Anomalous Three-Quantum Decay of Positrons in Alkaline Earth Oxides*

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The decay of positrons annihilating in alkaline earth oxides was investigated through lifetime and three-to two-quantum relative yield measurements. It was found that in these materials positrons display an anomalous behavior: the time annihilation spectrum is complex, exhibiting a long-life component, and the abundance of three-quantum events is exceptionally large. The observed abundance exceeds the value that can be calculated assuming positronium formation by a factor up to 15.

INTRODUCTION

R ECENT investigations[1,2] have shown the existence of a long-life component in the time spectra of positrons annihilating in alkali halides and in lithium hydride. In alkali halides this component is characterized by a mean life ranging between 4×10^{-10} sec and 8×10^{-10} sec, and occurs with an intensity between 25 and 85%. A static magnetic field gives rise to a quenching of this component, whose amount, however, is appreciably lower than what would be expected if one attributes this component to the decay of a free positronium atom. In LiH, the long-lifetime component exhibits a relative intensity of 12% and a mean life 1.5×10^{-9} sec. No further data are available at present.

These results appear to indicate that in ionic crystals, positron-bound systems can be formed and persist for an appreciable time. The matter has been quite recently investigated theoretically by Neamtan and Verrall,[3] who found that in a LiH crystal the formation and the persistence of a positronium-like system is energetically favorable.

In what follows, we report experimental results on positron annihilation in alkaline-earth oxides, which give further support to the assumption of the existence of a bound system formed by positrons in crystals displaying an ionic bond.

EXPERIMENT AND RESULTS

The positron emitter used was ^{22}Na and the alkaline-earth targets were specimens of multicrystalline analytical-reagent-grade chemicals. For the analysis of the time distribution spectra a nanosecond vernier time-delay analyzer was employed; the experimental method is the same used in previous researches[2,4] and we shall not describe it here. The results obtained are collected in Table I, and a typical time spectrum is

TABLE I. Experimental data on the long-lifetime component. τ and I are the mean life and the percentage abundance of this component; p_3 is the fraction of the three-quantum events.

Oxide	$\tau \times 10^9$ (sec)	I (%)	$p_3 \times 10^3$
BeO	3.46 ± 0.26	6.8 ± 0.2	61.8 ± 3.8
MgO	4.86 ± 0.52	1.8 ± 0.1	23.3 ± 1.1
CaO	3.75 ± 0.28	3.2 ± 0.3	7.5 ± 1.1
BaO	3.20 ± 0.27	0.64 ± 0.07	4.2 ± 0.7

shown in Fig. 1. Besides the short-life component, the existence of a long-life component is evident.

The three-quantum relative yield was obtained by employing a method similar to that of the "valley-to-peak" ratio, which gives a measure of the relative importance of the annihilations from triplet and singlet states and which is conventional in researches of positron annihilation in gases.[5]

Figure 2 shows the γ spectrum in the region from 350 to 530 keV obtained with the active sample sandwiched in BeO and in Al, respectively. In Table I the fraction
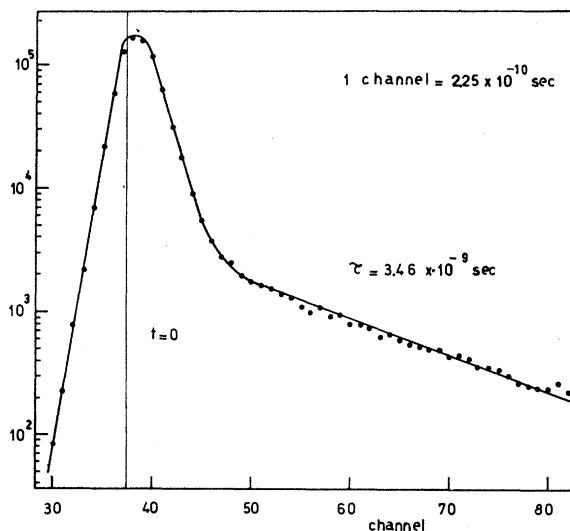

FIG. 1. Time spectrum of positrons annihilating in BeO.

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[2] A. Bisi, A. Fiorentini, and L. Zappa, Phys. Rev. **134**, A328 (1964).
[3] S. M. Neamtan and R. I. Verral, Phys. Rev. **134**, A1254 (1964).
[4] A. Bisi, A. Fasana, E. Gatti, and L. Zappa, Nuovo Cimento **22**, 266 (1961).
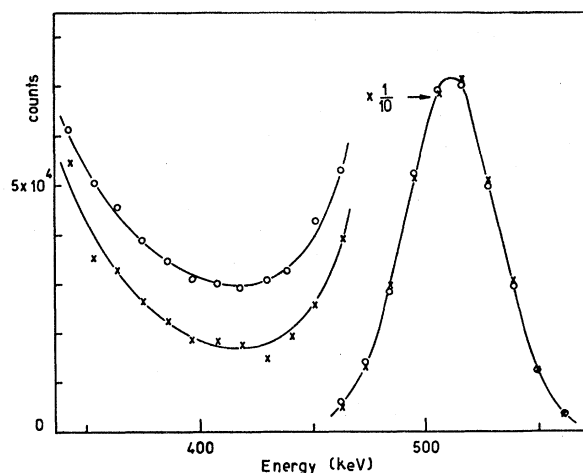[5] V. W. Hughes, S. Marder, and C. S. Wu, Phys. Rev. **98**, 1840 (1955).

FIG. 2. Annihilation radiation spectrum in the region 350–530 keV obtained with the active sample sandwiched in BeO (curve with open circles) and in Al (curve with crosses), respectively.

p_3 of the three-quantum events is also reported; this was deduced as described in what follows.

The fraction p_3 was obtained by means of relative measurements, i.e., by comparing, in the energy range 380–460 keV, the radiation intensity of positrons annihilating in the oxide under investigation with that in Al. In order to minimize any spurious effect due to absorption and scattering of the γ rays, all the targets had the same thickness (200 mg/cm², sufficient to stop all beta particles) and the same size; further, the active sample deposited on a very thin Al foil (0.18 mg/cm²) was put in a composite sandwich of four targets two of which, i.e., those of Al, operated as standard targets. The measurements consisted of several alternate recording runs, with the sandwich arranged with the oxide inside and Al outside, and conversely.

The measured intensity can be related to the p_3 value by means of the following equation:

$$\frac{I_v}{I_{v_0}} = \frac{1 + (p_3/p_2)(\epsilon_3/\epsilon_2)(I_p/I_v^*)}{1 + (p_3/p_2)_0(\epsilon_3/\epsilon_2)(I_p/I_v^*)},\qquad(1)$$

where I_v and I_v^* are the intensities in the "valley" (380–460 keV) with and without the contribution of three quanta, respectively; I_p is the intensity under the "peak"; ϵ_3 and ϵ_2 are the total detection probabilities of an event resulting from the annihilation through three and two quanta, respectively; p_3 and p_2 are the fractions of three- and two-quantum events, respectively.

Equation (1) puts explicitly in evidence that the measured intensity in the "valley" region consists of both a portion of the three-quantum spectrum and a portion of the two-quantum spectrum as seen from the scintillation spectrometer. For convenience this last portion is measured relative to the intensity I_p under the "peak" (511 keV).

In Eq. (1), the subscript 0 refers to the Al target.

The value of I_p/I_v^* was obtained, with an approximation quite sufficient for the present investigation from the spectrum of annihilation radiation of positrons in Al. ϵ_3 and ϵ_2 were calculated on the basis of the gamma-ray spectral shape from three-photon annihilation,[6] and of the detecting efficiency of the scintillation spectrometer.[7] Furthermore, we have assumed

$$(p_3/p_2)_0 = 3(C_T/C_s) = 1/372,$$

where C_T and C_s are the fundamental interaction rates in the triplet and singlet state, respectively.[4] The statistical factor 3 enters because the ^3S states contribute $\frac{3}{4}$ of all the collisions of the positron with free electrons at low energy.

A check on the whole procedure was carried out by measuring the ratio I_v/I_{v_0}, firstly with various metals from Be to Ag, and then with Teflon where an appreciable contribution to p_3 derives from the annihilation of positronium. In this last case p_3 was calculated taking into account the abundance and the lifetime of positronium[6] and by assuming that in one-half of all collisions of free positrons the external electrons have the parallel spin orientation. This drastic assumption on the relative spin population was made by us owing to the fact that in Teflon the positron annihilation takes place within atoms with closed shells, in which the electron spins are paired off. An exhaustive theoretical treatment of the annihilation of free positrons with bound electrons was given by Chang Lee[8] for the simplest case of the He atom. In metals, I_v/I_{v_0} was found to have the same value within the experimental error ($\sim6\%$); there appears, however, a slight increase at higher Z (about 1%), which we attribute to an additional effect due to production of external bremsstrahlung giving rise to an increase of the radiation intensity in the "valley" region. In Teflon, the p_3 value deduced from Eq. (1) [$p_3 = (5.87\pm0.50)\times10^{-3}$] agrees quite well with the calculated one [$p_3 = (5.47\pm0.15)\times10^{-3}$].

We wish to point out that the usefulness of this method is not as general as may appear at first sight. In fact, we have ascertained the appearance of instrumental effects which tend to lower the ratio I_v/I_{v_0} when targets of low density are used and to an extent which increases with decreasing distance between source and scintillator. We have not investigated such effects in detail, since their amount does not appreciably affect our results on alkaline-earth oxides where p_3 attains large values.

In order to correlate the results of lifetime measurements and three-quantum production, we make the simple and plausible assumption that in alkaline-earth oxides the positron becomes bound in a system which

[6] A. Ore and J. L. Powell, Phys. Rev. 75, 1696 (1949).

[7] S. H. Vegors Jr., L. L. Marsden, and R. L. Heath, IDO-16370, 1958 (unpublished).

[8] Chang Lee, Zh. Eksperim. i Teor. Fiz. 33, 365 (1957) [English transl.: Soviet Phys.—JETP 6, 281 (1958)].

is responsible both for the long-life component in the time annihilation spectrum, and for the three-quantum abundance. In particular, for a positronium-like system which, in its ^3S state, decays both with emission of three quanta and with emission of two quanta through a "pick-off" process, the following equation can be written:

$$p_3 = (1 - \tfrac{4}{3}f)C_T/(C_T + C_s) + f(\tau/\tau_3), \quad (2)$$

where the first term gives the contribution to p_3 arising from the free positrons on the same assumption of the relative spin population used above for Teflon, and the second that arising from the positrons bound in ^3S states whose percentage abundance is f; τ and τ_3 are the total (see Table I) and the partial three-quantum lifetimes, respectively. The abundance f is not equal to the abundance I of the long-lifetime component reported in Table I, owing to the fact that of the whole three-quantum spectrum, only that fraction used by us in operating the time analyzer contributed to the annihilation-time spectrum (330–530 keV). Taking into account this fact, the I values of Table I can be corrected to obtain the f values, which are reported in Table II together with the values of τ_3 deduced from Eq. (2).

As can be seen, the τ_3 values are much lower than the value $\tau_3 = 1.39 \times 10^{-7}$ sec characteristic of ortho-positronium in free space, so that the bound system formed by a positron in alkaline-earth oxides shows significant departures from a positronium atom. In fact, as can be seen from Table I, the three-quantum production in BeO exceeds by a factor 15 the values that one would expect for orthopositronium in free space.

At present, it is very hard to give even a tentative

TABLE II. Percentage abundance f and partial three-quantum lifetime τ_3 of the positronium-like system in the ^3S state.

Oxide	$f(\%)$	$\tau_3 \times 10^9$ (sec)
BeO	8.4 ±0.2	4.8±0.5
MgO	2.5 ±0.1	5.4±0.7
CaO	3.3 ±0.3	19 ±4
BaO	0.74±0.08	7 ±2

sketch of the nature of this bound system. For instance, if one compares the set of the collected data on alkaline-earth oxides with those obtained in the previous work on alkali halides,[2] it can be seen that the present situation is quite unfavored. Our present results cannot be connected with previous measurements, nor with theoretical previsions, which are lacking. Furthermore, it must be emphasized that in alkali halides, the collected data were adequate in number, and the abundance of the long-life component quite large, so that empirical regularities between annihilation rates and molecular density could be easily detected. No regular trend can be detected in the present case.

Furthermore, any attempts to gain information on the nature of the long-life component in alkaline-earth oxides through a phenomenological approach similar to that used for a positronium atom in an insulator (i.e., dependence of intensity and lifetime on factors like magnetic and electric fields, temperature, pressure, phase) imply long, troublesome, and scarcely accurate measurements because of the low abundance of the component (of the order of 1%). Perhaps the only material which could be investigated with some success is beryllium oxide.