Positronium fine-structure interval $\Delta\nu$ in oxide powders

M. H. Yam, P. O. Egan, W. E. Frieze, and V. W. Hughes
J. W. Gibbs Laboratory, Yale University, New Haven, Connecticut 06520
(Received 12 January 1978)

The fine-structure interval $\Delta\nu$ of ground-state positronium formed in SiO_2 and MgO powder pellets has been determined through measurements of microwave-induced transitions between Zeeman substates of orthopositronium. The $\Delta\nu$ values for Ps in the powders are less than $\Delta\nu$ for free Ps by the order of 1 part in 10^4, suggesting an attractive interaction between Ps and the powder grain surfaces within the pellet. No Zeeman transition was found for positrons stopping in a silicon single crystal or in fused quartz.

I. INTRODUCTION

The study of positronium (Ps or e^+e^-) in condensed matter is an active field of research[1-4] which seeks to learn about the formation and interaction of positronium in solids and liquids. It is known that Ps is formed in many molecular solids and liquids but is generally not formed in ionic solids and metals. Formation of Ps occurs in metal-oxide powders where almost free Ps may exist between the powder grains.[5,6] The metal oxides serve also to produce slow positrons[7] and positronium.[8] Hence, the interaction mechanisms of e^+ and Ps with metal oxides are of particular interest.

Positronium is identified from the energy and lifetime spectra of the annihilation γ rays, principally from the relatively-long-lifetime orthopositronium component. The effect of a static magnetic field on the annihilation spectra provides an additional tool for identifying Ps. The principal interaction of Ps with condensed matter that has been studied is the shortening of the lifetime of orthopositronium. The angular correlation spectrum of the 2γ annihilation indicates the presence of parapositronium and determines its kinetic energy.[9,10]

An additional, more refined tool for studying the interaction of Ps in condensed matter is the observation of the microwave-induced Zeeman transition which determines the effective fine-structure interval $\Delta\nu$ for Ps. A shift in $\Delta\nu$ from its value for free Ps is due to the interaction of Ps in matter. This microwave resonance type of experiment was introduced for Ps formed in γ-Al_2O_3 powder pellets.[11] We report here more extensive measurements of $\Delta\nu$ for Ps in SiO_2 and MgO powders. Preliminary reports of these results have been made.[12,13]

II. EXPERIMENTAL METHOD

The method of the experiment is the same as that reported earlier[11] for Ps in γ-Al_2O_3 and also for Ps in gases,[14-16] which determines the precise value for $\Delta\nu$ of free Ps. Indeed the experimental apparatus used in the most recent precision determination[16] of $\Delta\nu$(Ps) is used for the measurements reported here.

The Zeeman transition between the $M = \pm 1$ and $M = 0$ sublevels of ground-state orthopositronium is observed when positrons are stopped in con-

TABLE I. Physical properties of metal-oxide powders.

Sample	Specific surface area (m^2/gm)	Mean radius (Å)	Spread in radius (Å)	Density of solid bead (gm/cm^3)	Density of pellet [a] (gm/cm^3)
SiO_2(380) [b]	380 ± 30	35	18	2.20	0.51 ± 0.05
SiO_2(300) [b]	300 ± 30	35	18	2.20	0.51 ± 0.05
SiO_2(130) [b]	130 ± 25	80	40	2.20	0.51 ± 0.05
MgO(110)	110 ± 15	100	···	3.58 [c]	1.28 ± 0.13
γ-Al_2O_3(80)	80 ± 4	250	···	3.5–3.9 [c]	0.9

[a] A quantity $\rho^* = \rho_{pellet}/(\rho_{bead} - \rho_{pellet})$ is often useful for studies of these powders (Ref. 17). For our samples ρ^* is 0.30 ± 0.03 for SiO_2, 0.56 ± 0.06 for MgO, and 0.30–0.35 for γ-Al_2O_3.

[b] These SiO_2 samples are Aerosil 130, 300, and 380, supplied by Degussa Inc., New York, N. Y. and Hanau/Main, West Germany.

[c] *Handbook of Chemistry and Physics*, edited by R. C. Weast, 50th ed. (Chemical Rubber, Cleveland, 1969).

 © 1978 The American Physical Society

densed matter. In a magnetic field of about 7800 G the Zeeman transitions are induced with a microwave magnetic field and detected through an increase in the 2γ 0.5-MeV annihilation rate.

The physical properties of SiO_2 and MgO samples used in this experiment are given in Table I. The powder grains are spheres with an approximately Gaussian distribution in their radii. All samples are pressed into cylindrical pellets of 1.9-cm diameter and 0.2–0.6-cm thickness. The density of a pellet is determined after the microwave heating which occurs in the $\Delta\nu$ measurement.

In addition to these powders, fused quartz and a silicon single crystal are studied. The silicon crystal is 2.1 cm in diameter and 0.45 cm in thickness. The fused-quartz sample is 2.1 cm in diameter and 0.41 cm in thickness.

For the measurements of the powder pellets, the microwave cavity is shown in Fig. 1 with a fused-quartz sample holder 1.9-cm in inner diameter and 1.2 cm in depth cemented onto one lid of the cavity. Dimensions of the quartz holder are chosen so that the sample position coincides with the viewing region of the NaI(Tl) detectors. The majority of the positrons entering the microwave cavity stop in the sample; less than 1% stop in the quartz sample holder. Prior to the experiment the sample is placed in the cavity which is then evacuated to about 10^{-5} Torr. Microwave power of 5–10 W at 2.3 GHz is coupled into the cavity, and heating for 5–10 h. is employed to achieve a pressure below 10^{-6} Torr before data taking begins. For fused quartz and silicon a different quartz sample holder consisting of a back plate and tentacle like clips is used in the experiment.

For the measurements of SiO_2 and MgO powder pellets the ^{22}Na source strength is 0.5 mCi, and for the Si and fused quartz it is 2 mCi.

The principal difference in experimental technique as compared to the earlier measurement[11] on γ-Al_2O_3 is the use of pulsed 2.3-GHz microwave power produced by a positive-intrinsic-negative diode modulator at a rate of 1–5 kHz. The microwave induced signal at a particular value of static magnetic field H is defined by

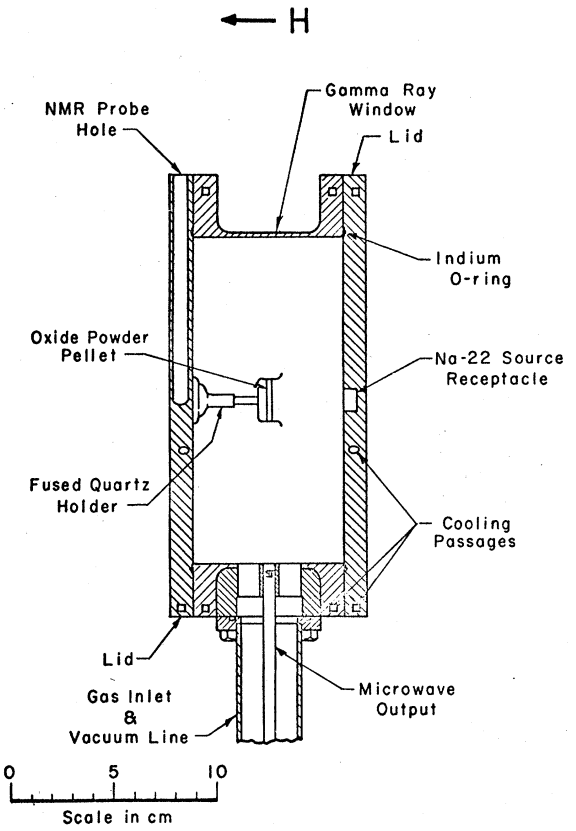

FIG. 1. Cross section of microwave cavity showing placement of pellet. The direction of the static magnetic field H is indicated.

$$\text{Signal} = \frac{2\gamma \text{ rate (microwaves on)} - 2\gamma \text{ rate (microwaves off)}}{2\gamma \text{ rate (microwaves on)} + 2\gamma \text{ rate (microwaves off)}} . \quad (1)$$

A resonance line is obtained by measuring the signal as a function of the static magnetic field H. The rapid modulation reduces the effect on the signal of relatively slower drifts or instabilities in experimental conditions, such as the cyclic variation in coincidence counting rate observed in the earlier experiment.[11] Background associated with nonresonant variation of the 2γ coincidence rate with static magnetic field is largely avoided. High-peak microwave power of up to 320 W could be coupled into the microwave cavity at the pressure of 10^{-6} Torr, and customarily peak powers of 40–60 W are used. No power leveling is required for the microwave system. The rectified output of a crystal microwave detector is used to gate appropriate scalers which collect the 2γ coincidence counts.

Resonance lines are obtained under computer control by varying the static magnetic field with the microwave frequency and power held fixed. Typical 2γ coincidence rates are 600–2000 sec^{-1} per detector pair. A resonance curve is obtained in a period of 5–10 h. A typical resonance curve is shown in Fig. 2. The full theoretical line shape

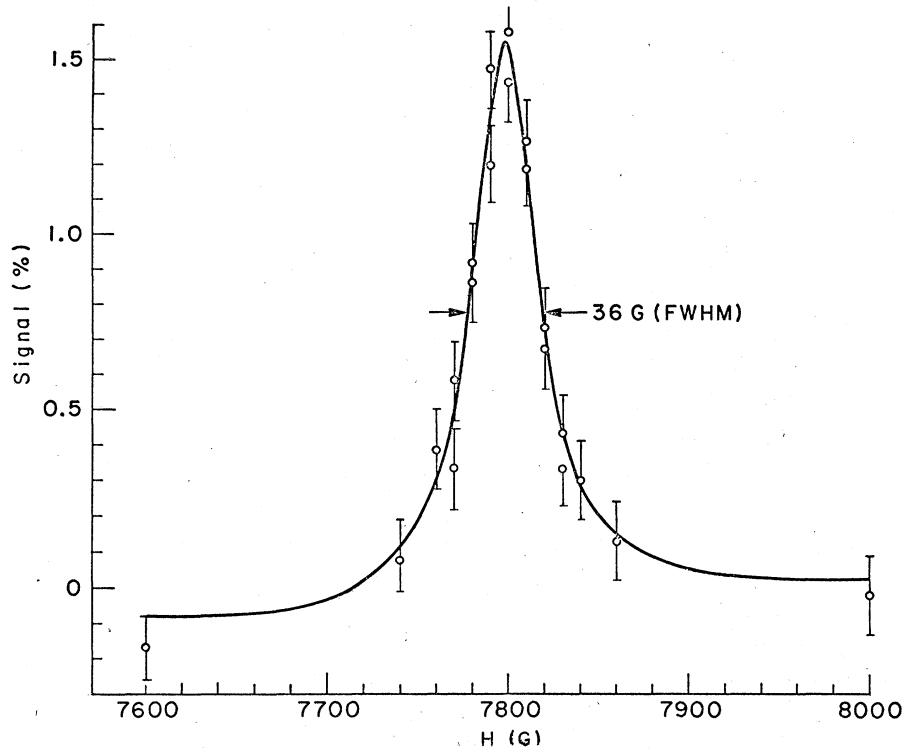

FIG. 2. Positronium Zeeman resonance signal observed for positrons stopped in a SiO_2-powder pellet [sample SiO_2 (380) of Table I] with a peak microwave power of 320 W. The solid curve is the theoretical line shape fit to the experimental points, whose statistical counting errors are indicated.

given in an earlier publication[16] is fit to the data points using a least-squares optimization method. To an adequate approximation the theoretical line shape is Lorentzian with its center at the resonance field value corresponding to the microwave frequency f,

$$f = \tfrac{1}{2}\Delta\nu[(1+x^2)^{1/2} - 1], \tag{2}$$

where $x = 2g'\mu_B H/h\Delta\nu$ in which g' is the gyromagnetic ratio of the electron bound in Ps and μ_B is the Bohr magneton. The linewidth is determined by the annihilation rates of the Ps states and by microwave power broadening and is essentially the same as for Ps in a gas.[14,16] The value of $\Delta\nu$ is obtained with Eq. (2). The error in our measured value of $\Delta\nu$ is primarily the statistical counting error and corresponds to the determination of the resonance magnetic field value to about 1 part in 50 of the linewidth.

III. RESULTS AND DISCUSSION

A summary of results for the oxide powders is given in Table II. We note that the values of $\Delta\nu$ for Ps in the powders are close to that for free Ps, $\Delta\nu$(Ps), but differ by significant amounts. The shifts $\Delta\nu$(Ps) $-\Delta\nu$ (powder) are all positive and of the order of 1 part in 10^4.

All the beads are assumed to be spherical and to have the same density, and hence the specific surface area (m^2/gm) of a bead will be inversely proportional to the bead radius. A pellet is assumed to consist of a uniform distribution of beads. We have data on pellets made from three different bead sizes of SiO_2, and the pellets are constructed so as to have the same density (Table I). In view of the variation in specific surface area and in bead radii assigned by the manufacturer, we should group together the SiO_2(380) and SiO_2(300) data points given in Table II. We then note that the shift in $\Delta\nu$ decreases significantly for the SiO_2(130) sample with its distinctly lower value of specific surface area. This result for SiO_2 as a function of bead specific surface area is consistent with the simple picture[6] that Ps exists in the interstices between the powder grains and that the shift in $\Delta\nu$, as in a gas, will be proportional to the number of collisions during the lifetime of Ps, which is proportional to the specific surface area. We note that the magnitude of the shift of $\Delta\nu$ in powders is comparable to and of the same sign as that due to Ps collisions in Ar

TABLE II. Positronium $\Delta\nu$ in oxide powders.

Sample	Average microwave power (W)	Peak microwave power (W)	Signal (%)	$\Delta\nu$ (GHz)	$\Delta\nu$(Ps) [a] $-\Delta\nu$(powder) (MHz)
SiO_2(380) [b]	40	320	1.5	203.270(14)(70 ppm)	+115(14)
SiO_2(380) [b]	50	100	0.8	203.265(16)(80 ppm)	+120(16)
SiO_2(300)	50	110	0.9	203.235(15)(75 ppm)	+150(15)
SiO_2(130)	60	130	0.7	203.313(17)(85 ppm)	+72(17)
MgO(110)	50	100	0.4	203.302(49)(120 ppm)	+83(49)
γ-Al_2O_3(80) [c]	30–50	30–50	0.3–0.5	203.308(24)(120 ppm)	+77(24)

[a] The value of $\Delta\nu$ for free Ps is taken from Ref. 16: $\Delta\nu$(Ps) = 203.3849(12) GHz.

[b] For these data, two different pellets made from the same batch of beads are used. One of the pellets is used for the condition of 320-W peak microwave power and the other for both the 320- and 100-W peak microwave power conditions.

[c] This sample was measured in Ref. 11. Because of current interest in Ps in low-density powders (Ref. 17), we made a preliminary, but unsuccessful search for a resonance signal in SiO_2 powder of density 0.05 gm/cm^3. The low- positron-stopping density in the powder and the breakage due to microwave heating of the thin window used to contain the powder are technical obstacles.

gas at about 10 atm. The sign of the $\Delta\nu$ shift in powders suggests that an attractive Ps surface interaction, perhaps a long-range Van der Waals force, may be involved. This Ps-surface interaction which leads to the $\Delta\nu$ shift is presumably also responsible for the increase in orthopositronium decay rate observed in powders.[5,6,17,18]

Although the peak resonance signal should be proportional to the Ps formation fraction in the powder, it is difficult to conclude much more about formation than that it is the same within about a factor of 3 for the various powder samples studied.[2]

With silicon and fused quartz no resonance signal was observed over the range of static magnetic field from 6 to 9 kG where a signal as large as 3×10^{-4} would have been seen.

Formation of muonium has also been observed in metal-oxide powders.[19]

ACKNOWLEDGMENTS

We are grateful to G. zu Putlitz of the University of Heidelberg and M. Leventhal of Bell Telephone Laboratories for supplying the metal-oxide powders, and the silicon and fused quartz, respectively. A. Rich and K. Marko of the University of Michigan collaborated on the search for a resonance in a low-density SiO_2 powder mentioned in Table II. This work was supported in part by the National Science Foundation under Grant No PHY-77 19615.

[1] *Positron Annihilation*, edited by A. T. Stewart and L. O. Roellig (Academic, New York 1967).

[2] *Proceedings of Third International Conference on Positron Annihilation*, edited by P. Hautojärvi and A. Seeger (Springer-Verlag, Heidelberg, 1975).

[3] I. Ya. Dekhtyar, Phys. Rep. 9, 243 (1974).

[4] J. Green and J. Lee, *Positronium Chemistry* (Academic, New York, 1964).

[5] W. Brandt and R. Paulin, Phys. Rev. B 5, 2430 (1972).

[6] R. Paulin and G. Ambrosino, J. Phys. (Paris) 29, 263 (1968).

[7] K. F. Canter *et al.*, J. Phys. B 5, L167 (1972).

[8] K. F. Canter, A. P. Mills, Jr., and S. Berko, Phys. Rev. Lett. 33, 7 (1974).

[9] L. A. Page and M. Heinberg, Phys. Rev. 102, 1545 (1956).

[10] S. Berko and F. L. Hereford, Rev. Mod. Phys. 28, 299 (1956).

[11] D. J. Judd *et al.*, Phys. Rev. Lett. 30, 202 (1973).

[12] P. O. Egan *et al.*, Bull. Am. Phys. Soc. 18, 1503 (1973). The $\Delta\nu$ shift reported in this preliminary experiment was considerably higher than the value reported in the present paper. The earlier experiment was of much lower precision and did not utilize pulsing of the microwave power.

[13] M. H. Yam *et al.*, Bull. Am. Phys. Soc. 21, 625 (1976).

[14] E. D. Theriot *et al.*, Phys. Rev. A 2, 707 (1970).

[15] E. R. Carlson, V. W. Hughes, and I. Lindgren, Phys. Rev. A 15, 241 (1977).

[16] P. O. Egan, V. W. Hughes, and M. H. Yam, Phys. Rev. A 15, 251 (1977).

[17] D. W. Gidley, K. A. Marko, and A. Rich, Phys. Rev. Lett. 36, 395 (1976).

[18] G. W. Ford, L. M. Sander, and T. A. Witten, Phys. Rev. Lett. 36, 1269 (1976).

[19] J. H. Brewer *et al.*, in *Muon Physics III*, edited by V. W. Hughes and C. S. Wu, (Academic, New York, 1975), p. 4.