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Fine-Structure Measurement in the First Excited State of Positronium*

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The 2^3S_1 - 2^3P_2 energy splitting of positronium has been observed by inducing rf transitions in a weak magnetic field (≈ 50 G) in the absence of a buffer gas and observing a resonant increase in the Lyman- α emission. We obtain 8628.4 ± 2.8 MHz, which is within 2 standard deviations of the theoretical value computed to order α^3R by Fulton and Martin.

In this Letter we report the first measurement of a fine-structure interval in the first excited state of positronium (Ps), based on our recent observation¹ of the Lyman- α radiation of Ps. The importance of the experimental determination of the $n=2$ Ps fine structure, which includes Lamb-shift-like terms, is well known²; this measurement represents the first experimental test of quantum electrodynamics applied to an excited state of a purely leptonic atom.

In contrast to hydrogen, the positronium $2S$ and $2P$ levels are nondegenerate already in order α^2R . The fine structure of these levels has been computed through order α^3R by Fulton and Martin,³ using the Bethe-Salpeter equation. From the several accessible fine-structure intervals we chose to study the 2^3S_1 - 2^3P_2 splitting, since it was predicted to lie in the X band and it involves the long-lived 2^3S_1 state (10^{-6} sec for 3γ annihilations—see term diagram, Fig. 2). Fulton and Martin obtain theoretically

$$\begin{aligned} \Delta\nu(2^3S_1-2^3P_2) &= \frac{23}{480} \alpha^2R (1 + 3.766\alpha) \\ &= 8625.14 \text{ MHz}, \end{aligned} \quad (1)$$

and estimate that the neglected terms of order α^4R may amount to several MHz. Present uncertainty in α produces only a 3-ppm uncertainty in

$\Delta\nu$.

Our measurement is based on the observation of an enhanced Lyman- α emission rate when $2^3S_1 \rightarrow 2^3P_2$ transitions are induced by an rf electric field at the proper frequency. The experiment is made possible by the production of Ps in vacuum, with use of a low-energy positron beam, at essentially zero field ($B \approx 50$ G), thus circumventing the problems associated with gas collisions and minimizing Zeeman splitting and motional Stark shifts.⁴

A beam of slow positrons (≈ 30 eV) is produced in an apparatus previously described in connection with the Lyman- α experiment,¹ and is guided into a cylindrical rf cavity as shown in Fig. 1. One side of the cavity has been replaced by parallel wires to allow the Lyman- α photons (2430 Å) to be detected by a uv-sensitive solar-blind photomultiplier (Amperex 56SBUVP, dark rate < 2 sec⁻¹). A semicylindrical, back-surface-aluminized, Suprasil-quartz mirror is used inside the cavity to improve the photon collection efficiency. The slow positrons collide with the Cu end face of the cavity and form Ps atoms leaving the surface, as reported earlier.⁵ Approximately 10^{-3} to 10^{-4} of the incident positrons form Ps in the $n=2$ state leading to the $2P \rightarrow 1S$ 2430-Å photon emission (optical decay rate Γ_{2P}

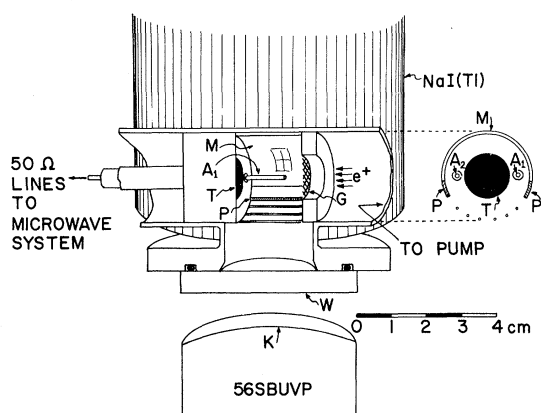


FIG. 1. Positron target chamber and microwave cavity. *G*, 95%-transmission tungsten grid; *T*, copper target; *M*, aluminized Suprasil quartz mirror; *W*, Suprasil quartz window; *K*, CsTe photocathode; *P*, support posts; *A*₁, input antenna; *A*₂, output antenna; NaI(Tl), annihilation detector coupled to an RCA 8575 phototube. Another NaI(Tl) detector (not shown) is placed above the chamber directly over the detector shown.

$= 3.14 \times 10^8 \text{ sec}^{-1}$), with the subsequent annihilation of the ground state ($1^1S_0 \rightarrow 2\gamma$, $1^3S_1 \rightarrow 3\gamma$). Two 7.62-cm-diam by 7.62-cm-long NaI(Tl) detectors placed above and below the target chamber are used to count the annihilation γ 's. As in Ref. 1, the time-delay spectrum between the Lyman- α photons and the subsequent annihilation γ 's is recorded by a time-to-amplitude converter and a multichannel analyzer (MCA), thus registering $2P \rightarrow 1S \rightarrow 2\gamma$ or $2P \rightarrow 1S \rightarrow 3\gamma$ events.

The rf electric field in the TM_{010} microwave cavity (center frequency 8860 MHz, $Q \approx 70$) is parallel to the incident slow-positron-beam velocity. X-band microwave power from a free-running Gunn-diode oscillator is fed into the cavity by antenna *A*₁ (Fig. 1). The rf electric field in the cavity is monitored by a small output probe, *A*₂, coupled to a cavity frequency meter and a thermistor power meter. The coupling to the input antenna is adjusted at each frequency for zero reflection, by use of a double-stub tuner. A coaxial switch is used either to turn the microwave power on and off or to switch between two Gunn oscillators operating at different frequencies. The input frequencies are measured and monitored by a heterodyne frequency meter (HP5245L/5255A), coupled via a 20-dB directional coupler

between the coaxial switch and the cavity input connector.

The Lyman- α -annihilation- γ coincidence rate reported in Ref. 1 has been increased some 200-fold, by mounting the uv photomultiplier directly in view of the target area with no filter or lens, by increasing the solid angle subtended by the NaI(Tl) detectors, and by using a stronger ^{58}Co fast-positron source ($\approx 500 \text{ mCi}$).

Data were obtained by switching every 1000 sec either the rf power (on and off) or the rf frequency ($f + \Delta$ and $f - \Delta$, with $\Delta = 30 \text{ MHz}$), while recording the Lyman- α -annihilation- γ time-delay spectrum in two halves of the MCA. At each frequency the input power level was adjusted to keep the transmitted power constant. All relevant counting rates as well as delayed coincidences were monitored continuously by a set of scalers and were recorded on paper tape. Typical single rates were $\approx 16\,000 \text{ sec}^{-1}$ for the NaI(Tl) counters and $\approx 8.3 \text{ sec}^{-1}$ for the uv phototube. The number of delayed coincidences (from 8 to 291 nsec following the prompt peak) was $\approx 0.18 \text{ sec}^{-1}$. The frequency f was changed usually after ≈ 40 pairs of counting periods (once every 24 h).

A typical time spectrum with the rf field off obtained during a 24-h run (12 h rf off) consists of a prompt peak with $\approx 56\,000$ counts and a delayed part containing ≈ 6000 counts separable into two components⁶: a long component which can be fitted by an exponential decay ($\tau \approx 120 \text{ nsec}$, $I \approx 60\%$) and a complex short component (average $\tau \approx 25 \text{ nsec}$, $I \approx 40\%$). As in Ref. 1 we interpret the large prompt component as due mainly to γ -ray-induced counts in the uv tube, and the delayed components as the measure of the 1^3S_1 annihilation following Lyman- α emission. We attribute the complex nature of the delayed components to the effects of wall collisions. With the rf field on an increase in the time-delayed coincidences is observed which follows a resonance curve; we interpret this effect as due to rf-induced $2S \rightarrow 2P$ transitions.

In Fig. 2 we plot the "microwave Lyman- α signal" defined as $S(f) = [N_{\text{on}}(f) - N_{\text{off}}] / N_{\text{off}}$, where N_{on} (N_{off}) is the counting rate in the time-delayed component of the uv-photon-annihilation- γ time spectrum, with the rf field on (off). The delayed rate was obtained from the MCA by summing all channels from 8 to 291 nsec after the prompt peak. The data were taken at a constant power reading of 0.41 mW. We also plot

$$S'(f) = [N(f + \Delta) - N(f - \Delta)] / [N(f + \Delta) + N(f - \Delta)] = [S(f + \Delta) - S(f - \Delta)] / [2 + S(f + \Delta) + S(f - \Delta)], \quad (2)$$

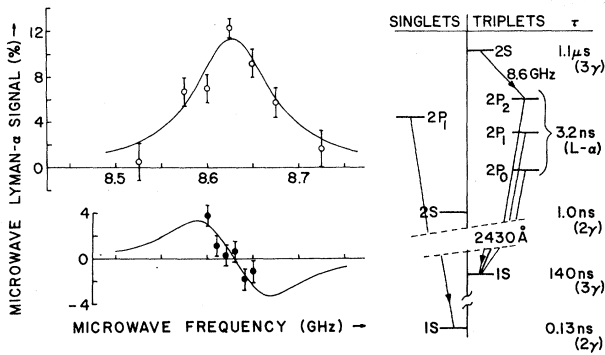


FIG. 2. The observed Lyman- α signal S (open circles) and logarithmic first-difference signal S' (solid circles) as a function of microwave frequency [see Eq. (2)]. The inset is the schematic term diagram for the $n=2$ and $n=1$ Ps states indicating the relevant transitions and the lifetimes τ for each level.

obtained in separate runs with $\Delta = 30$ MHz. A Lorentzian line shape $S(f) = \frac{1}{4}A\delta^2[(f - f_0)^2 + \frac{1}{4}\delta^2]^{-1}$ was maximum-likelihood fitted simultaneously to both sets of data by use of Eq. (2). The smooth curves in Fig. 2 are the results of the fit, yielding $A = (11.4 \pm 0.6)\%$, $f_0 = 8628.4 \pm 2.8$ MHz, and $\delta = 102 \pm 12$ MHz with $\chi^2/\nu = 12.1/10$.

The width δ is twice the ≈ 50 -MHz natural linewidth ($\Gamma_{2P}/2\pi$) of the 2^3P_2 state; we attribute this to power broadening and, to a smaller extent, to the effects of Ps collisions with the cavity walls. (The theoretical effect on δ of the 10^6 -sec $^{-1}$ annihilation rate of the 2^3S_1 state is small.) If we assume that all substates of the $n=2$ level are equally populated during production, and neglect possible geometrical differences in the detection efficiency between the originally produced $2P$ decays and the $2S \rightarrow 2P$ induced Lyman- α decays, we predict a maximum effect of 33% for $S(f_0)$. Subtracting the background due to accidentals in the time spectrum from $N(f)$ results in an observed effect of $A = (14.1 \pm 1.0)\%$. The change of A with power and the power broadening to the width agree with an order-of-magnitude computation which includes the natural decay rates, if an additional decay rate of $\approx 6 \times 10^7$ sec $^{-1}$ (possibly accounted for by wall collisions) is assumed for the $2S$ state. The largest uncertainty in this computation is the estimate of the electric field at the center of the cavity from the transmitted-power data, given the complex geometry of the cavity. The details of Ps-wall collisions are too little understood at present for a more detailed model computation. These uncertainties, how-

ever, influence mainly the width and the magnitude of $S(f)$, and would affect f_0 , the resonance frequency, only via a possible asymmetry of the actual line shape, an effect subject to further more precise experimentation.

To check further various parameters that might influence our results we have performed the following tests:

(1) We have changed the geometry of the wires on the side of the cavity and appreciably altered the resonance shape of the cavity; data taken at 8525, 8625, and 8725 MHz in this geometry are consistent with Fig. 2.

(2) We have changed the resonance frequency of the cavity to ≈ 8625 MHz ($Q \approx 140$) by removing the mirror, the cavity now consisting of a wire cage. A five-point resonance curve at 0.41 mW gives results consistent with the data of Fig. 2 (error bars approximately twice as large).

(3) In the above experiment with no mirror, the interior of the target chamber was smoked with MgO.⁷ The delayed-time spectrum shows a clear decrease in the short-lifetime intensity when compared to the time spectrum obtained without MgO. This effect supports the explanation of the complex decay of the delayed components in terms of wall collisions. In all our measurements, the difference time spectrum [(rf on) - (rf off)] also exhibits a $\approx 30\%$ short-lived component (≈ 10 nsec) and a ≈ 120 -nsec long component. Experiments are in progress to study in more detail the nature of these complex decay schemes.

(4) We tested the effect of rf power by measuring $S(8625$ MHz) at 0.13, 0.41, and 2.0 mW and obtained $(5.3 \pm 1.1)\%$, $(12.3 \pm 0.8)\%$, and $(16.3 \pm 1.1)\%$, respectively. At 2.0 mW three additional points taken at 8525, 8725, and 8860 MHz indicated a further broadening of the resonance. The last point was $S(8860) = (2.7 \pm 2.1)\%$. Since this datum point was taken at the resonance frequency of the cavity, its small value compared to $S(8625) = (16.3 \pm 1.1)\%$ at this power is further evidence that there are no spurious effects associated with the rf response of the cavity.

(5) Most of the experiments were performed with no uv filter in front of the uv photomultiplier tube, to improve counting rates. However, to confirm that the observed rf effect is indeed due to rf-induced Lyman- α photons, an interference filter (2430- \AA peak wavelength, ≈ 200 - \AA full width at half-maximum, and $\approx 25\%$ transmission) was used in one set of runs. The rf effect observed was $S(8625$ MHz) = $(12.6 \pm 2.8)\%$ at 1.4 mW,

in statistical agreement with the no-filter result at this power.

An rf effect $A = (0.68 \pm 0.09)\%$ at 0.41 mW is also observed in the singles rate of the uv photomultiplier. The small size of the effect is due to the high background of γ -induced counts in the uv photomultiplier and is in agreement with an estimate based on the observed delayed-coincidence rf effect.⁸ The NaI(Tl) singles-rate rf signal on the other hand was less than 0.01%. The large rf effect on the delayed coincidences coupled with the small singles-rate effect strongly supports our interpretation of the observed resonance.

The center frequency $f_0 = 8628.4 \pm 2.8$ MHz obtained from the data of Fig. 2 is to be compared with the predicted value given by Eq. (1) only after a correction is made to account for the small Zeeman and motional Stark shifts due to the 54-G-rms magnetic induction B in the cavity (produced by the slow- β^+ -beam solenoid).⁹ We estimate the Ps kinetic energy T to range between $T = 0$ and 1 eV. Using a quadratic interpolation from the computed values at 100 G,¹⁰ we find the combined Zeeman and Stark shifts for $\Delta\nu$ [Eq. (1)], averaged over the relevant substates, to be -4.65 MHz for $T = 0$ eV and -1.03 MHz for $T = 1$ eV (in the extreme case of the Ps velocity perpendicular to B). Thus the Zeeman- and motional-Stark-shift-corrected theoretical value of Eq. (1) ranges between 8620 and 8624 MHz, compared to our experimental value of 8628.4 ± 2.8 MHz. We note that the α^3R radiative correction (Lamb shift) in Eq. (1) corresponds to 231 MHz; thus our experiment already confirms this value to a few percent accuracy and may indicate the need for higher-order corrections.

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Note added.—We have now obtained a new measurement of the microwave effect by using a non-resonant circular-wave-guide target region in place of the resonant cavity of Fig. 1. Using a mica target and 120-mW rf power (TE₁₁ mode) traveling opposite to the positron beam we have

measured $S(f)$ at sixteen frequencies from 8375 to 8915 MHz with error bars approximately the same as the open-circle data of Fig. 2. We obtain a Lorentzian fit with $A = (11.7 \pm 0.9)\%$, $f_0 = 8652 \pm 8$ MHz, $\delta = 157 \pm 27$ MHz, and $\chi^2/\nu = 16.6/13$. [Although this χ^2/ν is reasonable, one datum point taken at 8825 MHz, $S(8825) = (5.8 \pm 1.2)\%$, deviates from the fitted curve by 3.1 standard deviations.] Given the new experimental conditions ($B \approx 80$ G), this value of f_0 is consistent with the Zeeman- and motional-Stark-shifted theoretical value plus a possible Doppler shift caused by ≈ 1 -eV positronium atoms leaving the surface of the target. Further measurements of the Doppler shift are in progress.

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¹K. F. Canter, A. P. Mills, and S. Berko, *Phys. Rev. Lett.* **34**, 177 (1975).

²See, for example, V. W. Hughes, in *Atomic Physics*, edited by V. W. Hughes, B. Bederson, V. W. Cohen, and F. M. J. Pichanick (Plenum, New York, 1969), p. 15.

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⁵K. F. Canter, A. P. Mills, and S. Berko, *Phys. Rev. Lett.* **33**, 7 (1974).

⁶In Ref. 1, because of much poorer statistics, this separation into two lifetimes was not possible and an overall lifetime of ≈ 50 nsec was quoted: The two sets of data are consistent with each other.

⁷From the observation of the long lifetime of 1^3S_1 Ps in MgO powder [R. Paulin and G. Ambrosino, *J. Phys. (Paris)* **29**, 263 (1968)] one would expect a very small annihilation probability for Ps collisions with MgO powder.

⁸It is interesting to note that this constitutes the first observation of Ps which does not rely on the detection of annihilation γ 's.

⁹In future experiments the magnetic field could be reduced and made uniform throughout the cavity.

¹⁰M. L. Lewis, private communication; see also Ref. 4.