case (0) than for cases (1) and (2) due to the presence of the NO gas,[16] but if the total number of positrons annihilating is the same in all cases, as can be expected, then Eq. (15) is still valid. The different angular distribution of the gamma rays from the three-quantum annihilations of the different magnetic substates of orthopositronium[11] need only be considered insofar as there is an appreciable fraction of three-photon annihilation from the $M=0$ state of orthopositronium; its effect is negligible in the present experiment. Background gamma rays from positrons annihilating in the cavity walls will be the same for cases (0), (1), and (2), and can be included in the number of free positron-electron annihilations $N_a$.

The effect of the small conversion of $M=0$ state two-quantum annihilations to three-quantum annihilations by the microwave field can be included by a modification of Eq. (14). If the ratio of the fraction of two-quantum annihilations $N_p$ converted to three-quantum decays to the fraction of three-quantum annihilations $N_0$ converted to two-quantum decays is denoted by $\eta$, the solution to the appropriately modified Eq. (14) is

$$P_T=P_1(1-N_p\eta/N_0)=y/(1+y), \qquad (17)$$

in which $P_T$ is the change in the number of 3-quantum decays divided by the number of $M=+1$ and $M=-1$ positronium atoms formed. Actually the quantity $\eta$ varies with position on the resonance curve, but it is approximately equal to $\gamma_t/\gamma_0$ (see Appendix I). Since $N_p\simeq N_0$, it is seen that $P_T$ differs from $P_1$ by about 7%. The experimentally determined value of $P_T$ obtained from Eq. (17) is to be compared with the theoretical value for $P_T$ given by Eq. (12).

[16] Gittelman, Dulit, and Deutsch, Bull. Am. Phys. Soc. Ser. II, 1, 69 (1956).

## 3. APPARATUS

The apparatus consisted of a cavity in which positronium was formed together with an associated gas handling system; a scintillation spectrometer to measure the energy spectrum of the annihilation $\gamma$ rays; a microwave system to provide a microwave magnetic field in the cavity and to monitor its power and frequency; an electromagnet and equipment to measure the magnetic field. Part of this apparatus has been described previously[11]; further information will be given in this section.

### 3.1. Magnetic Field

An electromagnet with a pole-face diameter of 9 in. and a gap of $2\frac{31}{32}$ in. was used. The coils were connected in series and were run from a 20-volt source of 10 series-connected 2-volt submarine batteries. The magnetic field of about 8000 gauss was obtained with a current of 220 amperes, which could be supplied by the batteries for some 12 hours before excessive drifting set in. Series-connected brass water-cooled trombones served for adjustment of the current and also for manual monitoring of the field during a run. Rose shims of conventional design[17] were used to improve the homogeneity of the field. The magnetic field was measured with a proton resonance probe[18] having a volume of about 0.5 $cm^3$ (0.1 normal solution of $CuSO_4$ in $H_2O$), and the frequency of about 34 Mc/sec was measured to 1 part in 20 000 with a General Radio 620 A Heterodyne Frequency Meter. The 60-cps field modulation was supplied by small auxiliary Helmholtz coils placed on either side of the proton sample; the modulation amplitude was sufficiently low so that the field at the probe was modulated by less than 1 part in $10^4$. The field inhomogeneity was about 5 parts in $10^4$ over a region 1 in. in diameter and $\frac{3}{4}$ in. in length (measured in the direction of the field). Photographic studies of the distribution of electrons from $G^{137}$ in the cavity with a pressure of 1 atmosphere and with a magnetic field of 8000 gauss indicated that this is the region from which the annihilating gamma rays are observed.

### 3.2. Cavity and Microwave System

Because the lifetime of positronium is very short due to the annihilation process and because the transition to be induced is a magnetic dipole transition, microwave magnetic fields of up to 10 gauss are required. A schematic diagram of the microwave system is shown in Fig. 2. The power source was a cw fixed-frequency magnetron (Raytheon QK390) with a rated power output of 850 watts at 2460 Mc/sec. The tube was generally

[17] M. E. Rose, Phys. Rev. 53, 715 (1938).

[18] R. V. Pound and W. D. Knight, Rev. Sci. Instr. 21, 219 (1950).

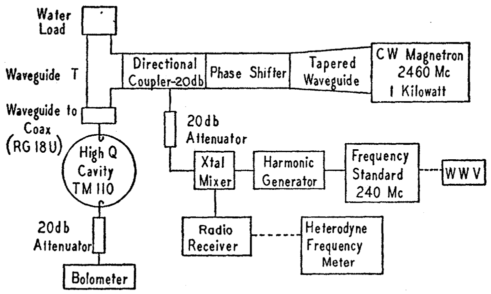


FIG. 2. Microwave system.

operated with an output power of about 600 watts. Since a substantial fraction of the magnetron output power must be delivered to the cavity, the coupling between the magnetron and the cavity was strong. Considerable difficulty was experienced in the design of the microwave circuit because of the interaction between the magnetron and the cavity; for the final circuit shown the VSWR is less than about 1.6 and a power between 100 and 200 watts is supplied to the cavity. The tapered waveguide adapts the magnetron output to an $S$-band wave guide. The phase shifter[19] was required for impedance matching to obtain stable magnetron operation. The water load served merely to dissipate power and thus to reduce the power reflected to the magnetron. The magnetron signal taken from the 20 db directional coupler was used to monitor the frequency. This signal was mixed with a harmonic at 2480 Mc/sec from the Columbia frequency standard and the beat frequency was observed on a National Company HRO-50 radio receiver which was calibrated against the General Radio 620 A Heterodyne Frequency Meter.

The cavity shown in Fig. 3 was a circular cylinder 14.745 cm in diameter and 3.912 cm in length with silver plating on the inside to a depth of 0.0005 cm. The cavity was mounted in the magnet gap so that its axis was in the direction of the static magnetic field. It operated in the $TM_{110}$ mode and the dimensions given correspond to a resonant frequency of 2480 Mc/sec. This mode has the advantages of providing a strong microwave magnetic field transverse to the static magnetic field at the center of the cavity and a field configuration independent of the $z$ (axial) coordinate, and of requiring reasonably small dimensions for a given wavelength. Throughout a region of $\frac{3}{4}$-in. radius from the axis of the cavity, which should include most of the observed positronium atoms, the microwave magnetic field is a linearly polarized field of constant amplitude to within an accuracy of about 10%. Coarse tuning of the cavity to the magnetron frequency of about 1% was accomplished by insertion of an appropriate amount of Teflon in the form of a spoked wheel $\frac{1}{8}$ in. in depth, which was held in place by friction. A circular depression at the center of this Teflon piece $\frac{5}{8}$ in. in diameter and $\frac{3}{32}$ in. deep was provided to hold the positron source which was clamped in place by a Teflon ring with an inside diameter of $\frac{7}{16}$ in. The source was a copper disk 0.001 in. thick and $\frac{5}{8}$ in. in diameter which had been bombarded at the Brookhaven pile to produce about a 90-mC source of $Cu^{64}$. Finer tuning was provided by movement of a Teflon plug $1\frac{3}{8}$ in. in diameter and 1 in. in height. The movement of about $\frac{1}{2}$ in. was controllable external to the cavity via a bellows arrangement and provided a tuning of about 1 part in $10^3$.

The input loop to the cavity had an area of about 0.5 $cm^2$ and was designed empirically so that the cavity presented at resonance a matched load of 50 ohms with a VSWR of less than 1.5. The input connector which held the input loop was a UG-30C/U type $N$ vacuum-sealed connector modified with a flange and O-ring groove so that it could be screwed into the cavity wall making an O-ring seal. The output coupling loop which was placed diametrically opposite the input loop had an area of about 0.025 $cm^2$ and was held by a similar connector. A third hole in the side of the cavity provided for evacuating and filling of the cavity. The cavity had a measured $Q$ of[20] about 11 000 as compared to the theoretical $Q$ of 18 000 when perturbing effects are neglected. If an input power of 150 watts is supplied to the cavity, the amplitude of the microwave magnetic field at the center is about 11 gauss. The output loop was connected through a 20-db attenuator to a bolometer. A typical value of the bolometer power reading was 30 mw, which implies a field at the center of the cavity of 22 gauss. This field determination was subject to considerable error as an absolute determination due to lack of knowledge about the exact effective output loop area

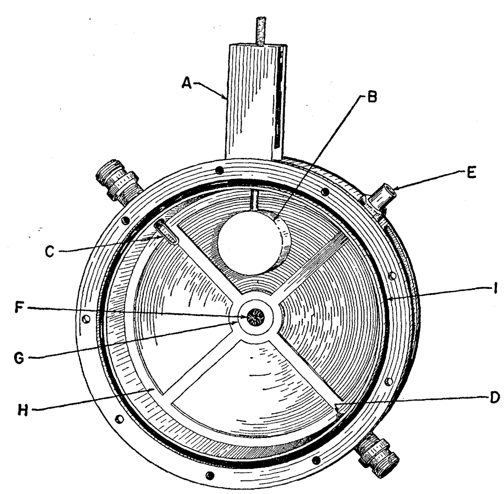


FIG. 3. Microwave cavity. $A$, external bellows control for tuning plug; $B$, Teflon tuning plug; $C$, rf input loop; $D$, rf output loop; $E$, gas inlet; $F$, radioactive source of $Cu^{64}$; $G$, Teflon source holder; $H$, Teflon piece for tuning and for support of source; $I$, O-ring.

[19] *Microwave Transmission Circuits*, edited by G. L. Ragan, Massachusetts Institute of Technology Radiation Laboratory Series (McGraw-Hill Book Company, Inc., New York, 1948), Vol. 9, p. 514.

[20] C. G. Montgomery in Massachusetts Institute of Technology Radiation Laboratory Series (McGraw-Hill Book Company, Inc., New York, 1947), Vol. 11, p. 333.

and due to uncertainty about the absolute calibration of the attenuator and bolometer. Copper tubing was soldered to the outside wall of the cavity to permit water-cooling.

### 3.3. Gas Handling System and Gamma-Ray Spectrometer

The gas handling system was the same as that described before.[11] The cavity was evacuated to a pressure of about $5\times10^{-5}$ mm of Hg, and then filled with the gas to pressures from $\frac{2}{3}$ to 2 atmospheres. Ordinary commercial-grade tank gases were used without purification.

The energy distribution of the annihilation gamma rays was observed with a NaI(Tl) scintillation spectrometer using a Dumont 6292 photomultiplier. The geometry associated with the collimation was the same as that used previously. The wall of the cavity through which the gamma rays reach the crystal was cut to a thickness of only $\frac{1}{8}$ in. to reduce Compton degradation. The pulse-height analyzer of the earlier reference was replaced by a commercial Atomic Instrument Company Model 520 twenty-channel differential pulse-height analyzer. Spectra of the type shown in Fig. 2 of reference 11 were observed. The ratio of the number of counts in the valley region from about 345 kev to 390 kev to the number of counts in the peak region from about 485 kev to 530 kev is essentially proportional to the ratio of the number of three-quantum annihilations to the number of two-quantum annihilations; and if a transition is induced in a strong static magnetic field from the $M=\pm1$ states of orthopositronium to the $M=0$ state of orthopositronium, this ratio will decrease.

### 4. PROCEDURE

The basic experimental data are values of the valley counts $V$ and the peak counts $P$, observed as a function of the static magnetic field $H_0$ with the microwave frequency and power fed into the cavity held constant. A typical resonance curve is shown in Fig. 4. For each point approximately 10 000 counts were taken for the valley region and 70 000 counts for the peak region. Both valley and peak regions were observed simultaneously with the 20-channel differential pulse-height analyzer. Hence the statistical accuracy of a single value of the valley to peak ratio is about 1%. A single point could be obtained in 4 to 10 minutes and an entire resonance curve involving some 25 points could be taken in 2 to 3 hours. A curve was usually obtained by varying the static magnetic field through the resonance in one direction and then back through the resonance in the opposite direction to obtain points located at field values intermediate between those of the first traversal. A range in field of about 2% was covered with a spacing between points of about 0.05% over a central range of 1% and with several points at the limits of the range.

The static magnetic field was measured with the proton resonance probe which was placed outside the cavity on the axis of the cavity and magnet about $1\frac{1}{4}$ in. from the center of the active volume of the cavity. The field was monitored to about 1 part in 20 000 during the counting time for each point by manually adjusting the trombone rheostat. This monitoring was found necessary primarily near the end of a run to compensate for battery drift. After taking a resonance curve the cavity was removed, and, without being turned off, the magnetic field was measured in the region of the field from which positronium annihilations were observed as well as at the position occupied by the probe while the resonance curve was being obtained. The field difference between these two positions was about 3 parts in $10^4$, and was quite constant from run to run. The variation of the magnetic field throughout the region from which positronium annihilations were observed was also measured occasionally after taking a resonance curve, and was found to be quite reproducible and equal to about 5 parts in $10^4$. Many auxiliary studies were made on the magnetic field inhomogeneity at times other than during a run, and it was found that the field inhomogeneity pattern was very reproducible from day to day. The pattern was independent of magnetic field value over the range of fields involved in the experiment, and, furthermore, it did not exhibit any hysteresis effects. Several slightly different positionings of the Rose shims were employed to obtain somewhat different inhomogeneity patterns for different runs.

The magnetron frequency was measured often during a run and was seldom found to vary by more than 1 part in 20 000. The output power from the cavity was measured for each point and was found to change in a random manner during a run, usually by about ±5% but occasionally by as much as ±10%. Zero drift of the bolometer was often troublesome, and a 1N21A crystal was sometimes substituted as a detector after the power had first been measured with the bolometer. Attempts to maintain the power level constant by tuning the cavity or by varying the phase shifter were unsuccessful,

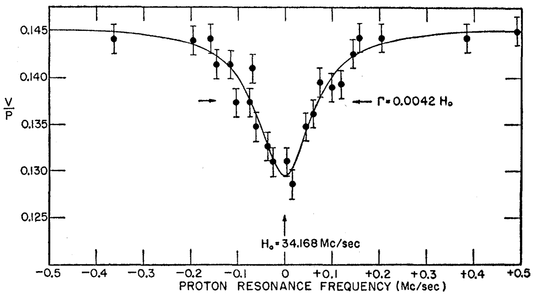


FIG. 4. Typical resonance curve. Values of the ratio of gamma-ray counts in the valley region (345 kev to 390 kev) to counts in the peak region (485 to 530 kev) are shown *versus* magnetic field, measured with a proton resonance probe, for a fixed microwave frequency of 2457.9 Mc/sec. Statistical counting errors are indicated for the experimental points. The solid curve is the Lorentzian curve of the form given by Eq. (11) which gives the best least-squares fit to the experimental points.

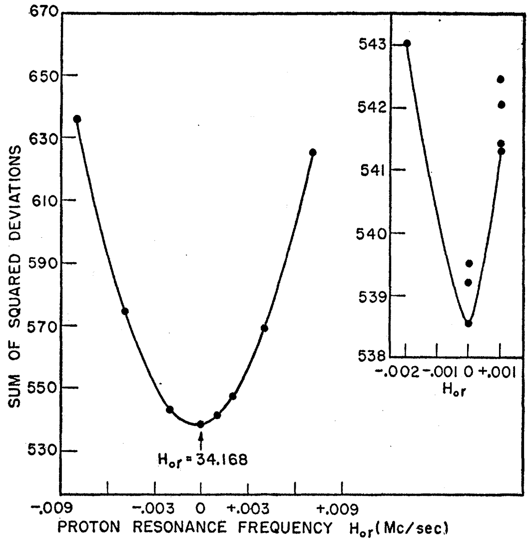


FIG. 5. Illustrating the least-squares fit of the theoretical Lorentzian curve of Eq. (11) to the experimental data. The curve on the left shows the sum of the squared deviations of the experimental points from the theoretical curve as a function of the assumed resonance value $H_{0r}$. Each point represents the smallest sum obtained by varying the other parameters $A$ and $\Gamma$ of the theoretical curve. The curve on the right shows the sum of the squared deviations over a small region of $H_{0r}$ near the resonance value chosen. The several values shown for a particular $H_{0r}$ correspond to different choices of the other parameters of the theoretical curve. The ordinates are in arbitrary units.

because such adjustments changed the magnetron frequency.

## 5. SUMMARY OF DATA AND RESULTS

### 5.1. Analysis of Data

Each experimental curve of the form of Fig. 4 was analyzed by fitting a Lorentz-shaped theoretical curve as given by Eq. (11) to the observed points by the method of least squares. The amplitude, width, and resonant field were varied to obtain a curve which gives the best least-squares fit to the observed points. This computation is very laborious to perform by the use of desk computers and it was programmed for an IBM 650 magnetic drum decimal computer.[21] The program was arranged with suitable intervals of parameter variation to obtain the resonant field to about 1 part in 30 000, which is about the accuracy with which the magnetic field was measured. This corresponds to choosing the resonant field to 1 part in 100 of the line width and to 1 part in 15 of the spacing between successive observed points. A typical curve showing the sum of the squares of the deviations as a function of the resonant field is given in Fig. 5. Checks were made to indicate that the final resonant field chosen is independent of the initial values taken for the parameters of the curve. Sufficient computations were made to be sure that no false minima were obtained. For each fitting some 400 sets of values of the parameters were tried, and the machine time required for the fitting of a single curve was about 2 hours. Computations made for several runs using different energy widths (number of channels) for the valley and peak regions yielded no significant differences.

The solid curve drawn through the experimental points in Fig. 4 is a Lorentzian curve of the form of Eq. (11) which represents the least squares fit to the experimental data. The center or minimum of the theoretical curve is taken as the resonant field $H_{0r}$ and the corresponding value of $\Delta\nu$ is computed using Eq. (4a). The observed change in $V/P$ at resonance corresponds by use of Eq. (16b) to a probability of about 0.25 for a positronium atom in the $M=\pm1$ substates of orthopositronium to die by two quantum annihilation.[22] This value of $P_1$ corresponds to a microwave field of 11-gauss amplitude, which is consistent with the accuracy of the field measurement described in Sec. 3.2. The line width between half-intensity points, $\Delta H_0/H_{0r}$, is seen to be $4.2\times10^{-3}$. The theoretical value for the half-width of the resonance line due to the annihilation lifetime is $3.6\times10^{-3}$ as given by Eq. (11) with the correction factor of order $\gamma_t/\gamma_0$ obtained from Eq. (12). For the high microwave field of about 10 gauss amplitude used, power broadening will increase the expected half-width to $4.0\times10^{-3}$. This figure is obtained by adding arithmatically to the natural width of Eq. (11) the extra width due to power broadening obtained from Eq. (13) and the extra width of order $\gamma_t/\gamma_0$ obtained from Eq. (12). The additional observed width of $0.2\times10^{-3}$ is attributed to the field inhomogeneity of about $0.5\times10^{-3}$ over the active region. The average fractional half-width of all the resonance lines given in Table I is $4.5\times10^{-3}$, which is in good agreement with the theoretical half-width as just computed.

A list of the data obtained is given in Table I. Eleven curves in all were obtained for the gases A, $N_2$, and $SF_6$ at several different pressures between $\frac{2}{3}$ and 2 atmospheres. Column IV indicates the microwave power level in the cavity. The power level varies by about a factor of 2, which represents the practical limit available, because lower powers gave insufficient signal and the higher powers indicated were the maximum ones achieved with our system. The number of points taken for the curves is indicated in column V. Relative weight factors are given in columns VI and VII. The weighting factor $w$ is proportional to the root-mean-square deviation for a point on the curve. The total relative weight assigned to a curve is proportional to the product of $w$ by the square root of the number of points on the curve. It varies from 1.0 to 3.1.

Values obtained for the width between half-intensity points and for the hyperfine structure separation corresponding to the resonant field chosen are given in

[21] We are indebted to the IBM Watson Scientific Computing Laboratory for the opportunity to use their computing machine.

[22] The values of 0.075 and 0.82 are taken for $R^0$ and $\rho$, respectively. These are typical measured values with an accuracy of about 10%. See reference 11 for further discussion of these quantities.

TABLE I. Summary of experimental data. Each row, which is designated by its run number, corresponds to a single resonance curve. Columns II and III list the gas and pressure at which the particular run was taken; column IV gives the average microwave power level in the cavity read by the bolometer in the circuit arrangement described in Sec. 3.2. Column V gives the number of points on the curve. Column VI lists the values of a partial weighting factor, $w$, which is proportional to the root mean square deviation of the observed points from the best-fit Lorentzian curve. Column VII lists the over-all weighting factors which are proportional to $w$ times the square root of the number of points. Column VIII lists the values of the total half-width between half-intensity points of the best-fit Lorentzian curve. Column IX gives the values of $\Delta\nu$.

| I | II | III | IV | V | VI | VII | VIII | IX |
|---|---|---|---|---|---|---|---|---|
| Run No. | Gas | Pressure (atmospheres) | Microwave power (bolometer reading in milliwatts) | Number of points | $w$ | Relative weight | Half-width ($\Delta H_0/H_{0r}$ in units of $10^{-3}$) | $\Delta\nu$ (in units of $10^5$ Mc/sec) |
| 1 | $SF_6$ | 1.2 | 28 | 19 | 1.2 | 1.4 | 4.2 | 2.03306 |
| 2 | $N_2$ | 2.0 | 27 | 24 | 2.0 | 2.6 | 4.2 | 2.03332 |
| 3 | $SF_6$ | 2.0 | 30 | 26 | 1.0 | 1.4 | 3.7 | 2.03262 |
| 4 | $N_2$ | 2.0 | 32 | 19 | 1.9 | 2.3 | 5.6 | 2.03372 |
| 5 | $N_2$ | 2.0 | 32 | 29 | 2.1 | 3.1 | 4.7 | 2.03362 |
| 6 | $N_2$ | 1.2 | 31 | 11 | 1.1 | 1.0 | 2.6 | 2.03260 |
| 7 | $N_2$ | 1.2 | 31 | 28 | 1.9 | 2.7 | 6.3 | 2.03338 |
| 8 | $SF_6$ | 0.7 | 24 | 16 | 1.5 | 1.6 | 4.4 | 2.03291 |
| 9 | $N_2$ | 2.0 | 17 | 16 | 1.8 | 1.9 | 3.7 | 2.03362 |
| 10 | A | 2.0 | 19 | 30 | 1.9 | 2.9 | 3.8 | 2.03343 |
| 11 | A | 2.0 | 20 | 19 | 1.6 | 1.9 | 4.0 | 2.03257 |

Half-width, $\Delta H_0/H_{0r}=(4.5\pm0.03)\times10^{-3}$ (average and standard deviation of weighted values)
$\Delta\nu=(2.03317\pm0.00012)\times10^5$ Mc/sec (average and standard deviation of unweighted values)
$\Delta\nu=(2.03327\pm0.00008)\times10^5$ Mc/sec (average and standard deviation of weighted values)

columns VIII and IX, respectively. The observed variations in $\Delta\nu$ and in half-width are believed to be due primarily to statistical variations in the number of valley and peak counts. The average half-width of $4.5\times10^{-3}$ is accounted for by the line width factors discussed for Fig. 4. The unweighted average of the values for $\Delta\nu$ is $(2.03317\pm0.00012)\times10^5$ Mc/sec, where the standard deviation of the group of values is given. The weighted average and standard deviation of the values of $\Delta\nu$ is $(2.03327\pm0.00008)\times10^5$ Mc/sec. A histogram of these weighted values together with a Gaussian curve which has been matched in area to the histogram and which has the central value and rms deviation of the data is shown in Fig. 6. The values of the weighted averages for the different gases A, $N_2$, and $SF_6$ are $2.0331\times10^5$ Mc/sec, $2.0335\times10^5$ Mc/sec, and $2.0329\times10^5$ Mc/sec, respectively. These values agree within their statistical accuracies, and hence indicate that there is no dependence of the $\Delta\nu$ value on the gas used to within the experimental accuracy. Nor are there any significant variations with pressure for the individual gases. The last three runs were taken with significantly lower microwave power than all the previous runs. The weighted average of these three runs is $2.0332\times10^5$ Mc/sec, which is in agreement with the average $\Delta\nu$ from all the data, and hence indicates that there is no dependence of $\Delta\nu$ on the microwave power level used.

### 5.2. Estimate of Error

The observed variation in the value of $\Delta\nu$ from run to run is believed to be primarily random in nature and due to the limited number of counts obtained for each point on a curve. A measure of the error to be expected from this cause is, of course, the standard deviation of the weighted $\Delta\nu$ values for all the runs taken as a group. Its value is $0.00008\times10^5$ Mc/sec as given in Table I.

The inhomogeneity of the magnetic field could contribute a significant systematic error. The variation of the magnetic field over the active region is about 5 parts in $10^4$. The value of the field in the region near the axis of the cavity and magnetic field and opposite the viewing hole for the gamma rays has been taken as the correct field value. Because the positronium decays arise mostly from this region and because the microwave power is highest near the axis, this choice of field value seems to be the reasonable one. The distribution of positronium decays is expected to be symmetrical about the axis, but no particular symmetry is expected along the axial direction. The distribution of magnetic field inhomogeneity need have no special symmetry throughout the active volume. With this situation obtaining, an

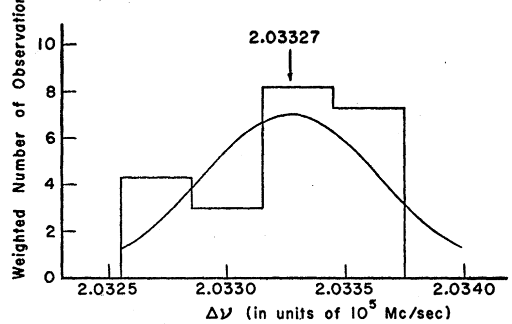


FIG. 6. Histogram of weighted $\Delta\nu$ determinations. The solid curve is a Gaussian curve which has been matched in area to the histogram and which has the rms deviation of the data. The weighted average value of $\Delta\nu$ is indicated.

asymmetry could be introduced in the resonance curve which would result in an apparent shift in the resonance value. The principal means of assuring that this error is small has been to take a considerable body of data, especially with some variation in the conditions of magnetic field inhomogeneity, and thus to tend to make this error a random one. No diamagnetic effects of any kind are sufficiently large to be a concern.

A deviation of the theoretical curve from the Lorentz form given by Eq. (11) could cause a systematic error. In the derivation of Eq. (11) from Eq. (10), higher order terms in $\Delta x$ and $x$ were neglected. Their inclusion would alter the Lorentzian form of the theoretical curve. Estimates of the effect of these terms on the curve indicate that the amplitude of the curve is changed by considerably less than the statistical error of the points. Furthermore, for five runs numerical computations were made of the resonant field using a theoretical curve corresponding to Eq. (11) but including higher order corrections in $x$ and $\Delta x$. The values of $\Delta\nu$ so obtained were in agreement with the values obtained from Eq. (11) to 1 part in 20 000. This agreement is within the accuracy to which the machine computation had been programmed. No calculations were made to fit the experimental data by the more exact theoretical line shape given by Eq. (12). All of the theoretical line shapes [Eqs. (10), (11), and (12)] are symmetrical about the resonance value of frequency or field, and it is believed that the same resonance value of field will be obtained with Eqs. (10) and (12) to well within the experimental accuracy. The principal importance of Eq. (12) is that it indicates that the half-width of the resonance curve should be somewhat larger than that predicted by Eq. (10).

A lack of proportionality between the change in the observed ratio of valley counts to peak counts and the fraction $P_1$ of positronium atoms formed in the $M=\pm 1$ states of orthopositronium which decay by two-quantum annihilation could also cause a systematic error. Exact computations were made for several runs in which $P_1$ was obtained from Eq. (15) rather than from the leading term of Eq. (16b). These calculations were made with the IBM 650 using both the theoretical line shape of Eq. (11) and the more exact form derived from Eq. (10) including higher order terms in $x$ and $\Delta x$. The values obtained for $\Delta\nu$ agree with the value obtained using Eq. (15).

Shifts in the energy levels given by Eq. (3) due to the annihilation process are of the order of $(\gamma_s/2\pi\Delta\nu)^2 \simeq 4\times 10^{-5}$ [23] and hence can be neglected. Furthermore, energy shifts due to the large microwave magnetic field are negligible, as is pointed out in Appendix I.

The final value for $\Delta\nu$ obtained from the present experiment is:

$$\Delta\nu = (2.0333 \pm 0.0004)\times 10^5 \text{ Mc/sec.}$$

The error estimate is about five times the standard deviation of the data. It corresponds to choosing the center of the resonance line to about 1/40 of its half-width. Since the resonance line is appreciably broadened by inhomogeneity of the magnetic field, which could introduce a systematic error, it is felt that the quoted error is a realistic upper limit of error.‖

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

## 6. DISCUSSION

The present paper has described a determination of the hyperfine structure separation of positronium in its ground state by a method essentially similar to that of Deutsch, Brown, and Weinstein.[5] The experiments of Deutsch *et al.* were done at static magnetic field values of 9000 gauss and 9500 gauss and yielded the values $\Delta\nu = (2.0335\pm 0.0005)\times 10^5$ Mc/sec and $(2.0338\pm 0.0004)\times 10^5$ Mc/sec, respectively. The experiment reported in the present paper was done at a magnetic field of about 8000 gauss and yielded the value $\Delta\nu = (2.0333\pm 0.0004)\times 10^5$ Mc/sec. All of these values for $\Delta\nu$ agree within the experimental limits quoted and hence confirm the theory of the dependence of the energy levels on magnetic field. The average of these three determinations of $\Delta\nu$ is $(2.0336\pm 0.0002)\times 10^5$ Mc/sec, in which a standard deviation of the three values is given.

The theoretical value for $\Delta\nu$ has been computed to order $\alpha^3$ ry by Karplus and Klein[4] [see Eq. (1)] and is equal to $2.0337\times 10^5$ Mc/sec. The experimental value is in excellent agreement with this theoretical value, and hence provides strong confirmation of the theory of positronium based on the Bethe-Salpeter equation. It may be noted that the first neglected term in the theory is of order $\alpha^4$ ry. Since the leading term is of order $\alpha^2$ ry, the first neglected term is of relative order $\alpha^2$ or 1 part in 20 000. Hence the magnitude of the $\alpha^4$ ry term is approximately equal to the present experimental uncertainty, and, since numerical factors in the theory might make it somewhat larger, an extension of the theory to the order $\alpha^4$ ry appears desirable at this time.

It is a pleasure to acknowledge several stimulating discussions with Professor Breit about the theory of the resonance line shape.

## APPENDIX I. THEORY OF RESONANCE LINE SHAPE

The theory of the resonance line shape is similar to that for the hydrogen fine structure transitions.[24] A quantitative expression for the positronium line shape

‖ *Note added in proof.*—The recent discovery that positrons originating from beta decay are preferentially polarized along their direction of emission [T. D. Lee and C. N. Yang, Phys. Rev. **105**, 1671 (1957); L. A. Page and M. Heinberg, Bull. Am. Phys. Soc., Ser. II, **2**, 172 (1957), and preprint of article for Phys. Rev.] does not alter the final value quoted for $\Delta\nu$. The fact that the magnetic substates of positronium are not all formed with equal probability because of the polarization of the positrons will change slightly some of the discussion of the theoretical line shape and of the analysis of the gamma ray spectra, but will not affect the value of $\Delta\nu$ quoted. See forthcoming note on the effect of the polarization of the positrons on positronium experiments.

[24] W. E. Lamb and R. C. Retherford, Phys. Rev. **79**, 549 (1950).

will be derived in this appendix. Halpern has discussed the effect of static and alternating magnetic fields on positronium decay, and his results will be useful.[23]

Consider the Hamiltonian

$$\mathcal{H}=\mathcal{H}_s+\mathcal{H}'(t),$$

in which $\mathcal{H}_s$ is the time independent part of the Hamiltonian including the interaction with the static magnetic field $\mathbf{H}_0$ in the $z$ direction, and $\mathcal{H}'(t)$ is the interaction with the microwave field $\mathbf{H}_1\cos\omega t$, which is linearly polarized in the $y$ direction [see Eq. (8)]. It is convenient to express the microwave field in the form:

$$H_1\cos\omega t\hat{y}=-(iH_1/4)(\hat{x}+i\hat{y})(e^{+i\omega t}+e^{-i\omega t}) +(iH_1/4)(\hat{x}-i\hat{y})(e^{+i\omega t}+e^{-i\omega t}), \quad (18)$$

in which $\hat{x}$ and $\hat{y}$ are unit vectors in the $x$ and $y$ directions. $\mathcal{H}'(t)$ has nonvanishing matrix elements between the $M=+1$ and the $M=0$ states of orthopositronium and between the $M=-1$ and $M=0$ states of orthopositronium.

Consider first the two magnetic sublevels $M=+1$ and $M=0$ of orthopositronium, which are eigenstates of $\mathcal{H}_s$ with energies $W_{1,1}$ and $W_{1,0}$, respectively. Denote their probability amplitudes by $a_1$ and $a_0$. The two state functions, $u_{1,1}$ and $\psi_{1,0}$, are normalized and are orthogonal to one another. We consider only that part of $\mathcal{H}'(t)$ which has a nonvanishing matrix element between these two states and which leads to a resonance term in Eq. (27). The states $M=+1$ and $M=0$ of orthopositronium annihilate with the emission of gamma radiation at rates $\gamma_t$ and $\gamma_0$, respectively. The equations of time-dependent perturbation theory are[25]

$$\begin{aligned} i\hbar\dot{a}_1&=a_0Ve^{+i\omega t}\exp[+(i/\hbar)(W_{1,1}-W_{1,0})t] -i\hbar(\tfrac{1}{2}\gamma_t)a_1,\\ i\hbar\dot{a}_0&=a_1V^*e^{-i\omega t}\exp[+(i/\hbar)(W_{1,0}-W_{1,1})t] -i\hbar(\tfrac{1}{2}\gamma_0)a_0, \end{aligned} \quad (19)$$

where

$$Ve^{+i\omega t}=(u_{1,1}|\mu_0 g(\mathbf{s}_- - \mathbf{s}_+)\cdot(-iH_1/4)(\hat{x}+i\hat{y})|\psi_{1,0})e^{+i\omega t}. \quad (20)$$

It will suffice for the purposes of this paper to use an approximate expression for $V$ which is obtained by using the value of $\psi_{1,0}$ given in Eq. (5) and obtained from first-order perturbation theory:

$$V\simeq+\frac{ix}{4\sqrt{2}[1+(\tfrac{1}{2}x)^2]^{\frac{1}{2}}}\mu_0 gH_1. \quad (21)$$

The notation used in Eqs. (19), (20), and (21) is defined in Secs. 2.1 and 2.2.

The decay of the state amplitudes due to the annihilation process is treated phenomenologically in Eqs. (19) by the introduction of damping terms. A complete treatment should include the description of the positronium annihilation into a final state consisting of photons. The phenomenological treatment of the annihilation process is similar to the treatment of the optically decaying $2p$ level in the theory of the Lamb shift measurement,[24] whose justification has been considered.[26] A detailed justification of the phenomenological treatment of the annihilation process in the positronium case has not been given. However, this treatment is believed adequate for the present application.

The general solutions of Eqs. (19) are:

$$\begin{aligned} a_1&=A_1\exp(-\delta_1-\tfrac{1}{2}\gamma_t)t+A_2\exp(-\delta_2-\tfrac{1}{2}\gamma_t)t,\\ a_0&=(-\hbar/iV)\{-\delta_1A_1\exp[(-\delta_1-\tfrac{1}{2}\gamma_t)t+i(\omega_{01}-\omega)t] -\delta_2A_2\exp[(-\delta_2-\tfrac{1}{2}\gamma_t)t+i(\omega_{01}-\omega)t]\}, \end{aligned} \quad (22)$$

where $A_1$ and $A_2$ are constants and

$$\delta_{1,2}=\frac{+i(\omega_{01}-\omega)+\tfrac{1}{2}(\gamma_0-\gamma_t)}{2} \pm\tfrac{1}{2}\{[i(\omega-\omega_{01})+\tfrac{1}{2}(\gamma_t-\gamma_0)]^2-4VV^*/\hbar^2\}^{\frac{1}{2}}, \quad (23)$$

and $\omega_{01}=(W_{1,0}-W_{1,1})/\hbar$.

Consider a positronium atom formed in the $M=+1$ state of orthopositronium. The initial conditions which determine the constants $A_1$ and $A_2$ are then:

$$a_1=1,\quad a_0=0,\quad \text{at}\quad t=0. \quad (24)$$

This gives

$$A_1+A_2=1,$$
$$\delta_1A_1+\delta_2A_2=0,$$

whence

$$A_1=\delta_2/(\delta_2-\delta_1),\quad A_2=-\delta_1/(\delta_2-\delta_1). \quad (25)$$

The fraction $P_1$ of these positronium atoms that decay by two-quantum annihilation is the quantity directly related to the experimental measurements:

$$P_1=\int_0^\infty dt|a_0|^2\gamma_{02}, \quad (26)$$

in which $\gamma_{02}$ is the two-quantum decay rate of the $M=0$ state of orthopositronium. By integration,

$$P_1=\gamma_{02}\frac{\hbar^2}{|V|^2}\frac{|\delta_1|^2|\delta_2|^2}{|\delta_2-\delta_1|^2}\left\{\frac{1}{\delta_1+\delta_1^*+\gamma_t}+\frac{1}{\delta_2+\delta_2^*+\gamma_t} -\frac{1}{\delta_1^*+\delta_2+\gamma_t}-\frac{1}{\delta_1+\delta_2^*+\gamma_t}\right\}. \quad (27)$$

This expression is exact provided only the two states $M=0$ and $M=+1$ of orthopositronium are considered

[25] W. Pauli, *Handbuch der Physik* (Verlag J. Springer, Berlin, 1933), second edition, Vol. 24, Part I, p. 158. The probability amplitudes $a_1$ and $a_0$ are defined similarly to the $a_n$'s in this reference.

[26] G. Wentzel, *Handbuch der Physik* (Verlag J. Springer, Berlin, 1933), second edition, Vol. 24, Part I, p. 752; G. Breit, Revs. Modern Phys. 4, 504 (1932); G. Breit and E. Teller, Astrophys. J. 91, 215 (1940).

and provided the nonresonant terms in Eq. (18) are neglected. The interaction with the microwave field has a matrix element connecting the $M=+1$ state of orthopositronium and the $M=0$ state of parapositronium, but this matrix element will contribute a nonresonant term to $P_1$ and can be neglected.

Consider next a positronium atom formed in the $M=0$ state of orthopositronium. The initial conditions are now

$$a_1=0, \quad a_0=1, \quad \text{at} \quad t=0. \tag{28}$$

The fraction $P_0$ of the annihilations of the $M=0$ state of orthopositronium that are two-quantum annihilations is found to be

$$P_0=\frac{\gamma_{02}}{|\delta_2-\delta_1|^2}\left\{\frac{|\delta_1|^2}{\delta_1+\delta_1^*+\gamma_t}+\frac{|\delta_2|^2}{\delta_2+\delta_2^*+\gamma_t}-\frac{\delta_1^*\delta_2}{\delta_1^*+\delta_2+\gamma_t}-\frac{\delta_1\delta_2^*}{\delta_1+\delta_2^*+\gamma_t}\right\}. \tag{29}$$

If only the two magnetic sublevels $M=-1$ and $M=0$ of orthopositronium are considered and if nonresonant terms are omitted, then expressions identical to Eqs. (27) and (29) are obtained for $P_{-1}$ and $P_0$, respectively.

Since the actual microwave magnetic field includes all the terms in Eq. (18), it is clear that all the three states of orthopositronium are coupled by interaction with the microwave field. Hence the expressions (27) and (29) obtained for $P_0$, $P_1$, and $P_{-1}$ by neglect of this actual coupling of the three states are incomplete. The application of Eqs. (27) and (29) will be for the case in which the microwave power is low, which corresponds to retaining only the lowest nonvanishing power of $V$. It is clear that the corrections to Eqs. (27) and (29) due to the coupling of the three states will involve higher powers of $V$ and hence can be neglected.

The quantity to be compared with experiment is the change, $P_T$, in the number of orthopositronium atoms which decay by two photon annihilation due to the microwave field divided by the number of $M=+1$ and $M=-1$ orthopositronium atoms formed:

$$P_T=\tfrac{1}{2}(P_1+P_{-1}+\Delta P_0), \tag{30}$$

in which $\Delta P_0=2(P_0-\gamma_{02}/\gamma_0)$ is the change in two-photon annihilation of the $M=0$ state due to the microwave field. The factor of 2 arises from the separate consideration of transitions between $M=0$ and $M=+1$ and between $M=0$ and $M=-1$ orthopositronium states in the approximation applicable to low microwave fields that the coupling of the three states can be neglected. This expression will be used only in the limit of low microwave power.

The principal effect of the nonresonant terms arising from the field of Eq. (18) is to produce a small shift in the energy levels of orthopositronium. This shift is of the order of $(H_1/H_0)^2$. For typical field values of $H_1=10$ gauss and $H_0=8000$ gauss, this shift is about 1 part in $10^6$ and hence is negligible.[27]

For further consideration of the line shape given by Eq. (30), the approximations applicable to the actual experimental situation can be employed. The microwave power level is sufficiently low so that an important simplification is possible. By reference to Eq. (23), it is seen that the expansion parameter is $(2|V|/\hbar)/[\frac{1}{2}(\gamma_0-\gamma_t)]$. In our case, with $H_1\simeq 10$ gauss and $H_0\simeq 8000$ gauss, this ratio is about 0.28. The error introduced in the expression for the resonance line will be of relative order $\frac{1}{2}\{(2|V|/\hbar)/[\frac{1}{2}(\gamma_0-\gamma_t)]\}^2$, which is about 0.04. An additional approximation will be made for the expression to be derived now, namely that $\gamma_t(=0.72\times10^7)\ll\gamma_0(=1.0\times10^8)$. The error introduced will be of relative order $\gamma_1/\gamma_0(=0.07)$. It should be noted that neither of these approximations alters the condition that the line shape will be symmetrlcal with respect to the variation of $\omega$ on either side of the resonance value $\omega_{01}$. If these approximations are employed in Eqs. (23), (27), (29), and (30), $P_T$ is found to be:

$$P_T=P_1=\gamma'/(\gamma_t+\gamma'), \tag{31}$$

in which

$$\gamma'=\frac{(\frac{1}{2}x)^2\gamma_s|V|^2}{\hbar^2[(\omega_{01}-\omega)^2+(x^2\gamma_s/8)^2]}. \tag{31a}$$

In deriving this expression, $\gamma_0$ is replaced by $(\frac{1}{2}x)^2\gamma_s$ in order to retain consistently the approximation $\gamma_t\ll\gamma_0$. The effect of power broadening of the resonance line is contained in Eq. (31). If the microwave power is sufficiently low so that $\gamma'\ll\gamma_t$, then $P_1$ becomes

$$P_1=\gamma'/\gamma_t. \tag{32}$$

This expression gives the non-power-broadened natural line shape.

A more exact expression for $P_T$ can be obtained by including the correction of relative order $\gamma_t/\gamma_0$. In this approximation $\Delta P_0$ contributes. Use of Eqs. (23), (27), (29), and (30), and retention of terms to the order $(\gamma_t/\gamma_0)^2$ in the power series expansion yield Eq. (12) of Sec. 2.4.

## APPENDIX II. EFFECT OF COLLISIONS

For the experiment reported in this paper, positronium atoms are formed in a gas having a pressure of about one atmosphere, which is required to slow down the positrons emitted at high energies from the radioactive source. Under these conditions the kinetic mean free time between collisions of a positronium atom with gas molecules is between $10^{-12}$ and $10^{-10}$ sec, which is short compared to the annihilation lifetime of orthopositronium. Hence it is important to consider the effect of collisions on line broadening and line shifts.

Since the transition studied is a magnetic dipole transition, the magnetic fields present during a collision

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

are of interest. The molecules of the gases used—A, $N_2$, and $SF_6$—have no electronic paramagnetism, and hence have a magnetic moment due only to the nuclei and to molecular rotation, which will be at most of the order of a nuclear magneton. At a distance of $a_0(\sim 0.5\times 10^{-8}$ cm) the magnetic field is about 50 gauss. The magnetic field resulting from the motion of the positronium atom with respect to the electric field of the gas molecule will be about 10 000 gauss at a distance of $a_0$ for a positronium atom with a velocity of $4\times 10^7$ cm/sec (which corresponds to a kinetic energy of 1 ev). The perturbation during a collision can be treated as a sudden perturbation[28] as regards transitions between the hyperfine levels of the ground state of positronium, because the collision time is short ($\sim 10^{-16}$ sec). A transition from a magnetic substate of orthopositronium to parapositronium, which is more probable than a transition between two magnetic substates of orthopositronium will have a probability of only about $10^{-9}$ per collision. Hence during the annihilation lifetime of $10^{-7}$ sec, in which some $10^4$ collisions occur, the transition probability is $10^{-7}$. This small value implies that there will be no appreciable line broadening due to these collisions.

The electronic diamagnetic moment induced in a gas molecule by the external magnetic field is considerably less than a nuclear magneton and hence has a negligible effect on the line broadening.

Although collisions with gas molecules do not contribute significantly to line broadening, a shift in the line center may occur in a manner similar to that discussed by Wittke and Dicke for their hydrogen hyperfine structure measurement.[29] The kinetic energies of the gas molecules and of positronium are too low to allow inelastic collisions which transfer positronium from the $1s$ to the $2p$ state. However, the strong electric fields due to the gas molecule which are present in a collision will cause a substantial perturbation of the positronium ground-state wave function which may be considered as primarily an admixture of $2p$ state. Owing to the high velocity of the positronium atoms and the correspondingly short collision time, the collision cannot be treated strictly as an adiabatic collision as was justified for the application of Wittke and Dicke, nor, on the other hand, is the sudden approximation applicable here. Because of the difficulty of the problem, only an order of magnitude theoretical result can be expected in a simple calculation, and the adiabatic treatment will be used. From the assumption that the positronium atom is entirely in a $2p$ state during a collision and from the observation that the hyperfine structure separations are considerably smaller in the $2p$ state than in the $1s$ state, it follows that a measurement of the ground-state hfs splitting which takes place over a time interval including many collisions will yield a value equal to the true hfs of positronium multiplied by the fraction of the time that the positronium atom is unperturbed by collisions. If the collision time is taken as $10^{-16}$ sec and the time between collisions as $10^{-11}$ sec, the measured hfs will differ from the true hfs of free positronium by 1 part in $10^5$. Because of the roughness of this approximate calculation the result should only be regarded as an indication that a measurement of the hfs splitting to such an order of magnitude requires an experimental study of the effect of the gas used and of its pressure on the hfs value determined. It has been pointed out in Sec. 5.1 that no systematic differences in $\Delta\nu$ were observed for the different gases and gas pressures used within the quoted error of 1 part in 5000. Line broadening due to these electric perturbations of positronium in collisions can only arise from statistical variations in the time average effect and is quite negligible.

Because of the ionization produced in slowing down the energetic positrons from the strong radioactive source, an electron-ion density of some $10^{10}/cm^3$ will be present in the cavity. This charge density is too small to influence appreciably either the line center or the line width.

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[28] L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1949), p. 207.

[29] J. P. Wittke and R. H. Dicke, Phys. Rev. **103**, 620 (1956).