

FORMATION OF POSITRONIUM IN 1S AND 2S STATES IN OXIDES

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An investigation of the annihilation of positrons in oxides of alkali-earth metals has revealed the presence of intense long-lived components  $\tau_2$  and  $\tau_3$  in the annihilation time spectra and a clearly pronounced narrow component in the angular distribution of the annihilation quanta in powdered BeO and MgO [1, 2]. It can therefore be concluded that an appreciable fraction of the positrons in these oxides form positronium atoms. The positron annihilation characteristics in SrO and BaO differ greatly in this series. Other oxides behave analogously, in some oxides, judging from the time spectra, positronium is produced (SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> [2-4]), whereas for a large group of oxides investigated by the method of angular correlation in [5], no such conclusion can be drawn.

These interesting facts call for an explanation. Notice should be taken in this connection of the definite correlation existing between the possibility of positronium formation and the width of the forbidden band of the oxides. Indeed, as seen from the table, in those oxides for which intense positronium formation has already been observed, the width of the forbidden band  $E_g$  exceeds 6.8 eV (the binding energy of the Ps-atom ground state in vacuum).

Forbidden band width  $E_g$  and fraction  $P_{Ps}$  of positronium formation for some oxides

Oxide	$E_g$ , eV [6]	$P_{Ps}$ , %	Oxide	$E_g$ , eV [6]	$P_{Ps}$ , %
BeO	10.4	16.5	PbO	2.3	*
MgO	8.7	16.0	Y <sub>2</sub> O <sub>3</sub>	2.1	*
CaO	7.5	4.4	La <sub>2</sub> O <sub>3</sub>	2.6	*
SrO	5.7	*	NiO	2.0	*
BaO	4.4	0.9 [7]	ZnO	3.3	*
SiO <sub>2</sub>	8-11	35	CuO	1.4	*
Al <sub>2</sub> O <sub>3</sub>	7.3	25			

\* - no intense Ps formation is observed (according to temporal or correlation measurement data).

Judging from the data in [6], B<sub>2</sub>O<sub>3</sub> ( $E_g = 9.0$  eV) is among the oxides with  $E_g > 6.8$  eV not listed in the table. It was therefore of considerable interest to measure the characteristics of positron annihilation in this oxide. We measured the positron lifetime spectrum in granulated B<sub>2</sub>O<sub>3</sub> powder (of "specailly pure" grade) using a setup with time resolution 1.0 nsec, in the range up to 10 nsec. The obtained time spectrum revealed the presence of an intense long-lived component with  $\tau_2 = 1.7 \times 10^{-9}$  sec (see Fig. 1a). It follows therefore that, in accord with the already noted correlation, positronium is produced in B<sub>2</sub>O<sub>3</sub>. On the other hand, in the mercury-oxide investigated by us, which has a narrow forbidden band ( $E_g = 1.0 - 1.2$  eV), no long-lived component  $\tau_2$  was observed. (Fig. 1). Thus, none of the hitherto

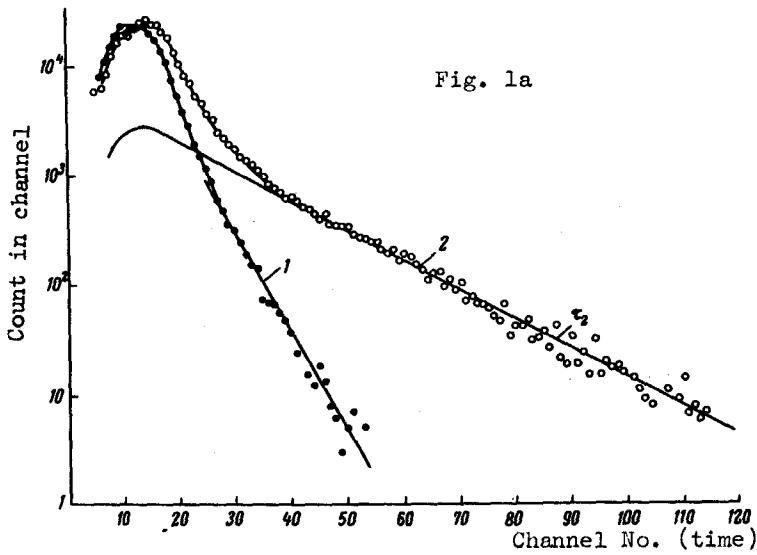


Fig. 1a

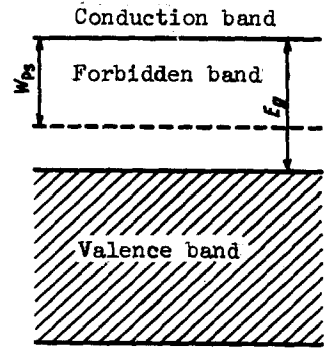


Fig. 2

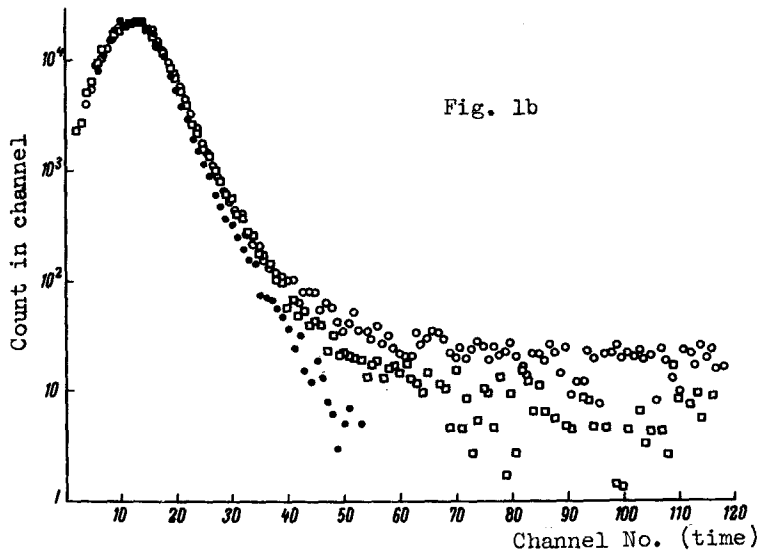


Fig. 1b

Fig. 1. Time spectra of positron annihilation in oxides (the random-coincidence background has been illuminated; the channel value is  $h = 1.04 \times 10^{-10}$  sec): a) 1 - HgO (pure), 2 - B<sub>2</sub>O<sub>3</sub> (specially pure);  $\tau_2 = (1.7 \pm 0.1) \times 10^{-9}$  sec,  $I_2 = 18 \pm 2\%$ . b) ● - HgO, □ - La<sub>2</sub>O<sub>3</sub>, ○ - Sb<sub>2</sub>O<sub>3</sub> (all "pure").

Fig. 2. Energy scheme of formation of thermalized positronium in oxides.

investigated oxides contradict the indicated correlation. This correlation is apparently not an accident. From the point of view of the band theory, the existence of a thermalized positronium atom is possible only in those oxides, for which the positronium energy level falls in the forbidden band (Fig. 2), since the positronium must be regarded as an impurity atom, and its energy level lies in the forbidden band at a depth  $W_{Ps}$  below the bottom of the conduction band. The binding energy  $W_{Ps}$  of the electron and positron in the Ps atom in the 1S state may also differ from 6.8 eV, owing to the polarizing action of the medium. The kinetic energy of the Ps atom in the oxide cannot exceed  $W_{Ps} = (6.8 - \Delta)$  eV ( $\Delta$  is the correction due to the polarizing action of the medium), for otherwise it will dissociate into an electron and a positron upon colliding with the lattice atoms. Consequently, the criterion for positronium formation can be written in the form

$$W_{Ps} \geq E_{e^+} + W_{Ps} - E_g \geq 0, \quad (1)$$

where  $E_{e^+}$  is the kinetic energy of the positron.

In first approximation, we put  $\Delta = 0$ . Then the criterion (1) at  $E_{e^+} < W_{Ps}$ , e.g., for thermalized positrons, can be given as

$$6.8 \text{ eV} \leq E_g$$

for the formation of positronium in the 1S state. Similarly,

$$1.7 \text{ eV} \leq E_g$$

for the formation of positronium in the 2S state.

It follows therefore that oxides with a forbidden band wider than 1.7 eV can serve as an object for the search of Ps atoms in the 2S state. It is necessary to bear in mind here that even if the excited Ps atoms are produced in the condensed phase, their experimental observation is difficult, for collisions with the atoms of the medium cause deactivation of the (2S)Ps within a time on the order of  $10^{-14}$  sec [8]. The deactivation time of (2S)Ps becomes larger than the proper lifetime  $\tau_2(^3S_1)$  only in gases at pressures below  $10^{-3}$  mm Hg, as demonstrated in [9]. In this sense, finely-dispersed powdered oxides are a particularly convenient object for the search of excited Ps atoms, since the ( $2^3S_1$ )Ps atoms produced on the surface or in the layer next to the surface ( $\sim 10$  Å thick) are able to go over into the free volume between the oxide particles. It is not excluded that the ( $2^3S_1$ )Ps atoms going into the free volume become annihilated with a larger lifetime, a possible upper limit of which is the proper lifetime  $\tau_2(^3S_1) = 1.12 \times 10^{-6}$  sec. Favoring this assumption are recent observations of the decay of ( $1^3S_1$ )Ps in powdered oxides ( $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{MgO}$ ) with a proper lifetime  $\tau_1(^3S_1) = 1.4 \times 10^{-7}$  sec [3, 4]. One can expect observation of 2S positronium atoms in measurements of the positron annihilation time spectra (in the time interval up to 10  $\mu\text{sec}$ ) in outgassed samples of powdered oxides having a forbidden-band width larger than 1.7 eV.

The data obtained by us on the positron annihilation time spectra in certain oxides offer indirect evidence of (2S)Ps formation. The positron annihilation spectra in powdered  $\text{HgO}$ ,  $\text{La}_2\text{O}_3$ , and  $\text{Sb}_2\text{O}_5$  are compared in Fig. 1b. We see that there is no intense  $\tau_2$  component in  $\text{HgO}$ ,  $\text{La}_2\text{O}_3$ , and  $\text{Sb}_2\text{O}_3$ . This agrees with the aforementioned assumption that (1S)Ps is not produced in oxides with a forbidden band  $E_g < 6.8$  eV ( $E_g$  for  $\text{HgO}$ ,  $\text{La}_2\text{O}_3$ , and  $\text{Sb}_2\text{O}_3$  the values of  $E_g$  are respectively 1.0 - 1.2, 2.6, and 4.2 eV [6]). A remarkable feature of Fig. 1b is that the counting rate of the delayed coincidences is larger in the region of larger times for  $\text{La}_2\text{O}_3$  and  $\text{Sb}_2\text{O}_3$  than in  $\text{HgO}$ . One cannot exclude the possibility that this increased counting rate is due to the presence of a rather long-lived component (perceived in our case as a constant plateau) connected with the formation of 2S positronium ( $E_g > 1.7$  eV), whereas  $E_g \leq 1.2$  eV in  $\text{HgO}$ , and the positronium is apparently not produced at all.

A preliminary analysis shows that the considerations advanced here concerning the possible formation of positronium may apply also to other substances (semiconductors, polymers, molecular crystals).

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ANGULAR ANISOTROPY OF THE FISSION OF  $Pb^{204}$  AND  $Pb^{208}$  BY ALPHA PARTICLES NEAR THE THRESHOLD

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Recently published results of the measurement of the angular anisotropy of the fission of  $Pb^{206}$  by alpha particles [1] have established that the energy gap in the spectrum of the internal excitations of the nucleus  $Po^{210}$  in the transition state is anomalously large,  $2\Delta_f = 4$  MeV. This is more than double the value  $2\Delta_g = 1.5$  for the equilibrium ground state, in which the  $Po^{210}$  nucleus is close to spherical. The contradictory character of the information [1-4] on the dependence of the parameter  $\Delta$ , which determines the role of the effect of paired nucleon correlation, on the nuclear deformation has stimulated us to perform similar measurements for the other even-even isotopes of lead,  $Pb^{204}$  and  $Pb^{208}$ .

The experiments were performed with the cyclotron of our institute, using a track procedure for registering the fission fragments. The experiments were so organized that simultaneous measurements could be made of the fission cross section  $\sigma_f$  and the angular distribution of the fragments  $W(\theta)$ . The alpha-particle energies were decreased to 29 - 38 MeV with the aid of aluminum foils. The accuracy with which the alpha-particle energy was measured was not worse than  $\pm 0.2$  MeV. The targets were prepared by an electrolytic method from separated  $Pb^{204}$  and  $Pb^{208}$ . The upper limit of the extraneous strongly-fissioning impurities was less than  $10^{-6}$  %, as estimated in reactor experiments, and guaranteed background-free measurements in the entire investigated energy range.

The results of the measurements of the angular anisotropy  $W(0^\circ)/W(90^\circ)$  of the fission of  $Po^{208}$ ,  $Po^{212}$ , and  $Po^{210}$  are shown in the figure as functions of the excitation energy in the transition state  $E^* = E_x - E_f$ , where  $E_x$  is the initial excitation energy of the compound nuclei and  $E_f$  is the height of the fission barrier. The values of  $E_f$  for  $Po^{208}$ ,  $Po^{210}$ , and  $Po^{212}$  were assumed equal to 19.8, 20.5, and 18.8 MeV, respectively (the last two according to [5], and for  $Po^{208}$  from the measured course of  $\sigma_f(E_x)$ ).

A comparison of the data shown in the figure reveals a striking difference in the energy dependence of  $W(0^\circ)/W(90^\circ)$  near threshold namely a sharp growth in the case of  $Po^{210}$ , and constancy in the case of  $Po^{208}$  and  $Po^{212}$ , similar to that observed for  $Tl^{201}$  and  $At^{213}$  in the  $(\alpha, f)$  reaction on gold in bismuth [6]. This discrepancy between the results of [1] and the aggregate of the experimental data for other nuclei may possibly be the consequence of an individual peculiarity of the  $Po^{210}$  nucleus and deserves a special study. All that matters to us here is that these difference in the course of the angular anisotropy should influence our ideas concerning the value of  $\Delta_f$ .

An estimate of the energy gap  $2\Delta_f$  in [1] was based on the following considerations. In