## Long Lifetime of Positrons in Ionic Crystals\*

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The time-annihilation spectrum of positrons in some ionic crystals was found to be complex. In alkali chlorides, besides the short-lifetime component typical of free-positron annihilation, the spectrum displays a long-lifetime component having a relative intensity of about 25% and a mean life ranging from  $4.3 \times 10^{-10}$ sec (LiCl) to 6.4×10<sup>-10</sup> sec (CsCl). A linear dependence appears to occur between the long-lifetime annihilation rate and the molecular density. In lithium hydride the long-lifetime component exhibits a relative intensity of 12% and a mean life of 1.5×10<sup>-9</sup> sec. Taking into account the difficulties faced by assuming positronium formation in ionic crystals, an attempt is made to interpret our results on the basis of the existence of positron compounds, like that of Wheeler ( $Cl^-e^+$ ) and that of Ore ( $H^-e^+$ ).

HERE has been no evidence, until now, for a long lifetime of positrons in ionic materials. A set of measurements conducted by us on positron annihilation in alkali chlorides and in lithium hydride have shown that in these materials positrons exhibit a complex timeannihilation spectrum. Besides the short-lifetime component ( $\sim 2 \times 10^{-10} \text{ sec}$ ) characteristic of positrons which annihilate when free, the spectrum displays a long-lifetime component (~6×10<sup>-10</sup> sec for alkali chlorides and 1.5×10<sup>-9</sup> sec for LiH) with a relative intensity of about 25% for alkali chlorides and 12% for LiH. If we accept the present view according to which in ionic crystals the formation of a positronium atom, stable with respect to subsequent dissociation, is energetically not permitted,1 we are led to interpret our results as evidence in favor of the existence of nonpositronium bound systems.

The positron emitter used was Na22; one drop of high specific activity solution of Na<sup>22</sup>Cl was evaporated on aluminium foil (0.18 mg/cm<sup>2</sup> thick) sandwiched between the specimens under investigation; their thickness was sufficient to stop all positrons. The alkali chloride specimens were multicrystalline analytical reagent-grade chemicals; the LiH specimen was a single crystal. The process was carried out in a glove dry box and the whole assembly of source-specimen was enclosed in a Lucite container hermetically sealed.

The same experimental method used in previous research<sup>2</sup> was adopted for the detection of the  $\gamma$  rays and for the analysis of the time-distribution spectra. The results obtained are collected in Table I, and a typical time spectrum is shown in Fig. 1. It is apparent that the short-lived positrons exhibit a lifetime  $\tau_1$  which is the same for all chlorides within the limits of experimental

Similarly, the relative intensity of the long-lived positrons appears to be rather insensitive to the change of positive ion within the chlorides; on the other hand, however, the lifetime  $\tau_2$  appears to increase from LiCl to CsCl, exhibiting a regular trend. An attempt to cor-

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relate the annihilation rate  $\lambda_2$  of the long-lived positrons with the molecular density, which is shown in Fig. 2, leads to the following results: (1) a linear dependence of the annihilation rate on the molecular density, and (2) an extrapolated value  $\lambda_0$  to zero density equal to  $0.84 \times 10^9 \text{ sec}^{-1}$ .

Table I. Positron lifetimes in ionic materials;  $\tau_1$ ,  $\tau_2$ , and  $I_2$  indicate the mean lifetimes of short-lived positrons, of long-lived positrons, and their intensity, respectively.

Ionic material	Molecular density ×10 <sup>-22</sup>	$\tau_1 \times 10^{10}$ (sec)	$\tau_2 \times 10^{10}$ (sec)	$\stackrel{I_2}{(\%)}$
LiCl	2.85	1.8±0.6	4.25±0.25	25±7
NaCl	2.23	$1.6 \pm 0.8$	$4.79 \pm 0.29$	$26 \pm 8$
KCl	1.60	$2.2 \pm 0.9$	$6.10 \pm 0.45$	$25\pm7$
RbCl	1.36	$2.2 \pm 0.9$	$5.90 \pm 0.19$	$30 \pm 6$
CsCl	1.42	$1.6 \pm 0.5$	$6.39 \pm 0.18$	$25\pm4$
LiH		$2.1 \pm 0.3$	$15.2 \pm 0.8$	$6.4 \pm 0.4$

Now we point out that a rate of 0.80×10<sup>9</sup> sec<sup>-1</sup> has been calculated by Simons<sup>3</sup> for the annihilation of positrons in the Wheeler compound (Cle-)e+ (a complex where the two particles in the parentheses are more closely bound to one another than the third) in its ground state by assuming that the probability of antiparallel spins is 1:4 as in the case of positronium. If the good agreement between our measured  $\lambda_0$  value and the theoretical one is not accidental, it is conceivable that the long-lived positrons in alkali chlorides annihilate through two competitive processes. One is the annihilation of the positron with its electrons in the system  $(Cle^{-})e^{+}$ ; the second should be a "pick-off" annihilation of the bound positron with an electron belonging to one of the surrounding atoms in the solid, or a compounddestroying process such as the transformation from  $(Cle^{-})e^{+}$  to  $Cl(e^{-}e^{+})$  whose lifetime is much shorter. This transformation could be induced in the solid due to the fact that the latter compound has a smaller energy than the former, as shown by Simons. In any of these cases the linear trend of Fig. 2 can be easily realized.

<sup>&</sup>lt;sup>1</sup> R. A. Ferrell, Rev. Mod. Phys. 28, 308 (1956). <sup>2</sup> A. Bisi, A. Fasana, E. Gatti, and L. Zappa, Nuovo Cimento 22, 266 (1961).

 <sup>&</sup>lt;sup>3</sup> L. Simons, Phys. Rev. 90, 165 (1953).
<sup>4</sup> J. A. Wheeler, Ann. N. Y. Acad. Sci. 48, 219 (1946).

As far as the long lifetime in LiH is concerned, it would seem straightforward to ascribe it to a bound state of a positron with a negative hydrogen ion, whose possibility of formation was first suggested by Ore.<sup>5</sup> This interpre-

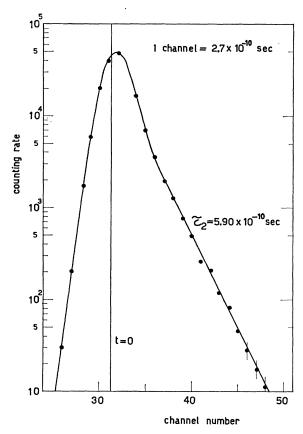


Fig. 1. Time spectrum of positron annihilation in RbCl.

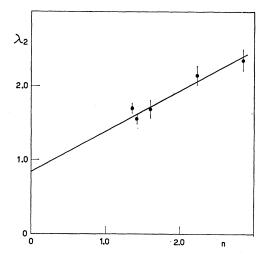


Fig. 2. Annihilation rate  $\lambda_2$  (units  $10^9~{\rm sec^{-1}}$ ) of long-lived positrons in alkali chlorides as function of molecular density n (units:  $10^{22}~{\rm cm^{-3}}$ ).

tation faces serious difficulties because the experimental value of the annihilation rate disagrees with that evaluated by Neamtan  $et~al.^6~(0.20\times10^{10}~{\rm sec}^{-1})$  for the annihilation of positrons from the  $({\rm H}^-e^+)$  ground state; the possibility remains however, as emphasized by Neamtan et~al., that such a bound system might be formed in solids but it would be described by wave functions which take into account the effect of the surrounding ions.

Finally, we wish to note that extensive data exist on the angular correlation of annihilation quanta in ionic materials, 7,8 for a correct interpretation of which the presence of a significant long-lifetime component cannot be ignored.

 $<sup>^{6}</sup>$  A. Ore, University of Bergen Yearbook No. 5 (Bergen, Norway, 1952).

<sup>&</sup>lt;sup>6</sup> S. M. Neamtan, G. Darewych, and G. Oczkowski, Phys. Rev. **26**, 193 (1962).

<sup>&</sup>lt;sup>7</sup> A. T. Stewart and R. H. March, Phys. Rev. 122, 75 (1961). <sup>8</sup> A. T. Stewart and N. K. Pope, Phys. Rev. 120, 2033 (1960). This article gives reference to earlier works.