

ANNIHILATION OF POSITRONS IN IONIC CRYSTALS

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CONTENTS

1. Average lifetimes of positrons in ionic crystals. 72
 2. Relative counting rate of three-photon coincidences 74
 3. Angular correlation of γ quanta in two-photon annihilation of positrons 74
 4. Positron states in ionic crystals 77
 Cited Literature. 79

THE study of the annihilation of positrons becomes more and more important as a means of investigating the electronic structure of matter. The registration of annihilation quanta makes it possible to determine the momentum distribution of the electrons, and also the mechanisms whereby electrons of a substance interact with positrons. By now, many experiments on the annihilation of positrons in solids have been performed. The main results of these experiments are given in the collection^[1] and in the reviews^[2-8]. In the present review we describe the status of research on annihilation of positrons in media of the ionic type. This question in itself is of considerable interest, as is evidenced by the increasing rate of publications on this subject. Recently, in the investigation of positron annihilation in ionic crystals, a number of new effects were observed, for example, an influence of the type of cation on the half-width of the angular correlation curves of the annihilation radiation in oxides of metals. In addition, it was found that in a number of oxides, a considerable fraction of the positrons forms positronium atoms, and in the case of finely-dispersed samples the temporal annihilation spectra reveal a component that differs little in duration from the lifetime of orthopositronium in vacuum. It was also observed that irradiation of ionic crystals by protons, γ rays, etc. leads to formation of new bound positron states. These effects are being intensely investigated at the present time.

The main experimental methods of observing annihilation of positrons in matter are as follows:^[1-8]: measurement of the positron lifetime, measurement of the relative counting rates of coincidence of three γ quanta, and observation of the angular correlation of the annihilation γ quanta. We consider in succession the results obtained by these methods.

1. AVERAGE LIFETIMES OF POSITRONS IN IONIC CRYSTALS

The lifetimes of positrons in ionic crystals were investigated in^[1,9-20]. At first^[1] only annihilation of positrons within a very short time, 10^{-10} sec, was discovered in ionic crystals. More careful investigations^[9,29], however, using apparatus with high resolution, led to the discovery of a multicomponent structure of the temporal distribution of positron annihilation in ionic crystals. We list the main results of these investigations:

a) **Metal hydrides**^[9-11,20]. The spectrum of the positron lifetimes in metal hydrides contains up to three components (Fig. 1). Gainotti et al.^[11] have established that in hydrides of alkali and alkaline-earth metals there takes place the relation

$$\frac{1}{\tau_i} = A_i + B_i n, \tag{1}$$

where n (cm^{-3}) is the molecular density of the hydrides, and A_i and B_i are parameters that are different for different groups of the metal ions. This relation is illustrated in Figs. 2 and 3.

b) **Metal halides**^[9,12-18]. A detailed study of the temporal distribution of the positron lifetimes in halides of metals of groups I and II has led to the observation of up to five components of the decay in the

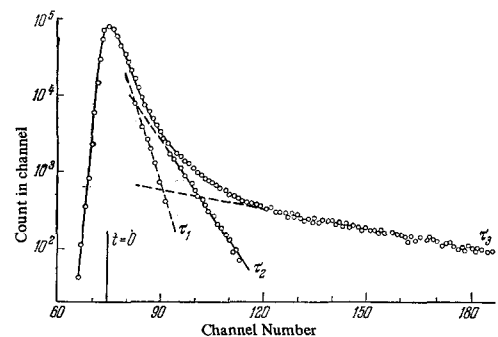


FIG. 1. Distribution of the lifetime of positrons in strontium hydride^[11] ($\tau_1 = 3.03 \times 10^{-10}$ sec, $\tau_2 = 6.91 \times 10^{-10}$ sec, $\tau_3 = 5.31 \times 10^{-9}$ sec).

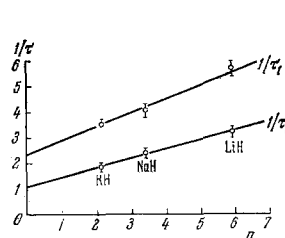


FIG. 2

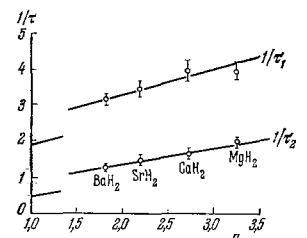


FIG. 3

FIG. 2. Positron annihilation rate τ^{-1} (10^9 sec^{-1}) against the molecular density n (10^{22} cm^{-3}) of alkali-metal hydrides^[11].

FIG. 3. Positron annihilation rate τ^{-1} (10^9 sec^{-1}) against the molecular density n (10^{22} cm^{-3}) of alkaline-earth metal hydrides^[11].

temporal spectra. By way of an example, Fig. 4 shows the positron lifetime spectrum of cesium bromide. It was established that the characteristics of the temporal spectrum (the lifetimes and intensities of the components) are determined mainly by the type of the halide anion and depend little on the type of the metal cation. The lifetimes τ_1 and τ_2 in alkali-halide crystals depend linearly on the cube of the radius of the anion, and the intensity (I_1) of the component τ_1 is a linear function of the square of the radius of the anion (Figs. 5 and 6). Singh et al.^[14] believe, however, that the lifetime τ_2 in alkali-halide crystals is determined not by the volume of the anion, but more readily by the ratio of the volume occupied by the anions in the crystal to the interstitial volume.

An analysis of the properties of the temporal spectra in halides of copper, silver, gold, and thallium has led the authors of^[15] to the conclusion that the annihilation rate ($1/\tau_1$) corresponding to the short-lived component increases linearly with increasing density of the halides of the foregoing metals, whereas the annihilation rate $1/\tau_2$ is practically independent of their density (Fig. 7). In addition, a correlation was observed between the intensity of the component τ_2 and the degree of ionicity of the chemical bonds in these compounds (Fig. 8).

Williams and Ache^[18] observed the appearance of a long-lived component (8.2×10^{-10} sec) with intensity 28% in the temporal spectra of annihilation in sodium halides (NaF and NaCl) bombarded with protons and γ rays. After thermal annealing of the bombarded samples, its intensity decreased strongly. It is therefore natural to assume that the origin of the long-lived component in bombarded sodium halides is connected directly with radiation defects.

c) Metal oxides^[16,19-29]. Even the first investigations of the temporal spectra in oxides of alkaline-earth metals have shown the presence of a component $\tau_2 \approx 3-5$ nsec. More careful measurements^[16,20] have made it possible to establish that the temporal spectra in oxides of certain metals contain, besides the component τ_2 , also a much longer-lived component with $\tau_3 = 50$ nsec. Measurements under vacuum conditions with powders of SiO_2 , MgO , and Al_2O_3 ^[21] have shown for τ_3 an unprecedented large value, which did not differ in practice from the calculated lifetime of atoms of orthopositronium in vacuum, namely $\tau_t^0 = 140$ nsec. The presence of long-lived components τ_2 and τ_3 in powders of BeO , MgO , SiO_2 , and Al_2O_3 was confirmed also in our investigation^[22]. It was also found that there was no τ_3 component in single-crystal and in sintered oxides (Fig. 9). The τ_3 component also drops out when the pores of highly dispersed oxide samples are filled with liquid^[23]. In powders of oxides, the intensities of the components τ_2 and τ_3 depend strongly on the per-unit surface area^[21], on the temperature, and on the pressure^[24]. A detailed investigation of the temporal spectra of annihilation in metal oxides carried out by Sen and Patro^[25,26] has made it possible to establish the presence of the components $\tau_2 \sim 10^{-9}$ sec and $\tau_3 \sim 10^{-8}$ sec in many other powders of metal oxides, the influence of air, moisture, and adsorbed gases on the lifetimes, and to a considerable degree on the intensity of the components τ_2 and τ_3

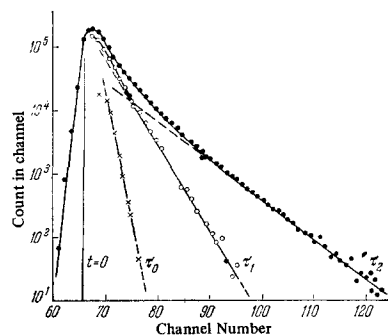


FIG. 4. Distribution of the lifetimes of positrons in cesium bromide^[13] ($\tau_0 = 1.26 \times 10^{-10}$ sec, $\tau_1 = 3.39 \times 10^{-10}$ sec, $\tau_2 = 6.86 \times 10^{-10}$ sec).

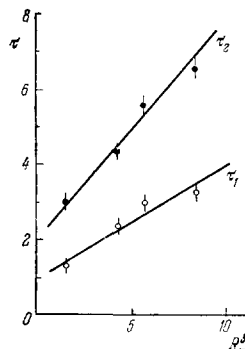


FIG. 5

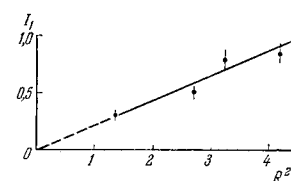


FIG. 6

FIG. 5. Average lifetime τ (10^{-10} sec) of positrons in lithium halides as a function of the cube of the anion radius R_- (\AA)^[13].

FIG. 6. Intensity of the component τ_1 of the temporal spectrum in lithium halides as a function of the square of the radius of the anion R_- (\AA)^[13].

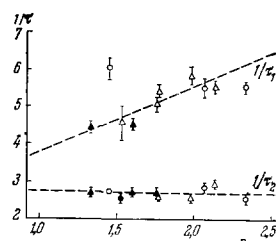


FIG. 7

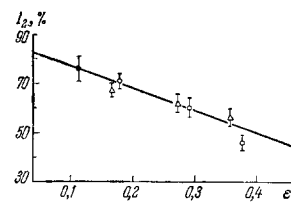


FIG. 8

FIG. 7. Positron annihilation rate τ^{-1} (10^9 sec^{-1}) against the molecular density n (10^{22} cm^{-3}) of halides of: Δ —copper; \circ —silver; \bullet —gold; \blacktriangle —thallium^[15].

FIG. 8. Intensity of the component τ_2 of the temporal annihilation spectrum as a function of the degree of ionicity (ϵ) of the chemical bonds in halides of copper (Δ), silver (\circ) and gold (\bullet)^[15].

and the two-quantum nature of the component τ_3 . The conclusion that the surface state has an effect on the characteristics of the long-lived components of the temporal annihilation spectra was confirmed by the results of Gol'danskii et al.^[27], who observed a noticeable influence exerted on these characteristics by the concentration of the OH groups on the surfaces of the oxides. The results of the foregoing investigations undoubtedly point to the formation of positronium atoms in powders of the oxides of many metals, the characteristics of the annihilation of which depend strongly on the state of the volume and the surface of the pores.

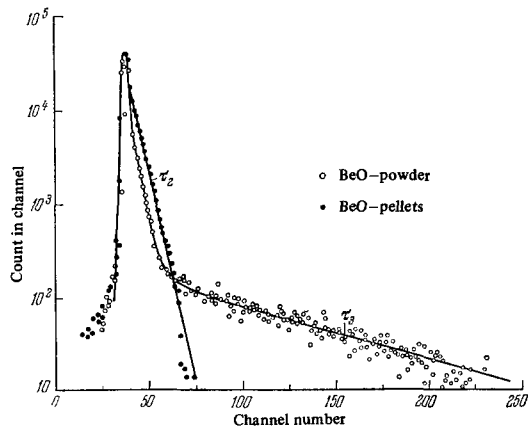


FIG. 9. Curves showing the distribution of the lifetimes of positrons in powdered and sintered beryllium oxide [22] ($\tau_2 = 4.5$ nsec, $\tau_3 = 80$ nsec).

The authors of^[26] measured the lifetime of the triplet positronium in certain oxides and observed agreement with the theoretical value $\tau_t^0 = 140$ nsec.

Interesting results were obtained^[28] by measuring the lifetime spectrum of the positrons in V_2O_3 in the temperature region 80–300°K, in which a semiconductor-metal transition takes place. A complicated temporal spectrum of annihilation was observed in V_2O_3 and was attributed by the authors of^[28] to annihilation of positrons from different positron band states. It turned out here that the lifetimes $\tau_1 = 1.94 \times 10^{-10}$ sec and $\tau_2 = 4.74 \times 10^{-10}$ sec remained constant in the investigated temperature region, thus directly indicating that the positron and electron wave functions are localized.

Besides hydrides, halides, and oxides, the lifetimes of the positrons were measured^[29] in certain compounds of the III-V and II-VI type. From the point of view of the characteristic temporal annihilation spectra, the III-V compounds (for example, AlN, AlP, InSb, BP) are similar to metals and elementary semiconductors, while II-VI compounds (CdTe, ZnTe, ZnSe) turn out to be closer to ionic compounds.

2. RELATIVE COUNTING RATE OF THREE-PHOTON COINCIDENCES

Experiments on the measurement of the temporal annihilation spectrum of positrons have shown that the annihilation process in ionic crystals has a complicated character. In particular, the presence of a long-lived component points to the possible formation of a positronium atom in ionic crystals. One of the essential proofs of the existence of positronium in matter is the determination of the effective cross sections of two- and three-photon annihilation. It is known^[30] that for free particles the ratio of the cross sections for three-quantum annihilation and two-quantum annihilation $\sigma_{3\gamma}/\sigma_{2\gamma} = 1/372$. In the case when positronium is produced in the medium, the ratio $\sigma_{2\gamma}/\sigma_{3\gamma}$ is much smaller than 372. From the known values of τ_2 and I_2 (in the absence of a second long-lived component^[22]), due to the formation of positronium, it is easy to calculate the relative probability of three-photon emission:

$$P_{3\gamma} = I_2 \frac{\tau_2}{\tau_1} + \left(1 - \frac{4}{3} I_2\right) \frac{1}{372}. \quad (2)$$

For $I_2 = 0$ (free annihilation), the probability $P_{3\gamma}$ is minimal and is equal to $P_{3\gamma}^0 \approx 0.27\%$.

An investigation of the counting rate of the coincidences of three γ quanta in ionic crystals was carried out in^[19,31-33]. It was established that for alkali-halide crystals with a lattice constant larger than 6 Å, the probability of three-quantum annihilation is much higher than 0.27%, and that this probability increases with increasing lattice constant. Even more surprising results were obtained for oxides of alkaline-earth metals, where the ratio $P_{3\gamma}/P_{3\gamma}^0$ reaches 22.4 (BeO). In addition, it turned out that the anomaly of three-quantum annihilation in MgO disappears on going from a polycrystal (powder) to a single crystal. The observed anomalies of 3γ annihilation demonstrate the possible existence of positronium atoms in ionic crystals.

The formation of positronium in matter can be revealed also by the magnetic quenching of orthopositronium*. Quenching of positronium by a magnetic field as a result of the ortho-para conversion should lead to a decrease of the 3γ -annihilation probability, to a weakening of the contribution of the long-lived component (I_2) of the temporal annihilation spectra, and to an enhancement of the narrow component (see below) of the correlation curves. Quenching of positronium was indeed observed in ionic crystals, but it turned out to be anomalously weak in the case of the KCl crystal^[12] (Fig. 10) and, conversely, anomalously strong in MgO powder^[35] (Fig. 11). Yet for other substances (for example, polymers and fused quartz) one observes^[6,34], as a rule, normal quenching of the positronium. Consequently, the properties of positronium in ionic crystals can differ greatly from those in other substances.

3. ANGULAR CORRELATION OF γ QUANTA IN TWO-PHOTON ANNIHILATION OF POSITRONS

Observation of the angular correlation of annihilation γ quanta makes it possible to assess directly the momentum distribution of the annihilating pairs (or electrons, if the motion of the positron can be neglected) and the formation of positronium atoms (from the appearance of the so-called narrow component in the correlation curves) in matter.

Measurements of the angular correlation of annihilation γ quanta in ionic crystals were carried out in^[36-66]. The following general laws in the properties of the angular distribution of annihilation radiation in ionic crystals of different classes were established:

a) Metal hydrides^[36,37]. The angular-correlation curves for hydrides of alkali metals (LiH and NaH) are shown in Fig. 12. The electron-positron density in the lithium hydride, calculated from these curves, deviates strongly from the electron density in the free ion H^- , unlike the case of alkali halide crystals^[41]. The angular-correlation curve for cerium hydride is

*A summary of the formulas and the main results on magnetic quenching of orthopositronium in matter can be found in the reviews of Gol'danskii^[6,34].

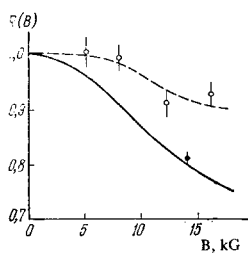


FIG. 10

FIG. 10. Magnetic quenching of long-lived component (I_2) of the temporal annihilation spectrum of positrons in KCl (O) and in lucite (●). Solid line—quenching for free positronium [12], ($R(B) = I_2(B)/I_2(0)$).

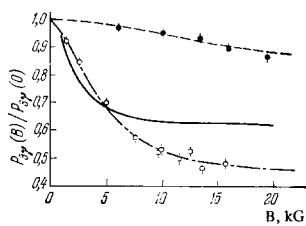


FIG. 11

FIG. 11. Magnetic quenching of three-photon annihilation of positrons in MgO powder (O) and in lucite (●). Solid line—quenching for free orthopositronium [35].

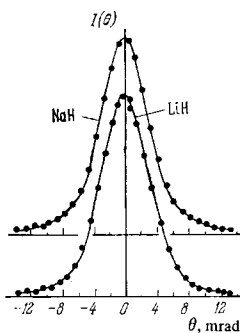


FIG. 12. Angular correlation curves for annihilation pairs of γ quanta in lithium and sodium hydrides [36].

much broader than for lithium hydride and for metallic cerium, and is the broader the larger the hydrogen concentration. Apparently, the half-width of the correlation curves in metal hydrides, like the lifetimes of the positrons (see Chapter 1), depends on their density.

b) Metal halides [$^{38-46}$]. Angular correlation curves in alkali-halide crystals have a form close to "triangular" (Fig. 13). The halfwidth of the correlation curves in these compounds depends on the nature of the anions but not that of the cations [39,41,42] (Table I); in halides of the same alkali metal, the correlation curves become narrower on going from fluorine to iodine. Thus, in sodium halides this narrowing amounts to almost 35%. However, in halides of other metals one can apparently expect deviations from these simple regulari-

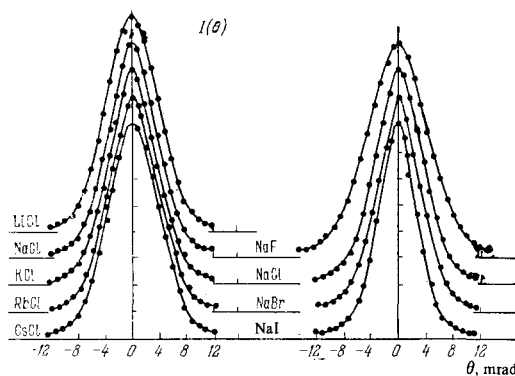


FIG. 13. Angular correlation curves of annihilation pairs of γ quanta in alkali-halide crystals [41].

ties (compare, for example, CuCl_2 and AgI with the chlorides and iodides of the alkali metals in Table I). In addition, a broadening of the correlation curves in KI was observed [46] on going from a polycrystalline sample to a single crystal ($\sim 7\%$). Careful measurements [43,44,46] have made it possible to observe anisotropy of the angular distribution of the annihilation pairs of γ quanta in alkali-halide single crystals. Thus, in single-crystal NaCl and KI, the correlation curve is broader by 8% in the [110] direction than for the [100] direction. Finally, we note that in alkali-halide crystals the narrow component in the correlation curves, characteristic of annihilation decay of atoms of parapositronium, is not observed.

c) Metal oxides [$^{22,46-62}$]. For this class of crystals the form of the correlation curves deviates noticeably from the "triangular" form characteristic of alkali-halide crystals (compare, for example, Figs. 13 and 14). Another difference lies in the appreciable variations of the half-widths of the angular-correlation curves on going from one oxide to the other [22,52] (see Table I). The difference in the half-widths of the correlation curves is also observed in a number of oxides of alkaline-earth metals having a structure of the same type [53]. Moreover, the half-width does not remain constant even in the series of oxides of the same metal, as is confirmed with iron oxides as an example (sintered FeO , Fe_2O_3 , and Fe_3O_4) [54]. The observed behavior of the half-width of the correlation curves in oxides can apparently be attributed to the change in the

Table I. Angular-correlation curves of annihilation pairs of γ quanta in sintered BeO and MgO and single crystals of Al_2O_3 and SiO_2 [22]

Crystal	$\Gamma, 10^{-3}$ rad	Crystal	$\Gamma, 10^{-3}$ rad	Crystal	$\Gamma, 10^{-3}$ rad	Crystal	$\Gamma, 10^{-3}$ rad
LiH	7.4	NaBr	7.4	SiO_2 ***	12.8	ZnO	12.2
NaH	7.1	NaI	6.8	V_2O_5	11.4	La_2O_3	11.7
LiF	12.0	KI	7.4	Cr_2O_3	14.8	Ce_2O_3	13.4
NaF	10.5	β -AgI	8.6	MnO	13.0	SnO_2	14.4
LiCl	9.2	BeO **	13.8	FeO *	12.7	PbO	9.6
NaCl	8.6	MgO **	12.8	Fe_2O_3 *	14.3	Na_2S	8.3
KCl	8.3	CaO	14.4	Fe_3O_4 *	13.7	FeS	10.3
RbCl	8.6	SrO	14.2	Co_2O_3	13.0	HoSb	8.5
CsCl	8.9	BaO	12.0	NiO	12.2	HoTe	8.6
CuCl_2 *	11.6	Al_2O_3 ***	15.3	CuO	12.4		

*Measurement by the authors.

**Sintered.

***Single crystals.

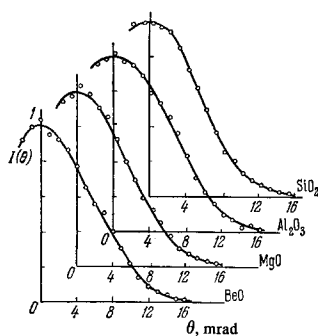


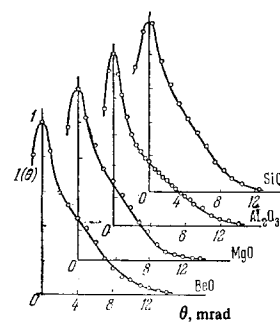
FIG. 14. Angular-correlation curves of annihilation pairs of γ quanta in sintered BeO and MgO and single crystals of Al_2O_3 and SiO_2 [22].

character of the chemical bonds in them when one cation of the metal is replaced by another (or when the degree of oxidation of the metal atom changes) (see Chapter 4). This assumption is also favored by such facts as the correlation between the half-width of the correlation curves and the position of the K edge of the x-ray absorption spectrum in iron oxides^[54] and with the position of the Mossbauer absorption line in alkali-silicate glasses^[61].

Interesting results were obtained in the study of the angular distribution of annihilation pairs of γ quanta in powdered metal oxides^[22,48,50,58]. In the correlation curves and in powders of certain oxides (BeO, MgO, Al_2O_3 , and SiO_2) there is observed an intense narrow component ($I_N = 10\text{--}20\%$) (Fig. 15), whereas in single crystals or in sintered samples of the same oxides, there is no narrow component. A comparison of the correlation curves plotted for the powders of MgO, ZnO, and Al_2O_3 in an oxygen atmosphere and in vacuum has shown^[50] that the intensity of the narrow component of the correlation curves in evacuated samples is greatly reduced. An intense narrow component is observed^[47,57-60] also in the correlation curves for monolithic samples of certain glasslike oxides (for example, SiO_2 , B_2O_3 , and GeO_2). The introduction of oxides of other metals into these glasses greatly decreases the intensity of the narrow component and can lead even to its complete vanishing^[61,62]. We note also that the halfwidth of the narrow component in the correlation curves decreases to almost one-half when the oxides go over from the highly-dispersed state to the glasslike state^[58]. This narrowing of the narrow component is analogous to a certain degree to the narrowing of the narrow component observed in the water-ice transition^[60]. The facts listed above indicate directly the formation of positronium atoms in oxides of certain metals in the highly-dispersed and in the glasslike states.

The use of a powerful source of positrons (1-6 Ci) and of high angular resolution of the apparatus (0.6 mrad) has enabled the authors of^[55,56] to observe the narrow component in the correlation curves of quartz single crystals ($I_N = 1\text{--}5\%$); this component could not be observed in all the preceding investigations. It is interesting that the long-lived component is missing from the temporal annihilation spectrum, a fact that can be explained, in the presence of formation of positronium atoms, as being due to the very high rate of the pick-off annihilation of orthopositronium. The irradiation of crystals in quartz by electrons of energy 4 MeV leads to weakening of the narrow com-

FIG. 15. Angular-correlation curves of annihilation pairs of γ quanta in powders of the oxides BeO, MgO, Al_2O_3 , and SiO_2 [22].



ponent (Fig. 16).^[57] In addition, experiments^[57] on the influence of neutron bombardment on the angular correlation of annihilation γ quanta in crystalline and fused quartz have revealed the important role played by the density factor in the formation of positronium in quartz, thus contradicting the conclusions of Colombino et al^[47].

d) Sulfides, selenides, tellurides, etc.^[45,46,51,63-66]

There are at present no systematic data for these compounds. Attempts were made^[45,46] to establish regularities in the behavior of the angular distribution of the annihilation radiation in crystals of isoelectronic series. For crystals with ion configurations of the neon-neon type (NaF, MgO, AlN, and SiC), no monotonic change in the half-width of the correlation curves was observed (the half-widths were respectively 9.7, 12.1, 11.1, and 12.0 mrad)^[46]. This fact is connected by the authors of^[46] with the corresponding change in the ionic charge on going from one compound to the other. However, such a comparison can hardly be applied to ionic crystals having different negative ions. For the isoelectronic xenon-xenon series (α -Sn, InSb, CdTe, and β -AgI), there is observed^[45] a change in the form of the angular-correlation curves, from the parabolic form (α -Sn) characteristic of metals to a form typical for ionic crystals (β -AgI). In chalcogenide compounds (As_2S_3 , As_2Se_3 , GeS_2), as well as for oxides^[62], there is observed^[66] a narrowing of the correlation curves and the absence from them of a narrow component in the crystal-glass transition.

Thus, by now numerous experiments have been carried out on the observation of the main characteristics of annihilation of positrons in ionic crystals, making it possible to conclude that the annihilation process has a complicated character. The explanation

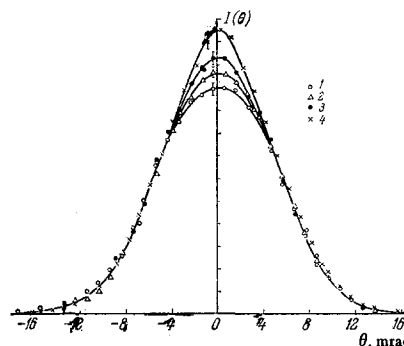


FIG. 16. Angular correlation curves in crystalline quartz bombarded by fast neutrons (1—not bombarded; 2— 6.2×10^{19} , 3— 9.3×10^{19} , and 4— 2.2×10^{20} neutrons/cm²) [57].

of the experimental data entails the need for analyzing the states of the positrons in the crystal. An important circumstance in this case is that the interaction of the moving positrons with the atoms of the ionic crystals leads to their thermalization. Elementary estimates show^[67,68] that the positron thermalization time in ionic crystals is $\sim 10^{-12}$ sec, whereas the shortest lifetime against annihilation is $\sim 10^{-10}$ sec. It follows therefore that the main characteristics of the annihilation depend on the interaction of the thermalized positrons with the medium, thereby greatly simplifying the problem.

4. POSITRON STATES IN IONIC CRYSTALS

An analysis of the experimental data on positron annihilation shows that in ionic crystals there can apparently be realized two main types of states from which the positrons annihilate*: bound positron states of the non-positronium type (compounds of positrons with negative ions^[3,13,41,51-53,67,69-73] polaron states of the positron^[74-76], positrons captured by point defects^[75,77,78], and positrons bound in the positronium atoms^[22,51,58,79-86]. Let us consider in sequence the annihilation of the positrons from these states.

Starting with the work of de Benedetti et al.^[87], it has been customary to assume that in ionic crystals the thermalized positrons are localized near the anions, forming quasiatomic e^+ + anion systems. Favoring this assumption is the fact that the lifetime of the positrons and the angular distribution of the annihilation radiation in alkali-halide crystals are practically independent of the type of the metal cation. To calculate the annihilation characteristics of the e^+ + anion system, several models were proposed, the main difference between which lies in the choice of the positron wave function $\chi(r)$. The relation $\theta_c = 15.53/a$ (θ_c is the "maximum" angle, the meaning of which is seen from Fig. 17), which follows from the Ferrel model^[3] ($\chi(r) = (4\pi)^{-1/2} \delta(r-a)$, where a is the Goldschmidt radius of the anion) is well satisfied in a number of cases^[38,45,64,65]. However, Ferrel's calculations did not result in agreement with experiment in the form of the correlation curves and in the estimates of the dependence of the lifetime (τ) of the positrons on the radius of the anion (in the indicated approximation we should expect a "triangular" form of the correlation function, and a proportionality $\tau \propto a^3$). In the model of Stewart and Pope^[41] ($\chi(r) = \text{const}$ in the region of the outer shells of the anions), the half-width of the correlation curves does not depend^[52] on the type of the cation, and this is seemingly confirmed with alkali-halide crystals as an example, but for other compounds (for example, oxides), a deviation from this rule was observed (see Table I). Satisfactory agreement between theory and experiment is obtained in the model of the "optical" positron^[71] ($\chi(r) \sim r \exp(-\alpha r)$ ^[88], where $\alpha = 3\eta/2$, and η is the effective charge of the anion in the crystal). In this model, the half-width of the correlation curves (Γ)

*In media of the ionic type, having a high concentration of negative ions, as shown by calculation^[72], the probability of annihilation of free positrons is negligibly small compared with the probabilities of two-photon annihilation of the bound positrons.

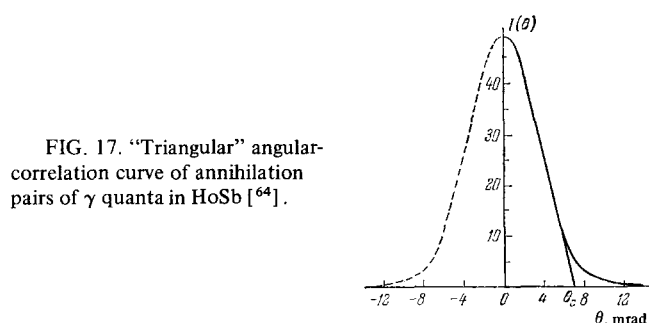


FIG. 17. "Triangular" angular-correlation curve of annihilation pairs of γ quanta in HoSb^[64].

and the lifetime of the positrons (τ) turn out to be connected with the magnitude of the effective charge of the anion in the crystal. In the approximation of the Slater wave functions of the electrons of the external n_s, p shell of the anion (see, for example,^[89]), this connection is given by the following expressions^[71]:

$$\Gamma_{n_s, p} = C_{n_s, p} (\alpha + \beta) \text{ mrad} \quad (3)$$

and

$$\tau_{n_s, p} = 2,5 \cdot 10^{-10} C'_{n_s, p} \frac{(\alpha + \beta)^{2n^* + 3}}{\alpha^3 \beta^{2n^* + 1}} \text{ sec}, \quad (4)$$

where $C_{n_s, p}$ and $C'_{n_s, p}$ are constants that depend on the number n, β is the argument of the Slater exponentials, and n^* is the effective principal quantum number. The effective anion charges calculated^[52-54,61,62,66,90] within the framework of the considered model agree well, as a rule, with the data of other methods (see, for example, Tables II and III). Agreement is observed also in the form of the correlation curves (Fig. 18). The charges for alkali-halide crystals calculated from formulas (3) and (4) are also in good agreement (to carry out such a comparison in other compounds (oxides, sulfides, etc.) it would be desirable to carry out in the future measurements of the shortest-lived components of the temporal annihilation spectra in these compounds; hardly any such

Table II. Effective charges of anions in ionic crystals, calculated from the short lifetime of the positrons^[13,15,17]

Anion	Substance	Positron annihilation	Spighetti method ^[91]
F ⁻	LiF	0,78	0,87
	NaF	0,68	0,93
	CaF ₂	0,69	0,74
	SrF ₂	0,70	0,65
	BaF ₂	0,70	0,70
Cl ⁻	NaCl	0,61	0,74
	KCl	0,66	0,80
	RbCl	0,79	0,84
	TlCl	0,79	1,08
	CuCl	0,84	1,10
O ²⁻	SiO ₂ Crystal	0,68 *)	0,98 **)
	SiO ₂ (fused)	0,56 *)	

*The charge was calculated by using the lifetime of positrons in quartz, as measured by Bell and Graham^[92].
 **)For quartz we used the charge value obtained by x-ray-spectral methods^[92].

Table III. Effective charge of oxygen ion in oxides of alkaline-earth metals, calculated from the half-width of the correlation curves (see Table I)

Oxides	Positron annihilation ^[52] , $\eta, -e$	Other methods* ^[91,92] , $\eta', -e$
BaO	1,14	1,1
MgO	0,99	1,01
CaO	1,23	1,18
SrO	1,20	1,22
BaO	0,85	0,85

*The values closest to η were chosen from the values of η' listed in the review^[91].

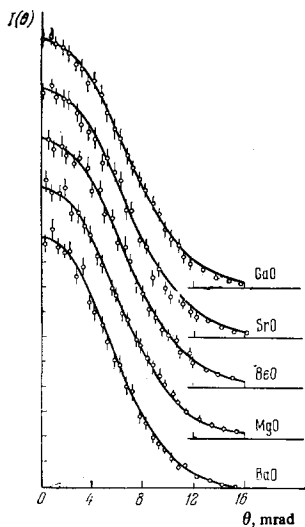


FIG. 18. Comparison of calculated (solid line) and experimental (circles) angular correlations for alkaline-earth-metal oxides [52].

measurements have been made to date). This gives grounds for assuming that the annihilation of the positrons can be used to determine the effective charges of the anions in media of the ionic type.

The models considered above for the e^+ + anion system do not explain fully the multicomponent structure of the temporal spectra of the annihilation. Such an explanation can be given in part^[13,58,59,61] by starting from the expected properties of the ground and excited states of the e^+ + anion system. We note also that a description of the annihilation process on the basis of the properties of the e^+ + anion system is not applicable to metal hydrides for which the dimensions of e^-H^- greatly exceed^[94] the lattice constant, in distinction from halides^[13] and oxides^[53]. The dependence of the average lifetimes of the positrons on the hydride density confirms this conclusion. A calculation of the characteristics of this case (for LiH) was given by Brandt et al.^[94].

In ionic crystals having defects (interstitial ions, vacancies), capture of positrons by defects is possible^[75,77,78] with formation of positron centers, such as, for example, e^+ + interstitial anion, positron in the cation vacancy (F^+ center), positron and electron (Ps atom) in the cation or anion vacancy (F^- center). An analysis of the results of experiments with irradiated alkali-halide crystals (see Chapter 1) and quartz (see Chapter 3) has led the authors of^[18,55,56] to the conclusion that F^- centers are produced in these crystals. The appearance of a narrow component ($I_N \approx 5\%$) in the correlation curves for an LiF crystal bombarded by γ rays from Co^{60} (dose 10–20 Mrad)^[95] was explained by us likewise as being due to the formation of F^- centers. An analysis of the kinetics of formation of F^- centers in the crystal shows that the concentration of the defects in the crystal and the cross section for the production of the F^- centers (σ_F) can be determined from the intensity of the long-lived component of the temporal annihilation spectrum and from the narrow component in the correlation curves. According to the estimates of^[95] we have $\sigma_F \sim 10^{-15} \text{ cm}^2$ for the LiF crystal.

The results of the experiments (see Chapters 1–3) have shown convincingly that positronium can be pro-

duced in ionic crystals, in contradiction to the opinion of Ferrel^[3] and Wallace^[4]. The question of the probability of formation of positronium in ionic crystals has not yet been finally resolved. It is possible that this probability is connected with the width of the forbidden band of the crystal^[86]. The facts reported in Chapters 1–3 indicate clearly that besides the energy factor, an important role in the existence of positronium in ionic crystals (and glasses) is played by the geometric factor. The formation of positronium atoms is observed, as a rule, only in the presence of defects (vacancies, pores, regions of decreased density, etc), thus indicating that these atoms are localized in defects. Brandt and Paulin^[96,97] propose that positronium atoms are produced in oxide powders in the interior of the particles, and that some of these atoms become annihilated during the process of diffusion, while another part falls into the volumes of the pores. According to estimates by these authors, the diffusion coefficient of orthopositronium atoms in SiO_2 is $\sim 2.6 \times 10^{-4} \text{ cm}^2/\text{sec}$, and the mean free path to annihilation inside the particles is $\sim 46 \text{ \AA}$, whereas, for example, in MgO we have respectively $\sim 14 \text{ cm}^2/\text{sec}$ and $\sim 270 \text{ \AA}$.

A very important question is that of the states, mechanisms, and kinetics of the loss of the positronium atoms in pores. It was assumed earlier that the positronium atoms are lost only in the volume of the pores. If the pores were filled with oxygen (or air), then the orthopositronium atoms would experience an ortho-para conversion on the paramagnetic molecules of the oxygen, as is manifested^[22] in experiment (See chapters 2 and 3) by an enhancement of the narrow component of the correlation curves, an anomalously strong magnetic quenching of the orthopositronium, and an increased rate of 3γ annihilation. However, experiments on the influence of the chemical composition of the oxide surfaces and the temperature on the characteristics of the long-lived components of the temporal annihilation spectra give grounds for assuming^[86] that some of the positronium atoms "stick" to the walls of the pores and form the surface states of positronium. An investigation of this question is so far in the initial stage, but even now one can advance the hypothesis^[27,86,88] that there are prospects for using the method of positron annihilation to study chemisorption and adsorption phenomena in porous systems.

As already noted (see Chapter 3), the half-width of the narrow component, i.e., the average kinetic energy of the parapositronium atoms, depends on the state of the oxides. This dependence can be attributed^[58] to differences in the dimensions of the defects if the positronium atom in the defect is regarded as a particle in the potential well of finite depth (as is usually assumed^[96] in the "free" volume models). By solving the inverse problem, it becomes possible^[58] to estimate the dimensions of the defects.

As to the anomaly of the magnetic quenching of orthopositronium and 3γ annihilation in alkali-halide crystals (see Chapter 2), qualitatively these anomalies can be explained by means of the exciton model^[6,82-84] of the positronium atom in the crystal. The indicated anomalies can be also explained differently, namely,

as being due to the effect of "compression" of the positronium atom in the crystal. Our estimates within the framework of the model of Sommerfeld and Welker^[100] show (see also^[99]) that the "compression" of the positronium atoms should lead to a decrease of its binding energy, to a shortening of its lifetime, to an increase of its kinetic energy (and consequently to a broadening of the narrow component^[60]), to anomalously weak magnetic quenching of the orthopositronium, and to a increased rate of 3γ annihilation.

Note added in proof. We present another list of recent papers^[101-120] which describe even more completely the complexity of the process of annihilation of positrons in ionic crystals:

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