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Defects in vitreous SiO₂ probed by positron annihilation

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Abstract

Annihilation characteristics of positrons in a single quartz crystal and vitreous silica glasses (v-SiO₂) were studied by measurements of two-dimensional angular correlation of positron annihilation radiations. From the measurements, it was found that positrons and positronium (Ps) atoms mainly annihilate from trapped states by vacancy-type defects in the v-SiO₂ specimen. For the v-SiO₂ specimen with a cylindrical porous structure, the annihilation of Ps with anisotropic momentum distributions was observed. This was attributed to the momentum uncertainty due to a localization of Ps in a finite dimension of pores. The present investigation showed possibilities for the detection of microstructures in v-SiO₂ by the positron annihilation technique.

1. Introduction

Defects in vitreous silica glasses (v-SiO₂) are known to be associated with optical absorption bands.¹ Because of the technological importance of v-SiO₂, significant studies of defects have been made, however, the nature of the intrinsic defects in SiO₂ has still open questions. The positron annihilation technique is now established as one of the best methods for the investigation of vacancy-type defects in crystalline solids. However, annihilation characteristics of positrons or positronium atoms (Ps) trapped by defects in SiO₂ are not well understood at this stage. In the present paper, we report a study of defects in v-SiO₂ by measurements of two-dimensional angular correlation of positron annihilation radiations (2D-ACAR).

2. Experimental

The specimens used in the present experiments were a single crystalline semiconductor grade α -SiO₂, a v-SiO₂ and a porous silica glass (PSG) with a cylindrical structure. The α -SiO₂ specimen was obtained from Poland. For the PSG specimen, the diameter of cylinders was 5 nm. The 2D-ACAR spectra were measured by the system constructed at the University of Tsukuba.² In order to know annihilation characteristics of positrons, lifetime spectra of positrons were also obtained by the conventional fast-fast system.

3. Results and Discussion

Figure 1 shows the momentum density of electron-positron pairs for the α -SiO₂ specimen. Berko *et al.*³ and Ikari and Fujiwara⁴ reported the 2D-ACAR spectrum for a single-crystal quartz. They observed a narrow peak located at zero momentum corresponding to the self-annihilation of para-Ps with an energy width near the resolution of the measuring system and peaks located at positions corresponding to a Bloch state of Ps. These results suggest that Ps formed in the α -SiO₂ specimen annihilates from the free state. Figure 2 shows the 2D-ACAR spectrum and its contour map for the v-SiO₂ specimen. An obtained homogeneous momentum distribution of electron-positron pairs is due to the annihilation of

Fig. 1. The momentum distribution of electron-positron pairs for the α -SiO₂ specimen.

Fig. 2. The momentum distribution of electron-positron pairs for the v -SiO₂ specimen (above) and its contour map (below).

Fig. 3. The momentum distribution of electron-positron pairs for the PSG specimen (above) and its contour map (below).

positrons in amorphous materials. However, no peak corresponding to the annihilation of Ps from the free state was observed. In order to discuss annihilation characteristics of positrons in the v-SiO₂ specimens in more detail, lifetime spectra were also measured. The results are summarized in Table I. The lifetime spectrum was decomposed into three components and the longest lifetime was obtained as 1.625 ns with the intensity of 47.7 %. This component can be associated with the pick-off annihilation of ortho-Ps. For the α -SiO₂ specimen, the intensity corresponding to the pick-off annihilation of ortho-Ps was only ~2 %. Because of the high sensitivity of positrons to vacancy-type defects, almost all positrons are considered to be trapped by vacancy-type defects in the v-SiO₂ specimen. Thus, the trapping of positrons by vacancy-type defects is considered to enhance the formation of Ps. The broadened spectrum for the v-SiO₂ specimen (Fig. 2) can be attributed to the broadening of the momentum distribution of Ps due to annihilation from such a localized state. Figure 3 shows the 2D-ACAR spectrum and its contour map for the PSG specimen with cylindrical pores of 5 nm diameter, where the cylinder lies parallel to the direction of p_y . In the contour map, a large anisotropy of the momentum distribution was observed at $|p| = 0 \sim 1$ mrad. This anisotropy can be attributed to the momentum distribution of Ps trapped in the cylindrical pores. Since Ps can move freely in the direction parallel to the cylinders (p_y), the observed broadening of the momentum distribution in the p_z direction can be attributed to the momentum uncertainty due to localization in a finite dimension in this direction in the cylindrical pores. The anisotropic peak corresponding to the annihilation of Ps trapped by the cylindrical pores in the 2D-ACAR spectrum was fitted to a Gaussian function. The obtained standard deviation in the p_y direction, σ_y , was obtained as 0.659 mrad and that in the p_z direction, σ_z , was 0.511 mrad. For the v-SiO₂ specimen, the peak corresponding to the annihilation of Ps trapped by vacancy-type defects were 1.63 mrad and 1.59 mrad, respectively. The value of σ_z/σ_y for the v-SiO₂ specimen is 1.03. However, σ_z/σ_y for the PSG specimen was 1.29. This suggests the localization of Ps in a finite dimension of pores. For the v-SiO₂ specimen, the large values of σ_y and σ_z are considered to be due to the trapping of Ps by vacancy-type defects. The lifetime spectrum for the PSG specimen was decomposed into four components (Table I). Since the longest component (13 ns) was not observed in the v-SiO₂ specimen, this lifetime can be associated with the lifetime of ortho-Ps trapped in the cylindrical structure.

Table I. The lifetimes and relative intensities for the v-SiO₂ specimen and the PSG specimen.

specimen	τ_1 (ps)	τ_2 (ps)	τ_3 (ns)	τ_4 (ns)
v-SiO ₂	130(5)	455(9)	1.625(4)	-
PSG	137(8)	417(6)	1.48(6)	13(2)
	I_1 (%)	I_2 (%)	I_3 (%)	I_4 (%)
v-SiO ₂	24.0(6)	28.2(4)	47.7(2)	-
PSG	19(1)	62.9(7)	12.5(3)	5.6(2)

Conclusions

We have studied the positron annihilation in the SiO₂ specimens by measurements of 2D-ACAR. For the v-SiO₂ specimen, it was found that positrons mainly annihilated in vacancy-type defects. The broadening of the momentum distribution of Ps was observed in the 2D-ACAR spectrum for the v-SiO₂ specimen. This was attributed to the localization of Ps in such defects. For the PSG specimen with a pore diameter of 5 nm, a large anisotropy in the 2D-ACAR spectrum was observed. This was attributed to the momentum uncertainty of Ps due to the localization in a finite dimension in the cylindrical pores.

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/1/ *The Physics and Technology of Amorphous SiO₂*, edited by R.A.B.Devine (Plenum Press, New York, 1988).

/2/ KONDO, H., T.Kubota, H.Nakashima, T.Kawano and S.Tanigawa, J. Phys. Condens. Matter 4 (1992) 4595.

/3/ BERKO, S., M.Haghoosie and J.J.Mader, Phys. Lett. A 63 (1977) 335.

/4/ IKARI, H. and K.Fujiwara, J. Phys. Soc. Jpn. 46 (1979) 92.