

TIME SPECTRA OF POSITRON ANNIHILATION
IN SOME LIQUIDS AND SOLIDS

by

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ABSTRACT

The change in the long lifetime (τ_2) for positron annihilation has been measured as a function of the temperature for Diphenylamine, N-Phenylbenzylamine and P-Bromotoluene. In each case the temperature range covers a first order phase transition of the substance under investigation.

The annihilation time spectra of positrons were studied in LiH and NaH. In both cases a complex spectrum was obtained. NaH showed a long lifetime of 2.6 nanoseconds and a short one of 0.43 nanoseconds while LiH showed a long one of 2.1 nanoseconds and a short one of 0.41 nanoseconds. The long components had an intensity of 10% in LiH and 15% in NaH.

CHAPTER I

INTRODUCTION

Several authors (Ruark¹, Ore²) have delved into the theoretical considerations of the interactions of positrons and electrons. They have predicted that a system consisting of a positron and an electron can exist metastably in vacuum and in a suitable nonvacuous environment. The bound system was given the name positronium by Ruark. The quantum mechanical argument closely parallels that for the hydrogen atom and to first order theory the "Bohr" radius of the ground state is 1.06 Angstroms and the energy of formation is 6.8 ev. in vacuum.

The ground state configuration consists of the 1^1S (singlet positronium) and the 1^3S (triplet positronium) substates. From statistical considerations it follows that the triplet substate should be three times as probable as the singlet. Spontaneous transitions between the ground state substates are absolutely forbidden in the absence of an external magnetic field, however both states are unstable against self-annihilation. The conservation laws infer that in the absence of an external electromagnetic field the singlet state decays with the emission of two quanta of electromagnetic radiation correlated at 180 degrees in the center of mass coordinates, whereas the triplet state annihilates with the subsequent appearance of three coplanar quanta. Single quantum annihilation may occur in the presence of a strong magnetic field.

The mean lifetimes against annihilation have been calculated for both states in a vacuum environment.

$$\tau_1 \text{ (singlet)} = 1.25 \times 10^{-10} \text{ sec.}$$

$$\tau_2 \text{ (triplet)} = 1.39 \times 10^{-7} \text{ sec.}$$

(Ore¹ and Powell³; Ferrell⁴).

It was pointed out by A. Ore that the formation of positronium in a nonvacuous environment is energetically favored only in a rather restricted range of positron velocities. When a positron enters a nonvacuous medium it generally does so with an energy which is greater than some excitation potentials of the atoms in the attenuating material. At such energies the cross sections for inelastic collisions are considerably larger than the cross section for the formation of positronium. As a result the positron will most probably engage in a number of inelastic collisions. The probability for the binding process becomes important in a fairly narrow range of velocities roughly centered about the "orbital" velocity of the outer atomic electrons of the environment. At lower velocities elastic collisions predominate.

Ore¹ has given the lower bound of the energy for which binding is possible in the case of an atomic gas. If E_i is the ionization energy for gas atoms, and if E_p is the binding energy of positronium (approximately 6.8 ev.) then the lower bound for the positron energy E' is given by:

$$E' = E_i - E_p$$

R. A. Ferrell⁴ has extended the theoretical treatment to a more general case.

Binding with free electrons is not possible in the absence of an external field.

Now experimentally one measures the mean lifetimes of positrons in matter. Annihilation from the bound state rather than direct annihilations of positrons was inferred by Deutsch and Shearer who noticed that the rate of annihilation was not proportional to the gas pressure for the short lifetime component. Backing for this argument was provided by the triple coincidence measurements by deBenedetti and Siegel⁵ and by Deutsch and Dulit⁶ who observed the quenching of the triplet state by an applied magnetic field.

However, in order that one may speak meaningfully about the absolute lifetimes of positronium in a given medium, one requires the assurance that the thermalisation time is small in comparison to the lifetime of the bound state. In other words, the inelastic collisions must be sufficiently effective in attenuating the initially fast positron to such velocities where the binding cross section is high. Calculations of thermalisation times have been carried out by deBenedetti et al.⁵, Lee-Whiting⁷ and Wallace⁸. They indicate a time of the order of 10^{-12} seconds for metals. Reliable estimates of thermalisation times of positrons in insulators and liquids do not

exist at present. It will be assumed here that it is reasonable to discuss absolute lifetimes where fast positrons from Na^{22} are employed.

Experimental investigations have displayed a rather varied behavior of the time spectrum of the annihilation radiation in a wide range of materials.

In metals only one lifetime is observed and this is rather insensitive to the type of metal under study. The magnitude of the lifetime stays in the range from $1-3 \times 10^{-10}$ seconds even though the electron densities may vary considerably from one metal to another.

Recently Bell has found a long component of very low intensity in some metals. In the case of ionic and valence crystals the results were similar. Here again, very recent measurements by Bisi et al.⁹ have displayed a long component of low intensity.

The situation is not at all analogous in many organic amorphous and molecular materials which have been studied. These substances reveal a complex time spectrum. One observes a short lifetime of the order of 10^{-10} seconds, and a longer one of about $1-3 \times 10^{-9}$ seconds. The fast rate is in reasonable agreement with the theoretically predicted value and is not sensitive to the environment. However in the case of the longer lifetime there is considerable deviation from the theoretical prognosis for triplet positronium in vacuum. Furthermore the magnitude of the long component is strongly dependent on

temperature and pressure as well as on externally applied magnetic fields. At points of first order phase transitions a sharp change is found to occur in the long lifetime.

The fractional amount of positrons annihilating from the triplet state, the so called I_2 , is also found to be sensitive to the environment. It decreases when a magnetic field is applied and when paramagnetic atoms or ions are added in small concentrations to the substance under study. An example of this latter effect is shown by Deutsch¹⁰, who added small amounts of NO and O₂ to his samples.

Similarly, in angular correlation measurements, where momentum distributions of the two photon annihilation process are investigated, two distinct momentum groups are often found. One is a narrow component which corresponds to low values of angular momentum; the other is a wider distribution and is due to pairs annihilating with higher angular momenta. Page et al.¹¹ have shown that the appearance of the narrow component is probably associated with the formation of positronium. Ferrell¹² has pointed out that pick-off annihilation is unlikely to fall into the low momentum region and hence the narrow distribution is likely due to annihilations from the singlet state.

On this basis the angular correlation bears out the quenching effect which was observed in the studies of the time spectra.

It should be noted here that the self-annihilation rate of positronium in the triplet state is proportional to the amount of

overlap of the wave functions of the positron and electron which form the bound pair. This overlap is not sufficiently sensitive to the temperature and pressure to account for the strong dependence on these parameters. Therefore, if we are to follow up the assumption that positronium is formed, we are forced to consider auxiliary mechanisms in order to account for the strong influence of electron density variations and magnetic fields.

One such mechanism is the "pick-off" process whereby the bound positron annihilates with an atomic electron. The probability of such a process will be directly proportional to the amount of overlap of the wave functions of the positron and the atomic electron and hence will be a function of the electron density in the immediate vicinity of the positronium atom. This will be sensitive to the pressure and temperature of the sample and therefore the pick-off process could be effective in shortening the long lifetime. The short lifetime should then also be affected but this effect will be lost in experimental uncertainty since the self-annihilation from the singlet state is itself highly probable.

Another possibility was examined by Ferrell¹². It concerns the triplet to singlet state conversion with the aid of a magnetic field, either molecular or microscopic in nature.

It further appears feasible that a positron compound is formed in some cases. Several groups have attacked this problem from

the theoretical standpoint. Wheeler et al.¹³ have examined the halogen chlorides. Neamtan et al.¹⁴ have considered the formation of H^-e^+ bonds. Bisi et al.⁹ have very recently shown that LiCl, CsCl, and LiH exhibit a complex decay scheme. The case of LiH shows a long lifetime of 1.5×10^{-9} seconds with an intensity of 6-7% and a short lifetime of 2×10^{-10} seconds.

In the case of these materials the short lifetime is regarded as being due to annihilations of free positrons while the long component is thought to emanate from the annihilation of positrons from the bound system $Li(H^-e^+)$. Again a density dependence is observed and to first order this is a linear relationship according to Bisi et al. Further evidence for positron compound formation was indicated by Lundholm et al.¹⁵

Another form of binding was inferred by Ferrell¹² from the following evidence:

On addition of diphenyl-picryl hydrazal, a negative free radical, the τ_2 value was reduced whereas the I_2 stayed constant within experimental error. In angular correlation measurements DeZafra¹⁶ noticed a broadening of the distribution without the formation of a narrow component. These observations could be reconciled by assuming that positronium atoms were bound to the D.P.H. molecule or radical.

CHAPTER II

APPARATUS

In these measurements Na^{22} sources were employed. When Na^{22} decays with the emission of a positron, Ne^{22} is formed in an excited state and undergoes a transition resulting in the emission of a 1.28 MeV gamma ray. The time lapse between the appearance of the 1.28 MeV gamma and the positron is too short to be measured with the apparatus of this experiment, hence the detection of the gamma ray is taken as the reference point in the lifetime measurements.

The annihilation of the positron with an electron is accompanied by two 0.511 MeV gamma rays correlated at 180 degrees. The lifetime of the positron is determined by measuring the time lapse between the advent of the 1.28 MeV gamma and that of the 0.511 MeV gamma.

The mechanics of the measurements are as follows:

Na^{22} sources were made by evaporating aqueous sodium chloride solution of high specific activity (2 mc/mg) on thin mica (5 mg/cm^2) sheets. These sources were then immersed in glass test tubes containing a few cubic centimeters of the particular substance under investigation. This source assemblage was then placed into the temperature regulation equipment which consisted in some cases of a furnace and in others of a cryostat or simply a thermos bottle. The detectors were then brought as close as possible to the location of the source as conditions permitted.

The schematic diagram of the electronic apparatus is shown in Figure 1.

Cylindrical plastic phosphors (NEE102), optically coupled to IP21 RCA photomultipliers served as detectors. On detection of gamma rays these photomultipliers deliver negative pulses of the order of ten volts. These pulses are then employed to trigger limiter circuits which are designed to put out fast rising, flat topped pulses. The rise time of the limiter pulse is about one nanosecond and the length of the pulse approximately two microseconds. These flat pulses are chopped to a length of 32 nanoseconds by shorting stubs and are subsequently fed to a time-to-pulseheight converter. The latter is designed to produce an output pulse whose height is directly proportional to the time interval during which two input pulses overlap. This is accomplished by directing the pulses from the two limiters onto two separate control grids of a 6BN6 pentode which is biased so as to give an output pulse only when both grids are activated simultaneously. The resulting pulse then travels to the control grid of a 5842 triode via an RC network. The RC network has an exponential voltage versus time response, however, only the closely linear initial section of this is employed. Hence the output of the triode becomes directly proportional to the time during which the 6BN6 conducts, that is, to the time during which the limiter pulses overlap.

The triode output is then amplified and fed to a Victoreen

BLOCK DIAGRAM OF APPARATUS

