Lifetime of Positrons in Superconducting Lead and Tin*

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The mean life of positrons was measured in superconducting lead and tin. Consistent results were obtained with the lead sample giving an average value for the mean life of $(0.57\pm0.07)\times10^{-9}$ sec. A null result was obtained with tin; the measured value of the lifetime was $(0.15\pm0.09)\times10^{-9}$ sec, as compared with the room temperature value of 0.15×10^{-9} sec. The data indicate that the lifetime change in lead is a superconductive phenomenon and not due simply to the low temperature of the sample. Conjectures are made to explain the different results obtained with lead and tin.

INTRODUCTION

NUMEROUS investigations have recently been made to measure the mean life of positrons in condensed materials. The work of Bell and Graham[1] provides the most extensive list of sample materials and allows certain general conclusions to be drawn. Two of these apply particularly to the work being reported upon here. First, the mean life in metals appears, within experimental error, to be constant at about 1.5×10^{-10} second. Second, the mean life measured in insulators varies considerably for different materials, but is almost always longer than that observed in metals by a factor of two or three. In some insulators, about 30 percent of all positrons appear to decay with a longer mean life than the others. The mean life of the longer-lived component decreases as the temperature decreases.

Now superconductors, in spite of their anomalous conductivity, in some ways more nearly resemble insulators than conductors. The infinite conductivity results from the presence of a certain number of electrons, termed superelectrons, which do not interact with their environment. Among the other superconductive effects, one would expect an increase in the positron life time due to the presence of the superelectrons which would not interact strongly with positrons or positronium, and hence not contribute to the annihilation. Dresden[2] indicates two processes which may cause the mean life of positrons in superconductors to differ from the mean life in normal materials. First, the wave function of an electron in a superconductor is certainly much different from that in a normal conductor, and the wave function of a positron may also be different. These differences should lead to an increase in the mean life of all positrons in superconductors. A crude estimate of the amount of this effect by Dresden (unpublished) indicated that a mean life of as much as 3×10^{-9} second for positrons in superconducting lead at 4.2°K might be expected. The second process indicated by Dresden gives rise to a complex lifetime, due to the formation of positronium. Positronium, formed in the triplet state, should not be converted to the singlet state by the superelectrons, and hence might remain much longer than in normal metals. However, it should be noted that all mechanisms, other than the interaction with conduction electrons, which convert triplet to singlet positronium in solids, will probably also be present in superconductors. The magnitude of both effects, on the basis of a superconductivity theory alone, is, of course, a function of how far below the transition temperature the sample is maintained, since thereby is determined the number of superelectrons.[3]

Investigations were then initiated to see whether or not any lifetime change could be observed. Lead was chosen as the first sample material, not only because calculations had been made in terms of it, but also its transition temperature (7.2°K) was sufficiently high to give a relatively large number of superelectrons at the boiling point of liquid helium.

EXPERIMENTAL DETAIL

The customary technique of delayed coincidences was used in carrying out the measurements. The coincidence circuit used initially was a modification of that proposed by de Benedetti and Richings.[4] The modifications consisted, principally, of the use of a negative high-voltage supply and the elimination of the limiter circuit. Stilbene crystals mounted on 5819 phototubes were used as detectors and RG 88/*u*, 185-ohm, cable was employed to secure the necessary delays. The resolving times obtained with this coincidence circuit were, approximately, 1.5×10^{-9} sec.

In later experiments, the coincidence circuit described by Bell *et al.*[5] was adopted, and the 5819 photomultiplier used instead of the 1P21. The circuit requires output of approximately three volts from the phototubes, and necessitates applying, therefore, voltages of about 1500 volts to the 5819. At these voltages

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[1] R. E. Bell and R. L. Graham, Phys. Rev. **90**, 644 (1953).

[2] M. Dresden, Phys. Rev. **93**, 1413 (1954).

[3] F. London, *Superfluids* (John Wiley and Sons, Inc., New York, 1950).

[4] S. de Benedetti and H. J. Richings, Rev. Sci. Instr. **23**, 37 (1952).

[5] Bell, Graham, and Petch, Can. J. Phys. **30**, 35 (1952).

the tubes exhibited a form of breakdown characterized by excessive noise and the disappearance of the output pulses. The reason for this was found to be connected with the amplification characteristics of the particular phototube, and the fault could be eliminated by applying stabilizing voltages to the last two dynodes. More complete details are to be published elsewhere.

In each case, both counters were allowed to accept both the nuclear and the annihilation radiations; therefore the delay curves are symmetric about zero delay.

The sample was a solid cylinder along the axis of which a hole had been drilled. $Na^{22}Cl$ in a HCl solution, as obtained from the Oak Ridge National Laboratory, was evaporated in the cylinder. Virtually all of the measurements in which the sample was to be superconducting were made with the cylinder placed in the bottom of a glass Dewar into which had been put liquid helium. The data for liquid nitrogen temperature were obtained from the same arrangement, except that now the sample was either submerged in liquid nitrogen or the chamber between the helium and nitrogen containers was filled with air, thus putting the sample at the nitrogen temperature. Because it was necessary to surround the helium Dewar with liquid nitrogen, the counters were about two inches from the sample, so that the coincidence counting rate was low. For this reason, accurate data for long delay times could not be obtained.

RESULTS

A preliminary experiment was carried out in order to ascertain whether any difference in the mean life between the normal and superconducting states of lead could be observed. The sample was submerged in liquid helium and the delays following the counters were adjusted to correspond to the peak of the delayed coincidence curve. Then the coincidence rate as a function of time was taken. If the mean life of the positron in the superconducting state were longer, the coincidence rate would be lower than the rate taken at a temperature above the transition point. With the lead at the temperature of liquid helium, then, the rate would be low, and would increase very rapidly when the helium in the Dewar had entirely evaporated.

The data are shown in Fig. 1 and clearly follow the indicated behavior. For about thirty minutes, while helium surrounded the lead, the rate was low; at the end of that time, when the helium was gone, the coincidence rate suddenly increased.

Obviously these results may be due to either of the processes described by Dresden. The change could be due to a small increase in the mean life of all positrons, or it could be due to a large increase in the mean life of about 30 percent of the positrons. Assuming that the change is due to a change in the mean life of all positrons, the amount of the lifetime change may be calculated. Such a calculation indicates a mean life of $(0.50\pm0.09)\times10^{-9}$ second. (The uncertainties given here are essentially the standard deviations due to the number of counts involved.) Although the statistics of the experiment are poor, the experiment indicates fairly conclusively that there is a lifetime change. Also, the method serves to give a quick and simple means of detecting lifetime changes of the type being discussed here. Surprisingly, in view of the statistics, the result given above checks quite well with the much more reliable results to be given below.

Satisfied that a change in the mean life could be measured and using the same experimental arrangements, we took several complete decay curves corresponding to the temperatures of liquid helium, liquid nitrogen, and the laboratory. The liquid helium and liquid nitrogen curves are shown in Fig. 2; the curves have been normalized to the same included area. The shape of the delay curves was the same for the liquid nitrogen and room temperature data. Assuming various

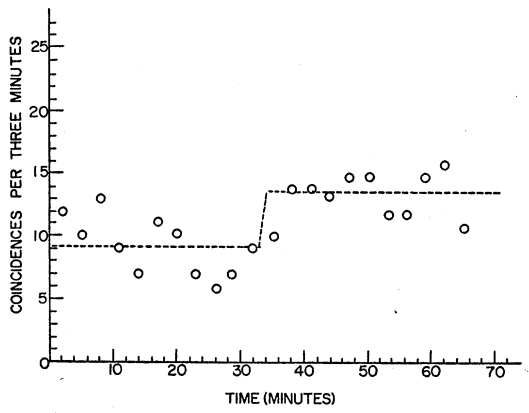

FIG. 1. Plot of coincidences-*vs*-time taken at the peak of a symmetric delayed-coincidence curve for a lead sample.

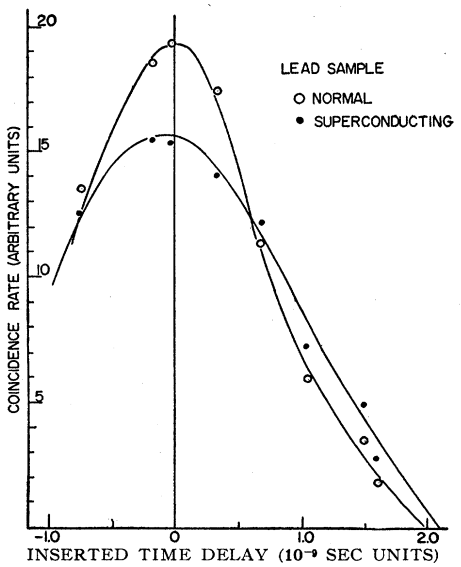

FIG. 2. Delayed-coincidence curves taken for a lead sample at liquid helium and liquid nitrogen temperatures.

single mean lifetimes, the corresponding exponential curves were "smeared-out" with the known shape of the prompt curve and fitted to the liquid helium data. The best fit was given by a mean life of 0.58 ± 0.09 second.

The integrated coincidence rates over the range of delays from -1.5×10^{-9} to $+1.5\times10^{-9}$ second were the same for the liquid nitrogen, liquid helium, and room temperature data. Thus, if a complex life time occurs, a lower limit may be set on the fraction of the positron having the longer mean life if the mean life is known; or an upper limit may be set on the mean life if the fraction of the positrons having that mean life is known. If it is assumed that 30 percent of the positrons have the longer mean life, then the mean life cannot be over about 10^{-9} second. Likewise, if a mean life of 3×10^{-9} second is assumed,[6] then less than 10 percent of the positrons are annihilated with this mean life. This is in agreement with the results of Madansky (private communication) who sets an upper limit of 1 percent on a long-lived component.

The use of lead as sample material has the obvious disadvantage that its transition temperature, 7.2°K, is so high there is no practical way of taking it just out of the superconducting state. Tin, on the other hand, with a transition temperature of 3.26°K presents no such difficulty. Consequently, delayed coincidence measurements were made with tin as the material in which the positrons decayed. Three sets of curves were taken: one with tin at room temperature; one at 4.2°K; and one at 2.4°K, obtained by reducing the pressure in the helium chamber.

Within the accuracy of the experiment, there was no change in the mean life of the positrons with temperature, and no superconducting effect occurred. Calculations of the number of superelectrons indicated, however, that the positron mean life in superconducting tin should be about nine-tenths of that found for superconducting lead.

Just before the measurements on tin were made, the coincidence circuit had been changed to one similar to that described by Bell.[5] In order to be sure that neither the earlier results in lead nor the later ones in tin were due to some sort of failure of the coincidence circuit, measurements of lead were repeated with same circuit with which the lifetime in tin had been measured. The data are similar to Fig. 2. The mean life computed as before is

$$\tau=(0.62\pm0.07)\times10^{-9}\text{ sec.}$$

As in the previous measurement with lead, a single mean life has been assumed. The limit on the presence of a longer mean life component is essentially the same as mentioned above; not over ten percent of the positrons can have a mean life as long as 3×10^{-9} second.

[6] W. E. Millett, Phys. Rev. **94**, 809 (1954).

DISCUSSION

The experiments reported here establish with little doubt that there is a change in the mean life of positrons in lead when the lead becomes superconducting, and that there is at most only a very small change in the case of tin. The time distribution of the annihilation of positrons in lead may be explained by a simple exponential decay with a mean life of $(0.57\pm0.07)\times10^{-9}$ second. However, the results are also consistent with a complex lifetime, provided that both components of the decay have a mean life of less than 10^{-9} second, or if only a small fraction of the positrons contribute to the longer mean life.

The behavior of positrons in solids is not at all well understood, but certain speculations can be made which may allow an explanation for the apparent disagreement between the results given above for superconducting lead and tin. The constancy and shortness of the lifetimes in metals are plausible on the basis of the abundance of free electrons in metals, these electrons being available to annihilate with the positrons either directly or through the formation of singlet positronium and to quench the longer-lived triplet state of positronium. In insulators a somewhat longer time is required before the formation of singlet positronium or the direct annihilation of positrons. Furthermore, triplet positronium is not so rapidly converted to the singlet state so that a second mean life, that of the triplet positronium, is observed. However, there are several mechanisms which can shorten the mean life of positrons in insulators. Two in particular might be mentioned.

The quenching of triplet positronium by external magnetic fields is well-known.[7,8] Crystalline fields could, however, also give rise to a quenching action, and one which would be dependent on the lattice structure. For example, there can be no fields or field gradients at lattice points or at the center of the unit cell in a cubic crystal (such as lead), whereas they can be present in more complicated crystals (such as tin). This may give a partial explanation for the results observed in lead and tin. Thus any long mean life component of positrons in superconducting tin may be masked by the presence of the crystalline fields.

A second effect which may tend to shorten both the long mean life and short mean life of the positron is allied to the concept of the covalent bond. Such bonding could arise from the formation of compounds between positronium and other atoms. Several compounds involving positronium have been shown to be stable,[8] while several others would appear to be stable from the analogy between positronium and hydrogen. In the latter class could be included the metal hydrides. This covalent binding could be very important in

[7] M. Deutsch and E. Dulit, Phys. Rev. **84**, 601 (1951).
[8] M. Deutsch, *Progress in Nuclear Physics* (Academic Press, Inc., New York, 1953), Vol. 3.

explaining the shortness of the lifetime observed in insulators and provide another means by which triplet positronium may be rapidly converted to singlet positronium. The length of the covalent bond when compared with the lattice parameters could also provide criteria determining whether or not such compounds could be formed.

Clearly a great deal of work must be done to be sure that the effects reported here are reproduced in other superconductors. The dependence of the observed lifetime change on the crystal structure of the superconductor could, for example, be checked. There are many other such problems immediately presented. Experiments are being continued here along these and other lines relating to positron life times in solids.

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Critical Field Phenomena in an Isotropic Paramagnetic Crystal

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Measurements have been made of the variation of magnetic moment with applied field, at constant entropy, for a spherically-ground single crystal of chromic methylammonium alum. Steady fields up to 500 oersteds were applied along a cubic axis of the crystal and the differential susceptibility measured ballistically by reversing a field of 1.7 oersteds in the primary of a mutual inductance surrounding the specimen. The total magnetic moment was obtained by integration. At entropies below that of the "Curie point" the differential susceptibility showed a pronounced maximum in a field of about one hundred oersteds. This behavior is compared with that observed in earlier measurements on the same specimen in transverse fields. The magnetic moment at constant external magnetic field H, plotted as a function of entropy, shows a maximum for small values of H. The locus of these maxima is evidently a transition curve separating regions of paramagnetic and antiferromagnetic behavior.

1. INTRODUCTION

SINCE the earliest investigations of the behavior of paramagnetic salts at very low temperatures, reached by means of the technique of adiabatic demagnetization, considerable interest has been centered on the phenomena associated with the appearance of a susceptibility maximum.[1] These include hysteresis and remanence observed in dc measurements, a complex ac susceptibility (ac losses), and a negative magnetocaloric effect—evidence of cooperative effects which have been generally ascribed to the onset of antiferromagnetic behavior.[2]

More recently Garrett[3] has reported measurements made on an anisotropic paramagnetic crystal, cobalt ammonium sulfate, in which an interesting effect was observed below the Curie point for fields applied along the K_1 (strong magnetic) axis—namely, that the differential susceptibility at constant entropy S *increased* with increasing magnitude of the polarizing field, reaching a fairly sharp maximum in fields of a few hundred oersteds. By measuring absolute temperatures in zero field, and by calculating increments of temperature as a function of field along an isentropic, using the thermodynamic formula

$$(\partial T/\partial H)_S = -(\partial M/\partial S)_H \qquad (1)$$

(where M is the magnetic moment of the specimen), Garrett was able to plot on a $T-H$ diagram a lattice of isentropic and isomomental curves. The locus of the minima of both sets of curves described a phase boundary which was identified as delineating the transition from antiferromagnetic to paramagnetic behavior, since the features of the diagram showed striking similarity to the predictions of a theory due to Sauer and Temperley[4] postulating antiparallel ordering at very low temperatures through dipole-dipole interaction.

In a recent investigation of the low-temperature magnetic properties of chromic methylammonium alum, a salt which is magnetically isotropic in the region of 1°K, Hudson and McLane[5] obtained indications of similar behavior of the susceptibility below the Curie point. The measurements were made with a transverse polarizing field, the measured susceptibility being assumed to be equal to M/H, as is true for an isotropic material. The indications were, however, that the salt was definitely not isotropic, at least in fields greater

[1] Kurti, Laine, Rollin, and Simon, Compt. rend. **202**, 1576 (1936).
[2] N. Kurti, J. phys. et radium **12**, 281 (1951).
[3] C. G. B. Garrett, Proc. Roy. Soc. (London) **A206**, 242 (1951).
[4] J. A. Sauer and H. N. V. Temperley, Proc. Roy. Soc. (London) **A176**, 203 (1940).
[5] R. P. Hudson and C. K. McLane, Phys. Rev. **95**, 932 (1954).