POSITRON ANNIHILATION IN FAST-NEUTRON-IRRADIATED QUARTZ

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The effect of fast-neutron irradiation on the angular distribution of annihilation γ pairs in crystalline and fused quartz is investigated. It is found that the half-width of the correlation curves varies with the quartz density. From an analysis of various positron states in quartz preceding annihilation and on the basis of the experimental data it is concluded that the variation of the half-width results from the appearance in the correlation curves of a narrow component whose intensity depends on the radiation dose. The narrow component is due to annihilation decay of parapositronium atoms produced in irradiated quartz.

INTRODUCTION

In their investigation of positron lifetimes with respect to annihilation in fused quartz Bell and Graham[1] found that the short-lived component with $\tau_1 \approx 3.5 \times 10^{-10}$ sec is accompanied by a long-lived component with the average lifetime $\tau_2 \approx 18 \times 10^{-10}$ sec ($I_2 \approx 29\%$). In crystalline quartz only a short-lived component with $\tau_1 \approx 2.0 \times 10^{-10}$ sec was observed. In[1] the τ_2 component was attributed to the formation of positronium (Ps) atoms in fused quartz. These results were confirmed in[2], where it was found that the τ_2 component corresponds to the nearly twice greater probability of three-photon annihilation ($P_{3\gamma}$) in fused as compared with crystalline quartz. Telegdi et al.[3] measured the probability ratio of 3γ annihilation in fused quartz and aluminum (free annihilation) and obtained $P_{3\gamma}^{qu}/P_{3\gamma}^{Al} \approx 2.29$. The data on magnetic quenching of $P_{3\gamma}$ in quartz[3] agree well with the hypothesis that Ps atoms are formed in fused quartz if we assume that the principal mechanism of ortho-Ps quenching consists in ortho-para conversion and pick-off annihilation. In the absence of a magnetic field the principal mechanism that shortens the lifetime of ortho-Ps in fused quartz to $\sim 10^{-9}$ sec is pick-off annihilation (in the vacuum the lifetime of the positronium triplet state with respect to annihilation is $\tau_t^0 \approx 1.4 \times 10^{-7}$ sec).

The existence of positronium atoms creates a narrow component in the angular correlation curves of annihilation γ pairs in fused quartz;[4-6] the narrow component was not observed in the case of crystalline quartz. As expected, the narrow component was enhanced in a magnetic field, but the angular distribution in crystalline quartz was not influenced by a magnetic field.[5] However, the observed intensity of the narrow component, $I_N \approx 17.6\%$,[4-6] did not agree with the intensity of the long-lived component of the lifetime spectrum, $I_2 \approx 29\%$[1] (when ortho-para conversion of positronium does not occur we should have $I_2 = 3I_N$). This discrepancy was later removed by Green and Bell,[7] who found that I_2 (and also τ_2) depends on the purity of the fused quartz. Thus for "Vitreosil" quartz (99.8% pure) $I_2 \approx 53\%$ ($\tau_2 \approx 15 \times 10^{-10}$ sec) was obtained, while for "Vycor" (96% SiO_2) the result was $I_2 \approx 32\%$ ($\tau_2 \approx 17 \times 10^{-10}$ sec). Moreover, the authors of[4-6] neglected the possibility that the broader component might be narrowed by a change in the character of the chemical bonds following the crystal-glass transition; the narrow component could then appear to be enhanced.

No appreciable temperature dependence of τ_2 and I_N in fused quartz was observed in[1,8]. The experiments of Colombino et al.[9] showed that density could not be the only factor involved in the formation of positronium. Four modifications of quartz were studied: fused quartz (2.1–2.3 g/cm^3), crystalline quartz (2.65 g/cm^3), tridymite (with density close to that of fused quartz), and cristobalite. A narrow component in the correlation curves was observed only in the case of fused quartz.

Later investigations[10] showed that the time spectrum of positronium annihilation in fused quartz contains three, rather than only two components; the lifetime corresponding to the third component lies between the previously observed values τ_1 and τ_2. An intense narrow component was observed in highly-dispersed crystalline quartz: $I_N \approx 13\%$.[11]

The foregoing review shows that some questions regarding the process of positron annihilation in quartz, and specifically the relation between positron atom formation and the structure or density of quartz require further study. In the present work we have investigated the influence of fast-neutron irradiation on the annihilation properties of quartz. Similar investigations have been conducted previously only for molecular crystals and polymers (see Gol'danskii's review in[12]). Quartz is of interest in this respect because we are enabled to observe how the annihilation properties change during the continuous transformation from the crystalline to the amorphous phase, without breakdown, of a sample that is being irradiated with neutrons.

EXPERIMENTAL TECHNIQUE AND RESULTS

We measured the angular distribution of annihilation γ pairs using apparatus with parallel-slit geometry;[13] the geometric resolution was $\Delta\theta = 1$ mrad. The positron source was ~ 4 mCi of $Na^{22}Cl$. In this apparatus there is a simple relationship between the z component of the momentum possessed by the center of mass of an annihilating pair and the angle θ, which is the measured

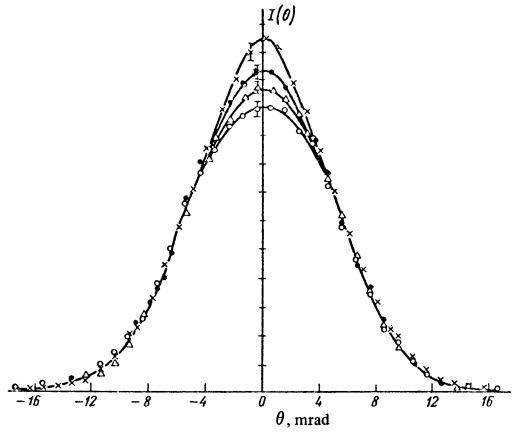

FIG. 1. Angular correlation curves for crystalline quartz (Y cut) after different fast-neutron doses: ○—unirradiated; △—6.2×10^{19}, ●—9.3×10^{19}, and ×—2.2×10^{20} neutrons/cm^2.

Table I. Half-widths of correlation curves for unirradiated and irradiated crystalline and fused quartz

Quartz	Γ, mrad (±0.2 mrad)			
	Unirradiated	9.3×10^{19} neutrons/cm^2	6.2×10^{19} neutrons/cm^2	2.2×10^{20} neutrons/cm^2
Single crystal				
X	12.8	11.9		11.0
Y	12.8	12.4	11.7	11.0
Z	12.6	12.0	11.4	11.1
Fused				
KI	8.0	11.8	11.1	10.8
KV	8.1	11.4	11.3	11.2

deviation of the opening angle of the annihilation γ pair from π: $p_Z \approx m_0 c\theta$ (m is the electron rest mass and c is the velocity of light).

Angular correlation curves of $I(\theta)$ were plotted for unirradiated single crystals (X, Y, and Z cuts) and fused quartz, fast-neutron irradiated single crystals (X, Y, and Z cuts), KI fused quartz (containing 10^{-1}—10^{-3} wt.% of random impurities), and KV fused quartz (having 5×10^{-3} wt.% hydroxyl content in addition to random impurities). The integral fast-neutron flux was 6.2×10^{19}, 9.3×10^{19}, and 2.2×10^{20} neutrons/cm^2, respectively.

The counting rate of annihilation γ-pair coincidences was recorded at 0.5-mrad angular intervals; at the peaks of the $I(\theta)$ curves the average number of counts per experimental point was ~2000. The background random coincidences was ~2% of the peak count and was subtracted from the correlation curves after their half-widths had been determined [i.e., the total width at half maximum of each $I(\theta)$ curve].

Single crystals of unirradiated quartz having different cuts do not differ appreciably with regard to the shape and half-width of the correlation curve (Table I). We obtained a considerably larger value, $\Gamma = 12.8$ mrad, than other authors,[4,9] whose result for crystalline quartz was $\Gamma \approx 11$ mrad. For fused quartz our result $\Gamma = 8$ mrad agrees with[9] within error limits.

With increase of the radiation dose the angular distribution for irradiated quartz single crystals becomes narrower; for a dose of 2.2×10^{22} neutrons/cm^2 the relative diminution of Γ is 14%. For fused quartz the half-width at first increases steeply, to about 50% increase at 6.2×10^{19} neutrons/cm^2. With subsequent

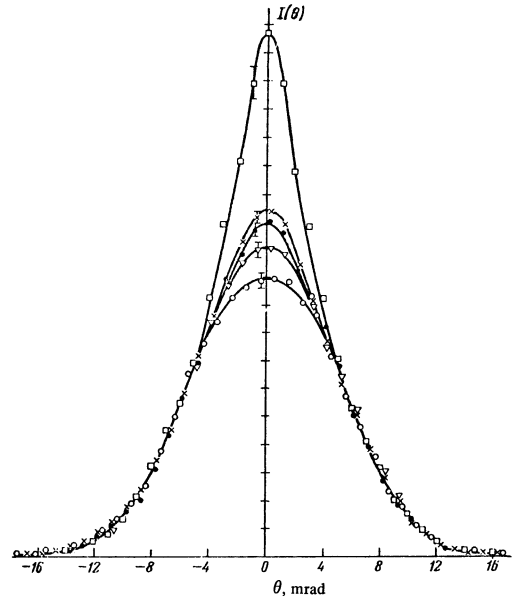

FIG. 2. Angular correlation curves for KI fused quartz after different fast-neutron doses: □ and ○—unirradiated fused and crystalline quartz, respectively; ▽—6.2×10^{19}, ●—9.3×10^{19}, and ×—2.2×10^{20} neutrons/cm^2.

increases of the dose the half-width diminishes somewhat and practically coincides with Γ for crystalline quartz at equal doses. KV and KI fused quartz exhibit considerably different behaviors. The angular distribution for KV quartz remains practically unchanged when the radiation dose is increased from 6.2×10^{19} to 2.2×10^{20} neutrons/cm^2.

As the radiation dose is increased the intensity I_N of the narrow component in the correlation curves of monocrystalline and fused quartz exhibits opposite behavior to that of the half-width (Table II). I_N was determined by the method of Page et al.,[4-6] whereby the experimental $I(\theta)$ curves merged in the angular region $\theta > 5$ mrad (Figs. 1 and 2). We note that $I_N \approx 21\%$ for unirradiated fused quartz is in good agreement with the results obtained by other authors: 18—20% in[4-6,8] and 22% in[9]. De Zafra's method[8] yielded approximately the same results.

DISCUSSION OF RESULTS

It has been firmly established that following a heavy dose of fast-neutron irradiation ($\geq 10^{20}$ neutrons/cm^2) crystalline quartz is transformed into an amorphous phase resembling quartz glass.[14,15] Following a dose of 1.5×10^{20} neutrons/cm^2 the density of crystalline quartz is reduced about 14%. Subsequent increase of the dose to 4×10^{20} neutrons/cm^2 is accompanied by only a small change of the density.[16] The density of fused quartz irradiated with 5×10^{19} neutrons/cm^2 increases almost 3% but diminishes somewhat for higher doses. For $\geq 10^{20}$ neutrons/cm^2 the densities of crystalline and fused quartz approach the same limiting value. The different behaviors of the density in the cases of crystalline and fused quartz are accounted for as follows: Under irradiation the crystal structure of quartz breaks down and disintegrates, whereas in fused quartz partial ordering of the structure occurs during the initial stage of irradiation.

Table II. Intensity of the narrow component in correlation curves of crystalline and fused quartz irradiated with different doses of fast neutrons

Quartz	I_N, %			
	Unirradiated	6.2×10^{19} neutrons/cm^2	9.3×10^{19} neutrons/cm^2	2.2×10^{19} neutrons/cm^2
Single crystal (Y cut)	0.0	3.0	5.7	8.4
Fused KI	21.4	4.4	6.5	8.7

Irradiation creates different kinds of defects and aggregations of defects in quartz (vacancies, interstitial atoms etc.)[15]. Therefore when interpreting the experimental data on positron annihilation in irradiated quartz we must take into account the possibility that various bound states of positrons will be formed.

Fast positrons introduced into a target are subject to ionization energy losses, annihilation in flight, the formation of positronium atoms, and elastic scattering on ions. The positrons lose energy in all these processes and are slowed down. The positron thermalization time in ionic media is of the order 10^{-11}—10^{-12} sec,[17] which is much shorter than the short positron lifetime with respect to annihilation in quartz (see above). Some of the thermalized positrons undergo free annihilation; the remaining positrons are captured by negative ions or defects. Gol'danskii et al.[18] have shown that in ionic media the annihilation probability of free positrons is negligibly small compared with the probability of the two-photon annihilation of bound positrons. On the basis of the foregoing, we considered the contributions in quartz to the annihilation of positrons in the following states: a) positronium atoms, b) the quasiatomic system $e^+0^{\eta^-}$ (where η is the effective charge of the oxygen ion), c) positrons bound to interstitial 0^{2-} ions, and d) F_+ centers (positrons in silicon vacancies).

The formation of positronium atoms is observed in fused quartz and also in irradiated crystalline and fused quartz. The principal proof thereof is found in the fact that the half-widths of the angular correlation curves depend on the density of the irradiated quartz. The reduced density of fast-neutron irradiated crystalline quartz leads to the appearance of the narrow component and to the enhancement of that component as the radiation dose is increased (Table II). The enhanced density and partial ordering of irradiated fused quartz reduce drastically the intensity I_N of the narrow component; the minimum of I_N corresponds to the maximum density of fused quartz (irradiated with $\sim 5 \times 10^{19}$ neutrons/cm^2). As the density of fused quartz diminishes under additional irradiation we observe a corresponding enhancement of the narrow component.

We also note that the half-width of the narrow component in unirradiated fused quartz and in irradiated samples of both fused and crystalline quartz has an identical value $\Gamma_N = (4.2 \pm 0.3)$ mrad within experimental error limits. This indicates that a single mechanism is responsible for the narrow component in the correlation curves of quartz and confirms the foregoing conclusion.

Our calculated annihilation properties of positrons from different states in irradiated and unirradiated quartz show that the angular distribution of annihilation γ pairs depends mainly on the annihilation decay of positronium atoms and of quasiatomic systems that consist of e^+ and an oxygen ion. The first of these mechanisms gives rise to the narrow component in the correlation curves. The intensity of the narrow component is augmented as the quartz is amorphized by irradiation, although in the investigated range of radiation doses this component does not attain the same magnitude as in unirradiated fused quartz. The large half-width, $\Gamma_N \approx 4$ mrad, of the narrow component indicates that parapositronium atoms in quartz (irradiated or unirradiated) are not thermalized or are captured by defects, thus increasing the zero-point energy.[8]

The broader component in quartz results from the formation of the metastable quasiatomic $e^+0^{\eta^-}$ system, whose half-width is determined by the effective charge η of the oxygen ions in quartz. In irradiated quartz a contribution to the broader component can also come from positrons captured by interstitial oxygen ions. The pick-off annihilation of orthopositronium atoms also contributes to the broader component.

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