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Observation of a Positronium Zeeman Transition in γ-Al_2O_3†

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The Zeeman transition between the $M=\pm1$ and $M=0$ states of orthopositronium in its ground state has been observed in a solid for the first time, when positrons were stopped in a γ-Al_2O_3 powder pellet, thus providing strong evidence that essentially free orthopositronium exists in γ-Al_2O_3. The value for the positronium fine-structure interval is $\Delta\nu(\gamma\text{-}Al_2O_3) = 203.308 \pm 0.024$ GHz (120 ppm). The difference $\Delta\nu$(free Ps) $-\Delta\nu(\gamma\text{-}Al_2O_3)$ is 88 ± 25 MHz.

Evidence for the formation of positronium by positrons stopped in powders of metal oxides has been obtained by several methods. Lifetime spectra[1,2] of the annihilation γ rays contain a component with a lifetime approximately equal to the mean life of free orthopositronium,[3] $\tau_0 \simeq 1.4\times10^{-7}$ sec. This long-lifetime component has been associated with three-γ-ray annihilation.[2,4] The 3γ annihilation is quenched by a magnetic field[5,6] in the same way as the 3γ annihilation from free orthopositronium.[7] A formation theory has been presented[8] according to which positrons stop inside the powder grains and form positronium, which then diffuses through the grain surfaces to exist as free positronium in regions outside the powder grains. This Letter reports the first observation of the Zeeman transition between the $M=\pm1$ and $M=0$ sublevels of orthopositronium when positrons are stopped in a γ-Al_2O_3-powder pellet, and provides strong evidence that essentially free orthopositronium exists in γ-Al_2O_3 powder.

The transition was observed by the technique and with the apparatus used to measure the positronium fine-structure interval $\Delta\nu$ in gases.[9,10] Positronium was formed by stopping positrons in a γ-Al_2O_3 powder pellet in a magnetic field of about 7800 G, Zeeman transitions from the $M=\pm1$ to the $M=0$ orthopositronium sublevels were induced with a microwave magnetic field, and the transitions were detected through an increase in the 2γ annihilation rate.

Only minor modifications (Fig. 1) of the apparatus used for a precision measurement of $\Delta\nu$ were necessary to permit introduction of the γ-Al_2O_3 pellet. The γ-Al_2O_3 pellet was 1.9 cm in diame-

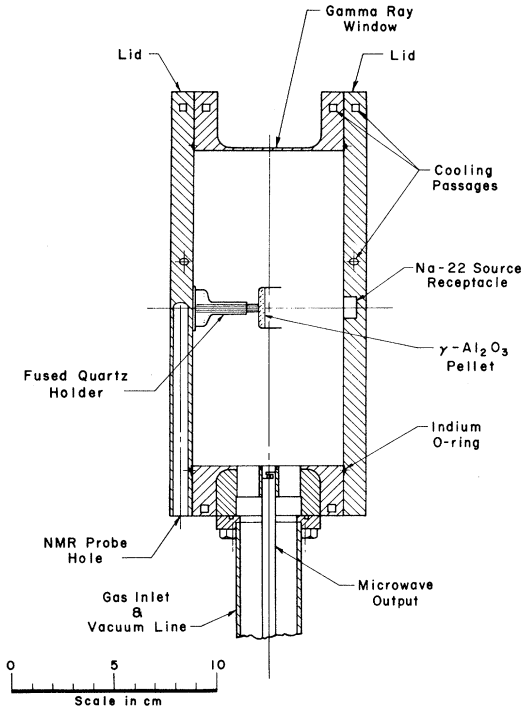

FIG. 1. Diagram of microwave cavity showing placement of γ-Al_2O_3 powder pellet.

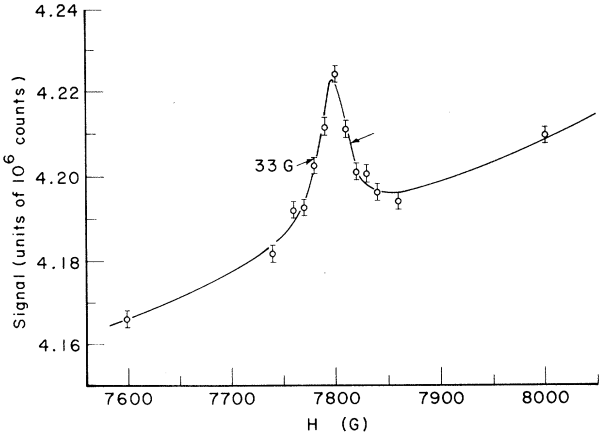

FIG. 2. Positronium Zeeman transition resonance for positrons stopped in γ-Al_2O_3 powder pellet. Plot shows coincidence counting rate versus magnetic field. Data were taken with a microwave frequency of 2323.180 MHz and an input power of 52 W.

ter and 0.3 cm thick and was supported in the center of the microwave cavity by a fused-quartz holder attached to the center of one lid of the cavity. The pellet was pressed from γ-Al_2O_3 powder[11] with particles of 500 Å mean diameter and it had a density of 0.9 g/cm^3 and an internal surface area of 80 ± 4 m^2/g.[12] A long-lifetime component and magnetic quenching of this component were observed in annihilation lifetime spectra for positrons stopped in pellets of this γ-Al_2O_3 powder.[13]

The positron source was 2 mCi of Na^{22} located in a hole in the cavity lid opposite the sample. Up to 52 W of microwave power was coupled into the cavity producing microwave magnetic fields of up to 7.7 G in the TM_{10} mode. Unambiguous mode identification could not be made because of the presence of the dielectric holder and pellet in the cavity. The upper limit on input power was set by microwave breakdown in the cavity. An ion pump was used to evacuate the cavity for several days before any data were taken. The cavity was pumped continuously while data were taken, and the cavity pressure was maintained between 10^{-5} and 10^{-4} Torr, depending primarily on the pellet outgassing rate under microwave heating. Three or four pairs of NaI(Tl) γ-ray detectors were operated in coincidence to detect pairs of two 0.51-MeV γ rays 180° apart in direction with 0.2 μsec time resolution. The coincidence counting rate from each pair was scaled, and the experimental signal was the sum of the coincidence rates from all detector pairs.

Data were taken by varying the magnetic field H with fixed microwave frequency and observing the change in the 2γ coincidence counting rate. With fixed conditions the coincidence counting rate still varied with time by about 0.1% to 1% depending on microwave power level and with a mean period of 50 min, possibly as a result of target charging effects associated with the positrons. A resonance curve is shown in Fig. 2. The circles are experimental points showing the number of coincidence counts recorded in a 300-sec period and are shown with 1-standard-deviation statistical error bars; the entire curve was obtained in about 1 h. The solid curve is the result of a least-squares fit to the data points of a Lorentzian with a linearly increasing background. The nonresonant background arises from the magnetic focusing of the positrons and the increase of the $2\gamma/3\gamma$ branching ratio with increase in field, and it is approximately linear in H over the resonance region. The microwave frequency and the field at the center of the resonance are used to compute $\Delta\nu$.[9] Resonance signals were detected with microwave input powers as low as 7 W but adequate resonance curves were obtained only with powers greater than about 30 W. The results from fits to four resonance curves at three power levels are shown in Table I, togeth-

TABLE I. Ground-state fine-structure interval for positronium in γ-Al_2O_3. Average $\Delta\nu(\gamma\text{-}Al_2O_3) = 203.308 \pm 0.024$ GHz (120 ppm).

Power (W)	$\Delta\nu$ (GHz)	Resonance linewidth FWHM (G)	Signal (%)
32	203.389 ± 0.058	29.7 ± 1.7	0.67 ± 0.05
37	203.252 ± 0.058	33.2 ± 2.0	0.65 ± 0.05
52	203.301 ± 0.030	33.2 ± 1.0	0.89 ± 0.04

er with an average value for $\Delta\nu(\gamma\text{-}Al_2O_3)$.

A search was made for a resonance with only the empty holder in the cavity. The measured resonance signal amplitude of $(0.02 \pm 0.07)\%$ confirmed that no orthopositronium was being observed in the holder. A Zeeman transition resonance was also observed with a γ-Al_2O_3 powder pellet which had been sintered in a vacuum furnace for $2\frac{1}{2}$ h at 1050°C, resulting in a reduction of surface area to 35 ± 4 m^2/g. The resonance signal amplitude for this curve was about $\frac{1}{2}$ that for an unsintered pellet at the same microwave power.

The average value of $\Delta\nu$ for positronium in γ-Al_2O_3 given in Table I is

$$\Delta\nu(\gamma\text{-}Al_2O_3) = 203.308 \pm 0.024 \text{ GHz} \quad (120 \text{ ppm}). \quad (1)$$

The indicated error is 1 standard deviation and is due principally to statistical counting errors. For comparison the value for $\Delta\nu$ for free positronium is[10]

$$\Delta\nu(\text{free Ps}) = 203.396 \pm 0.005 \text{ GHz} \quad (25 \text{ ppm}). \quad (2)$$

The difference

$$\Delta\nu(\text{free Ps}) - \Delta\nu(\gamma\text{-}Al_2O_3) = 88 \pm 25 \text{ MHz} \quad (3)$$

indicates that orthopositronium in the γ-Al_2O_3 pellet was perturbed. The perturbation is probably due to collisions of the orthopositronium atoms with γ-Al_2O_3 surfaces inside the pellet. The difference in Eq. (3) is the same magnitude as the fine-structure density shift of positronium in Ar gas at 6 atm pressure.[9] The measured resonance linewidths in Table I agree with the expected microwave power-broadened linewidths for free orthopositronium in a magnetic field of 7800 G. The resonance signal amplitudes shown in Table I indicate that about 23% of the positrons stopped in the unsintered γ-Al_2O_3 pellet formed positronium. This value is consistent with earlier reported values of 20% to 30% positronium formation.[8] We calculate that about 12% of the positrons stopped in the sintered γ-Al_2O_3 pellet formed positronium. The reduction in positronium formation with reduction in surface area in the sintered material is in qualitative agreement with the theory of positronium diffusion in γ-Al_2O_3 powder.[8]

Our observation of a positronium Zeeman resonance in a solid for the first time opens up a new and precise technique for studying the interactions of positrons and positronium in solids.

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