CURRENT STATUS OF EXPERIMENTS ON AND THEORY OF POSITRONIUM*

Positronium[1] is the atom consisting of an electron and a positron. It is a light isotope of hydrogen as it contains one electron and one positively charged particle lighter than the proton.

Study of the properties and interactions of this simple atom is of interest from several viewpoints. It is composed of leptons only. Its *energy levels*—in particular, the fine-structure intervals—and its lifetimes and decay modes are of great significance for quantum electrodynamics, elementary particle physics, and atomic physics[2,3,4].

The *formation* of positronium and its *interactions with other atoms and molecules* are interesting topics in the fields of atomic collisions and chemistry[5,6,7,8,9]

The *behavior* of positronium in *liquids* and *solids* is a large field of study important to the physics and chemistry of condensed matter, including low-temperature phenomena[7,10,11,12].

What Has Hitherto Been Measured

Until recently, the only *energy intervals* that had been measured were the fine-structure interval and Zeeman effect in the ground state of positronium. Within the last year, observation of the first excited state of positronium has been both claimed and disputed (see Subsequent Developments). The *annihilation rates* of both the 1^1S_0 and 1^3S_1 ground state of positronium have also been measured.

The methods used for studying this unstable atom have relied on its annihilation or decay characteristics. The energy interval measurements are based on absorption or stimulated emission at microwave frequencies, and they require first, an initial unequal population of the two states involved in the transition and second, a means for detecting an induced change in the state populations. Table 1 lists the annihilation characteristics of the ground state of positronium. The different lifetimes of orthopositronium ($^3S_1$ state) and of parapositronium ($^1S_0$ state) result in unequal state populations, and their different annihilation characteristics provide the means for observing an induced change in state populations.

Table 1
Positronium Annihilation

	Parapositronium ($^1S_0$)	Orthopositronium ($^3S_1$)
Lifetime (Mean)	1.25×10^{-10} sec	1.375×10^{-7} sec
Mode	Two γ-Rays E_γ = 511 KeV 180° Emission	Three γ-Rays E_γ(max) = 511 KeV; Continuous Coplanar Emission

* From: Hughes, Vernon, *Positronium and Muonium*, *Physik 1973*, German Physical Society Conference (Physik Verlag GmbH, Germany, 1973) pp. 123-155 (in English).

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4. Hughes, V. W., *Atomic Physics III*, edited by S. J. Smith and G. K. Walters (Plenum Press, New York, 1973), p. 1.
5. Hughes, V. W., *J. Appl. Phys.* 28, 16 (1957).
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12. Brandt, W. and Paulin, R., *Phys. Rev. B* 5, 2430 (1972).

Positronium Fine Structure

Breit Interaction

(a)

Pair Annihilation

(b)

$$\Delta W(1\ ^3S_1 - 1\ ^1S_0) = \left(\frac{4}{6} + \frac{3}{6}\right)\alpha^2\ \mathrm{Ry}$$

Fig. 1

Lowest order Feynman diagrams and energy separation for ground state positronium fine structure.

13. Bethe, H. A., and Salpeter, E. E., *Quantum Mechanics of One- and Two-Electron Atoms* (Springer-Verlag, Berlin, 1957).
14. Fulton, T., Owen, D. A., and Repko, W. W., *Phys. Rev.* A 4, 1802 (1971).
15. Owen, D. A., *Phys. Rev. Letters* 30, 887 (1973).

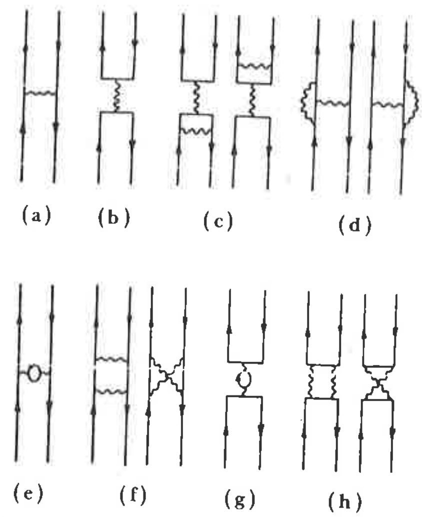
Positronium and Quantum Electrodynamics

From the viewpoint of quantum electrodynamics, positronium is an ideal atom since it is the bound state of an electron and its antiparticle the positron. I shall review the current status of our knowledge of the fine-structure interval in the ground state and of the annihilation rates in the ground state.

The energy levels of positronium can be calculated from the Bethe, Salpeter (or Gell-Mann, Low) equation[13] as a series expansion in the fine-structure constant α. Figure 1 shows the lowest order Feynman diagrams which contribute to the fine-structure interval in the ground state of positronium. Diagram (a) is a single photon exchange which gives rise to the spin-spin interaction and contributes the amount $4/6\ \alpha^2$ Ry to the fine-structure interval. Diagram (b) also involves a single photon but represents the virtual annihilation of the electron and the positron and contributes the amount $3/6\ \alpha^2$ Ry to the interval. Figure 2 shows the Feynman diagrams which involve two virtual photons and contribute to the order α^3Ry. Theoretical calculations have been carried through the order $\alpha^4\ ln\alpha$Ry, and the present complete theoretical expression for $\Delta\nu$ is given in Table 2[14, 15] p. 115.

Fig. 2. *Feynman diagrams for α^2Ry and α^3Ry energy separation terms for ground-state positronium fine structure.* (From R. Karplus and A. Klein, *Phys. Rev.* 87, 848 (1952).)

(a). *One-quantum exchange.*
(b). *(^3S only) One-quantum annihilation.*
(c). *(^3S only) One-quantum annihilation and exchange.*
(d). *Self-energy and magnetic moment corrections.*
(e). *Vacuum polarization.*
(f). *Two-quantum exchange.*
(g). *(^3S only) One-quantum annihilation plus vacuum polarization.*
(h). *(^1S only) Two-quantum annihilation.*



Zeeman Energy Levels

The Zeeman energy levels of the ground state of positronium are shown in Figure 3. At zero magnetic field there are two energy levels separated by the fine-structure interval $\Delta\nu$ of about 200 GHz.

The M=±1 magnetic sublevels of orthopositronium are degenerate and independent of H; the M=0 magnetic sublevel of orthopositronium has a leading term in H which is proportional to H^2. The Zeeman transition indicated by the arrow has been studied. Its frequency is given by:

$$f_{01} = \frac{\Delta\nu}{2}\left[(1+x^2)^{1/2} - 1\right] \qquad (1)$$

in which $x = 2\mu_B^e g' H/(h\Delta\nu)$ where μ_B^e is the electron Bohr magneton, g' is the electron g-value in positronium, and H is the external magnetic field.

The principle of the experiment involves inducing the Zeeman transition with microwave power and detecting the occurrence of the transition through the change in character of the annihilation γ-rays[16, 17, 18]. This change occurs because in the magnetic field of about 8000 G, the M=±1 orthopositronium state decays by three γ-ray annihilation with a mean life of 1.4×10^{-7} sec, whereas the M=0 orthopositronium state is an admixture of $^3S_1$ and $^1S_0$ states and annihilates predominantly into two γ-rays with the shorter mean life of 10^{-8} sec.

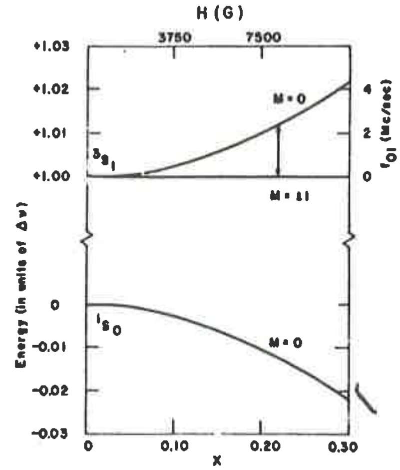
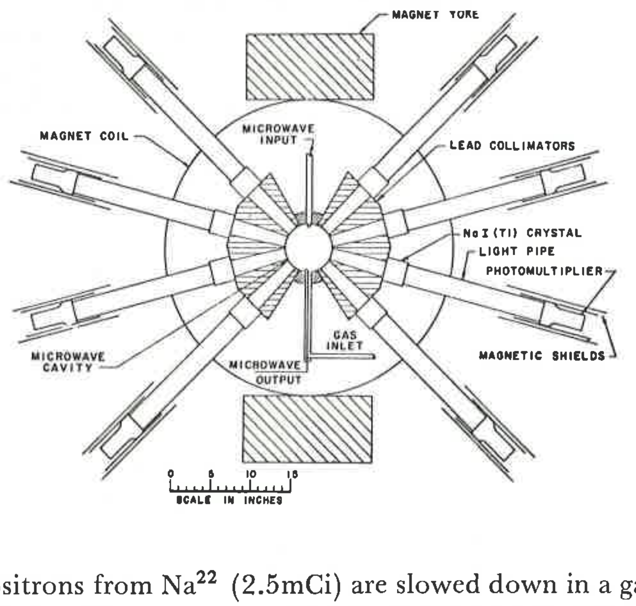


Fig. 3 ↑

Zeeman energy levels of positronium in its ground n=1 state. The transition observed is indicated by the arrow. *See also p. 85.*

Most Recent Measurement of Δν

The experimental arrangement for the most recent precision determination[19] of Δν is shown in Figure 4. Positronium is formed



← Fig. 4

Schematic diagram of experimental apparatus for positronium fine-structure measurement.

when positrons from Na^{22} (2.5mCi) are slowed down in a gas (nitrogen or argon). A uniform magnetic field is provided by an electromagnet with a 38 cm pole diameter. Microwave power of about 100 watts is

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17. Hughes, V. W., Marder, S., and Wu, C. S., *Phys. Rev.* **106**, 934 (1957).
18. Theriot, Jr., E. D., Beers, R. H., Hughes, V. W., and Ziock, K. O. H., *Phys. Rev.* A 2, 707 (1970).
19. Carlson, E. R., Hughes, V. W., Lewis, M. L., and Lindgren, I., *Phys. Rev. Letters* **29**, 1059 (1972).

mCi = milli-curie

1 Ci = 3.7×10^{10} sec^{-1}

supplied to the high Q microwave cavity operating in the TM110 mode. Eight scintillation counters are used as four counter pairs to observe the two 0.5 MeV annihilation γ-rays in coincidence. The coincidence counting rate is observed as a function of magnetic field for a fixed microwave frequency. Figure 5 shows a resonance

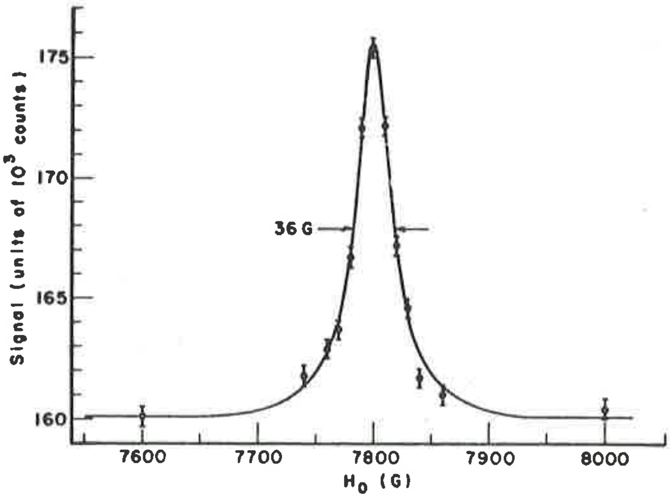

Fig. 5→

Resonance curve for positronium Zeeman transition. The microwave frequency was about 2300 MHz, and the N_2 gas pressure was 0.3 atm.

curve for the Zeeman transition. A linear background has been subtracted before plotting the experimental points. The solid curve is the theoretical lineshape (approximately a Lorentzian) fit to the data points. The linewidth (full width at half maximum) is about three parts in 10^3 and is due to the natural width associated with annihilation and microwave power broadening. From the observed resonance values of microwave frequency and magnetic field together with the Breit-Rabi formula, Eq. (1), we obtain a value for the fine-structure interval $\Delta\nu$.

The observed values of $\Delta\nu$ versus the gas pressure for argon[18] and nitrogen[19] are shown in Figures 6 and 7. The dependence of $\Delta\nu$

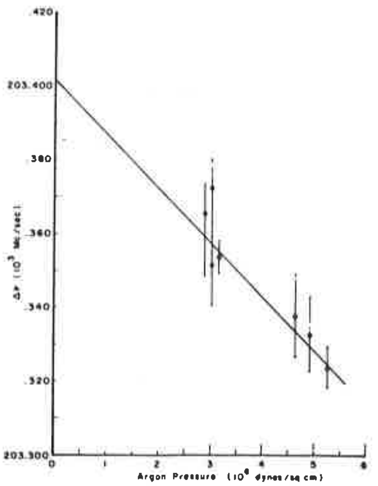

Fig. 6↑

Measured values of $\Delta\nu$ versus argon pressure. Solid curve is a straight line fit to the data.

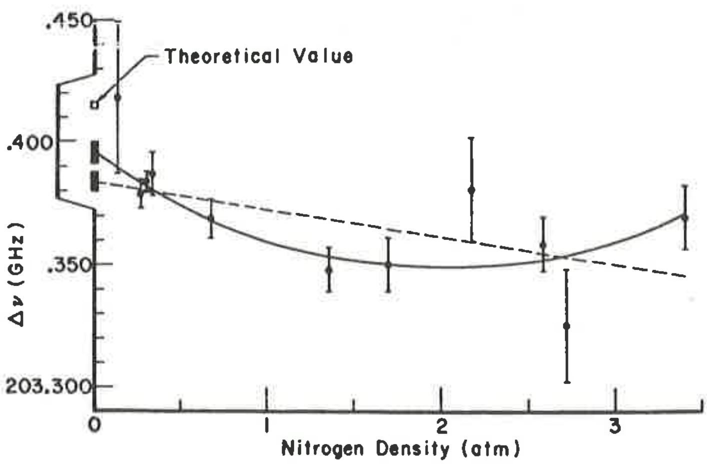

Fig. 7→

Plot of $\Delta\nu$ versus N_2 gas density in units of atmospheres at 23°C. The solid and dashed lines are the quadratic and linear fits, respectively. The solid bars show one standard deviation errors for the fitted values $\Delta\nu(0)$.

on pressure is due to the distortion of positronium in collisions with nitrogen. Fits to the experimental points of $\Delta\nu$ versus pressure indicate that not only a term proportional to pressure P but also a term proportional to P^2 is required. Extrapolation to zero pressure yields $\Delta\nu$ for free positronium. Both the linear and the quadratic fractional $\Delta\nu$ pressure shift terms $[(1/\Delta\nu)\ (\partial\Delta\nu/\partial P)]$ for positronium in argon and nitrogen are more than an order of magnitude larger than the corresponding values measured for muonium in argon and krypton[20, 21]. This comparison provides an example of the great difference between positronium and muonium or hydrogen collisions.

Table 2 gives the latest experimental and theoretical results

Table 2
Positronium Fine Structure

$$\Delta\nu\ (1^3S_1 - 1^1S_0)$$

$$\Delta\nu_{expt} = (203.403 \pm 0.012)\text{GHz (60 ppm)}$$

$$\Delta\nu_{expt} = (203.396 \pm 0.005)\text{GHz (25 ppm)}$$

$$\Delta\nu_{theor} = \frac{1}{2}\alpha^2 cRy\left[\frac{7}{3} - \frac{\alpha}{\pi}\left(\frac{32}{9} + 2ln2\right) - \frac{3}{2}\alpha^2 ln\alpha + \frac{\alpha^2}{2} ln\alpha\right]$$

$$= (203.404 \pm 0.000\ 7)\text{GHz (3.4 ppm)}$$

$$\Delta\nu_{expt} - \Delta\nu_{theor} = (-0.008 \pm 0.005)\text{GHz (25 ppm)}$$

$$\alpha^{-1} = 137.036\ 02 \pm 0.000\ 21\ \text{(1.5 ppm)}$$

for $\Delta\nu$. The accuracy of the experimental value is limited principally by the statistical counting error, and the one standard deviation error is 25 parts per million (ppm). The error indicated in the theoretical value is that which arises from the uncertainty in the fundamental atomic constants[22]—principally α—and does not include any estimate of the magnitude of uncalculated higher order radiative correction terms. The experimental and theoretical values agree within 1.6 standard deviations of the experimental error. This agreement constitutes perhaps the best test of the validity of the Bethe-Salpeter equation for the bound state of two leptons, since characteristic relativistic and recoil terms are important for positronium.

A more adequate comparison of the experimental value with theory really requires the calculation of the α^4 Ry term which can be of the order of 50 ppm. The calculation of this term is a formidable problem, but several groups are working on it employing modern approaches and techniques for evaluating radiative contributions to atomic energy levels[23]. At Yale, our experiment is continuing to

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21. Favart, D., McIntyre, P. M., Stowell, D. Y., Telegdi, V. L., DeVoe, R., and Swanson, R. A., *Phys. Rev. Letters* 27, 1340 (1971).
22. Taylor, B. N., Parker, W. H., and Langenberg, D. N., *Rev. Mod. Phys.* 41, 375 (1969).
23. Kroll, N. M., *Atomic Physics III*, edited by S. J. Smith and G. K. Walters (Plenum Press, New York, 1973), p. 33.

improve the accuracy in the determination of $\Delta\nu$ to 10 ppm or better through improved statistics with a stronger (10mCi) Na^{22} source and through the on-line use of a PDP-11-40 computer for control, stabilization and data accumulation in the experiment. Further improvements in the future may be possible in the determination of $\Delta\nu$ by the method of separated oscillating fields that yields resonance lines narrower than the natural linewidth associated with the annihilation rates of positronium[24, 25].

24. Ramsey, N. F., *Molecular Beams* (Clarendon Press, Oxford, 1956).
25. Hughes, V. W., *Quantum Electronics*, edited by C. H. Townes (Columbia University, New York, 1960), p. 582.

Annihilation Rates

The annihilation rates of both the $^1S_0$ and $^3S_1$ ground states have been measured. The parapositronium annihilation rate has been obtained from observations of the linewidths of the Zeeman transition discussed above versus the microwave power[18]. Figure 8 plots

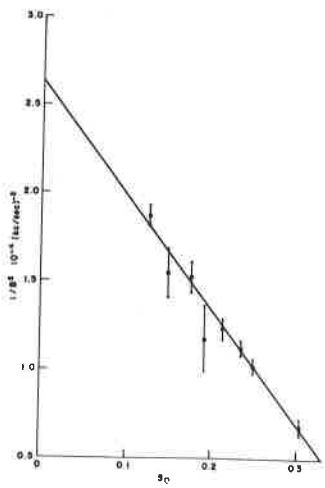

Fig. 8

Measured values of the inverse square of the linewidth $1/B^2$ versus signal height S_o for the positronium Zeeman transition. The solid curve is a straight line fit to the data points.

data of the inverse square of the linewidth *versus* signal height, and the solid line is the theoretical fitted line. The extrapolation to zero signal height yields the natural linewidth determined principally by the rate of the two γ-ray annihilation of the M=0 state. A value of the annihilation rate γ_p of $^1S_0$ parapositronium at zero magnetic field can be readily calculated from the observed natural linewidth of the resonance line and is given in Table 3.

The $^3S_1$ orthopositronium annihilation rate has been measured in straightforward timing experiments[26, 27]. The experiment involves forming and measuring the lifetime of orthopositronium in a gas. Since the orthopositronium lifetime is reduced due to collisions in the gas, an extrapolation to zero pressure is required to obtain the annihilation rate of free orthopositronium.

Table 3 gives the experimental and theoretical results. The

Table 3

Annihilation Rates of Ground State Positronium

(1 std. dev.)

	Parapositronium ($^1S_0$) (10^{10} sec^{-1})	Orthopositronium ($^3S_1$) (10^7 sec^{-1})
Experimental	0.799 ± 0.011	0.727 5 ± 0.001 5
Theoretical	0.798 54	0.721 19
$\gamma_{expt} - \gamma_{theor}$	+0.000 46 ± 0.011	+0.005 9 ± 0.001 5

$$\gamma(^1S_0)_{theor} = \frac{\pi\alpha^5 mc^2}{h}\left[1 - \frac{\alpha}{\pi}\left(5 - \frac{\pi^2}{4}\right)\right]$$

$$\gamma(^3S_1)_{theor} = \frac{4\alpha^6 mc^2}{9h}(\pi^2 - 9)$$

$$\alpha^{-1} = 137.036\,02 \pm 0.000\,21 \ (\pm 1.5 \text{ ppm})$$

parapositronium annihilation rate has been measured with an accuracy of 1.4% and the orthopositronium annihilation rate with an accuracy of 0.2%. The theoretical expression for the annihilation rate of parapositronium into two γ-rays includes the $\alpha^6 mc^2/h$ correction to the leading term, which arises from a Coulomb correction and a radiative correction[28]. For the three γ-ray annihilation only the $\alpha^6 mc^2/h$ leading term has been computed[29], although several Feynman diagrams contributing to the order of $\alpha^7 mc^2/h$ have been evaluated[30]. The agreement between the experimental and theoretical values for parapositronium is satisfactory. For orthopositronium the difference between the experimental and theoretical values is four standard deviations of the experimental error. Since the term of next order in α in the theoretical expression is of the order of magnitude of this difference, it is important that this difficult calculation

26. Beers, R. H., and Hughes, V. W., *Bull. Am. Phys. Soc.* 13, 633 (1968). The value given in Table 3 is an unpublished, revised value, which will be published soon.
27. Coleman, P. G., Griffith, T. C., and Heyland, G. R., *Journal of Physics* E 5, 376 (1972); private communication from T. C. Griffith.
28. Harris, I., and Brown, L. M., *Phys. Rev.* 105, 1656 (1957).
29. Ore, A., and Powell, J. L., *Phys. Rev.* 75, 1696 (1949).
30. Holt, J. M., "Radiative Corrections to the Decay of Orthopositronium," Ph.D. thesis, Yale (1970).

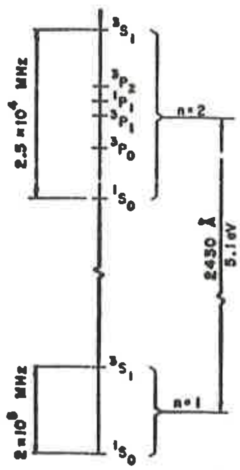

Fig. 9

Positronium energy levels, showing the fine structure levels of the n=2 state.

be done. It is interesting to note that this uncalculated term includes a Feynman diagram involving photon-photon scattering, in which one of the four photons is virtual. Photon-photon scattering has always posed a challenge to experiment[31], and the orthopositronium annihilation rate may be a measurable quantity to which photon-photon scattering makes a significant contribution.

Significance of the First Excited State of Positronium

Measurements on the excited n=2 state of positronium, particularly its fine structure and Lamb shift intervals, are of great interest for quantum electrodynamics. The energy level scheme is shown in Figs. 9, 10 and 11, including its Zeeman effect and motional Stark effect[32, 33].

Various unsuccessful searches have been made to observe the optical radiation of 2430 Å from the 2P→1S transition in emission[34, 35, 36, 37]. Following a pioneering unpublished experiment by Kendall[38], experiments are in progress at Bell Telephone Laboratories and at Yale University to try to populate the n=2 state by optical excitation from the 1S state. The Yale group has recently

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34. Bennett, W. R., Thomas, W., Hughes, V. W., and Wu, C. S., *Bull. Am. Phys. Soc.* **6**, 49 (1961); Hughes, V. W., *J. Appl. Phys.* **28**, 16 (1957).
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36. Fagg, L. W., *Nucl. Instr. Methods* **85**, 53 (1970).
37. Leventhal, M., *Proceedings of the National Academy of Sciences* **66**, 6 (1970).
38. Kendall, H. W., "The First Excited State of Positronium," Ph.D. thesis, M.I.T. (1954). Unpublished.

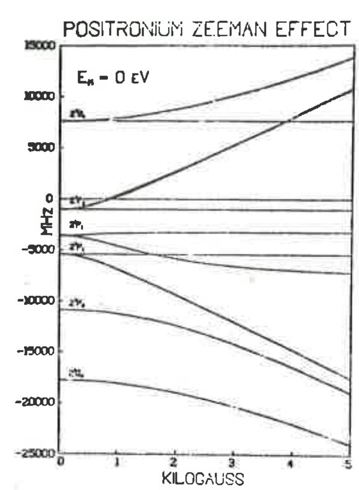

Fig. 10

Fine-structure energy levels in the n=2 state of positronium as a function of magnetic field with kinetic energy T=0 e V.

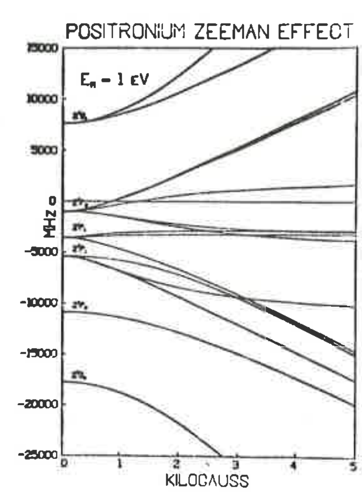

Fig. 11

Fine-structure energy levels in the n=2 state of positronium as a function of magnetic field with kinetic energy T=1 e V. (The maximum motional Stark effect in the plane perpendicular to the magnetic field is given.)

reported[39] evidence for the formation of the n=2 state using a white light source and also a tin arc. The experimental arrangement is shown in Figure 12. The signal is a change in the two γ-ray coincident rate when a broad band optical filter transmitting from 2000 Å to 3000 Å is inserted. The observed signal was 0.15% (±0.03%), whereas the predicted signal was about 0.1%. Higher light intensity and better wavelength selection are needed to confirm this result.

Spectroscopic measurements on the n=2 state will be very difficult because of the large perturbations and quenching of the n=2 levels by an external magnetic field and in collisions with the stopping gas. Theoretical values for the fine-structure intervals of the n=2 state have been calculated to the order of $\alpha^3 Ry$[40].

As a final topic for positronium, I would like to remark that recently, for the first time, the Zeeman transition $M=\pm1 \rightarrow M=0$ for orthopositronium was observed in a solid[41]. Fig. 13 shows a resonance curve observed in γ-Al_2O_3. The resonance has also been

39. Varghese, S. L., Ensberg, E. S., Hughes, V. W., and Lindgren, I., *Bull. Am. Phys. Soc.* **18**, 1503 (1973); *Phys. Lett.* **49**, 415 (1974).
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41. Judd, D. J., Lee, Y. K., Madansky, L., Carlson, E. R., Hughes, V. W., and Zundell, B., *Phys. Rev. Letters* **30**, 202 (1973).
42. Egan, P., Carlson, E. R., Hughes, V. W., Mourino, M., Varghese, S. L., Leventhal, M., and zuPutlitz, G., *Bull. Am. Phys. Soc.* **18**, 1503 (1973).

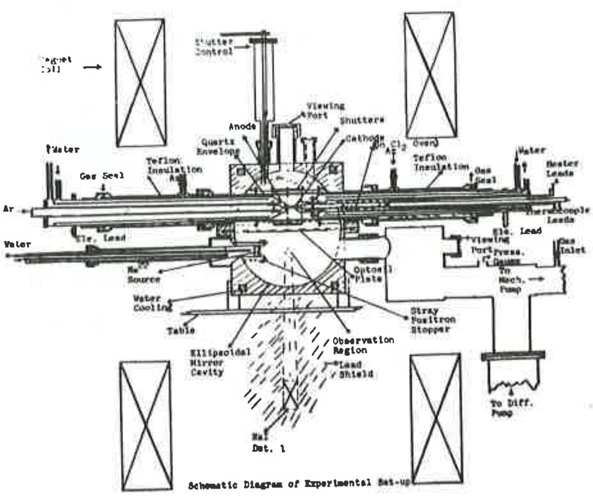

Fig. 12

Schematic diagram of experimental arrangement used at Yale to try to produce the n=2 state of positronium by optical excitation from the n=1 state.

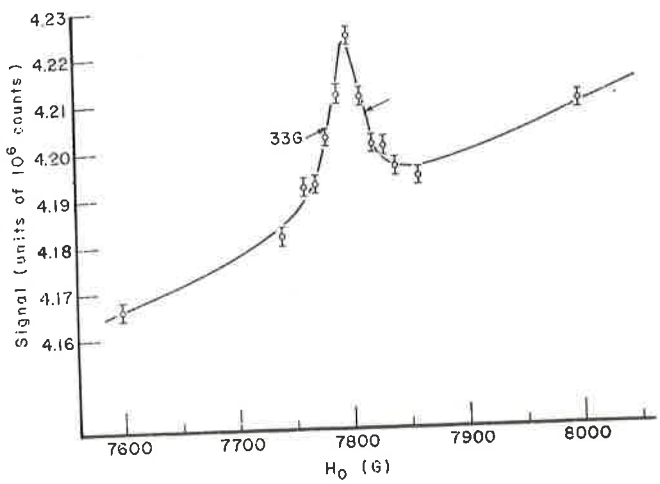

Fig. 13.

Positronium Zeeman transition resonance for positrons stopped in γ-Al_2O_3 powder pellet. Plot shows coincidence counting rate versus magnetic field. Data were taken with a microwave frequency of 2323.180 MHz and an input power of 52 W.

found in SiO_2 powder but not in fused quartz[42]. The resonance frequencies appear to be shifted to higher values by between 440 ppm and 2000 ppm compared to free positronium. These two powder materials—γ-Al_2O_3 and SiO_2—are porous on the microscopic scale and positronium may exist in the interstitial regions.

Vernon Hughes