Annihilation of Positrons in Superconducting Lead*

S. M. SHAFROTH AND J. A. MARCUS
Physics Department, Northwestern University, Evanston, Illinois
(Received November 30, 1955)

A search was made for changes in the two-quantum annihilation rate when positrons are annihilated in superconducting lead compared to normal lead. The change of state was brought about either by turning off or on a magnetic field or by varying the temperature of the sample. The ratio of the two-photon coincidence rate in normal lead to that in superconducting lead was found to be 1.0011 ± 0.0016 in the magnetic field experiment and 1.0003 ± 0.0011 in the temperature-varying experiment.

1. INTRODUCTION

DRESDEN[1] suggested that the character of positron annihilation radiation might be altered when the metal in which the annihilations were occurring changed from its normal state to its superconducting state. In the latter case, he indicated that the lifetimes of all positrons might be longer and more three-quantum radiation migh be present. Millett[2] investigated the lifetime in lead and found that some positrons have a mean life of $(3.5\pm0.5)\times10^{-9}$ sec when the lead becomes superconducting. Stump and Talley[3] investigated the lifetime effect in lead and tin. They found a slight increase in lifetime for superconducting lead, but no change for tin. Graham, Paul, and Henshaw[4] also investigated the lifetime effect in lead, vanadium and liquid helium. They found no change in lifetime for lead or vanadium in excess of their experimental error of 2×10^{-11} sec for these substances above and below the superconducting transition temperatures. In liquid helium they found a "strong" component with mean life $\tau=(2.7\pm0.3)\times10^{-9}$ sec, which may explain why other observers have reported a long lifetime in superconducting lead. In the present work, a preliminary report[5] of which has already appeared, a search was made for changes in the ratio of two- to three-quantum annihilation rates when lead was changed from normal to superconducting.

2. EXPERIMENTAL PROCEDURE AND RESULTS

The technique for measuring this ratio which was similar to Pond's[6] consisted of measuring the two-quantum coincidence rate resulting from positron annihilations in normal lead compared to the rate in superconducting lead. If a decrease in the two-quantum rate is found, it should be accompanied by a corresponding increase in the three-quantum rate, and vice versa. Electronic drifts were compensated for by frequent changing from one state to the other. Two scintillation counters in a colinear geometry were connected in coincidence. A Na^{22} source was enclosed between two thin lead disks, and placed midway between the two counters, as shown in Fig. 1. The source was made by running 1 millicurie of $Na^{22}Cl$, obtained from Oak Ridge, through an ion exchange column[7] to get as high a specific activity as possible and then evaporating it onto a thin (4 mg/cm^2) aluminum disk. Another disk was glued to the first by means of collodion so that the active salt was completely contained. The rate of two-quantum annihilation in the covered source was estimated by measuring its rate in the bare source[8] and adding to it the two-quantum annihilation rate due to positrons scattered back into the source by the lead. The saturation backscattering coefficient for positrons on lead with 2π geometry is 0.51.[9] We assumed that all backscattered positrons annihilate in the source. This leads to a value of about 0.7 for the fraction of the positrons annihilating in the source when surrounded by the lead disks. The lead disks were made to change from the normal to the superconducting state by two different procedures.

Procedure *A*

In the first procedure, the temperature of the disks was held constant at 4.2°K and an external magnetic field was turned off or on to produce or destroy the superconducting state. The source and 0.008-in.-thick lead disks were supported from the bottom of a metal Dewar which contained liquid helium (Fig. 1). A thin-wall copper radiation shield in the Dewar vacuum space was maintained at liquid nitrogen temperature. This served to reduce the rate of helium evaporation without the increase in scattering of gamma rays which would have resulted from the use of a shielding nitrogen Dewar. Even though considerable precautions were

* This work was supported in part by the Northwestern University Graduate School Fund.
[1] M. Dresden, Phys. Rev. **93**, 1413 (1954).
[2] W. E. Millett, Phys. Rev. **94**, 809(A) (1954).
[3] R. Stump and H. E. Talley, Phys. Rev. **96**, 904 (1954).
[4] Graham, Paul, and Henshaw, Bull. Am. Phys. Soc. Ser. II, **1**, 68 (1956).
[5] S. M. Shafroth and J. A. Marcus, Phys. Rev. **99**, 664(A) (1955).
[6] T. A. Pond, Phys. Rev. **93**, 478 (1954).
[7] We are greatly indebted to Dr. Henry Newman of the Chemistry Department for having performed this operation.
[8] In order to compare the bare-source rate with the covered-source rate, a correction (amounting to about 20% in the coincidence rate) for absorption of the gammas by the various intervening materials which are present in the covered-source experiments was applied.
[9] H. H. Seliger, Phys. Rev. **88**, 408 (1952).

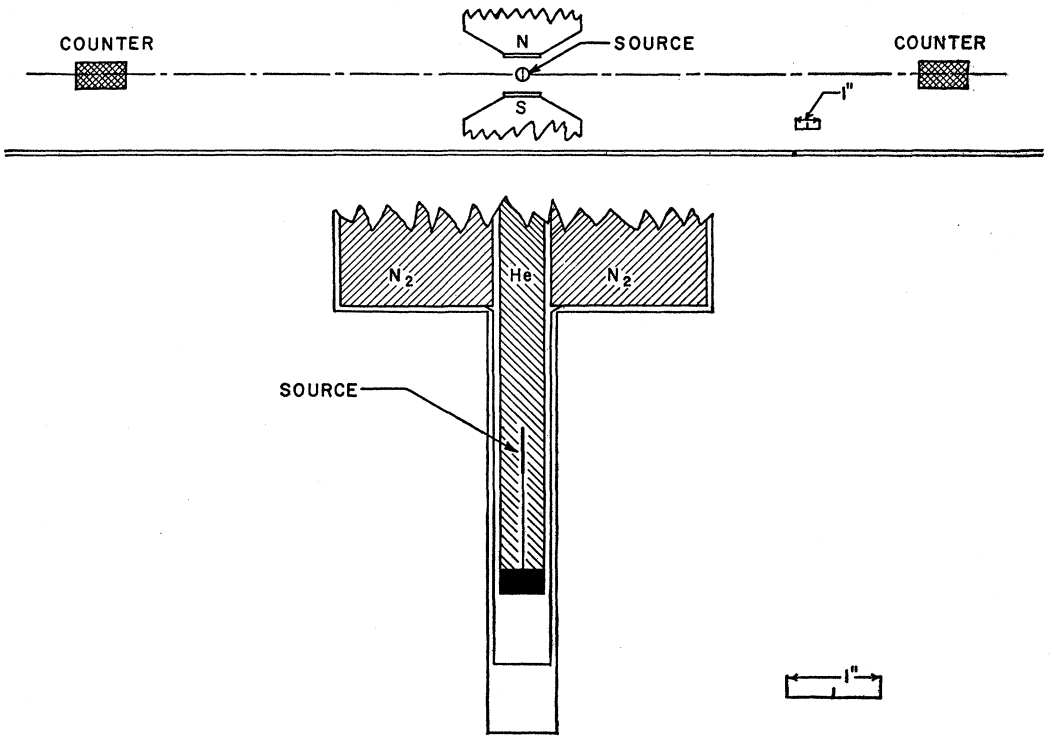

FIG. 1. Top: experimental arrangement used for measuring the two-quantum radiation and for changing the magnetic field in the region of the lead sample. Bottom: detail of the helium Dewar.

taken to shield the counters from the fringing field of the electromagnet, it produced small changes in the counting rates of the order of 0.1%. The correction for this effect was measured at room temperature. The scintillation counters consisted of Larco Plastiflour B crystals 2 in.$\times 1\frac{1}{8}$ in. in diameter, RCA 5819 phototubes, and Hewlett-Packard wide-band amplifiers. The coincidence circuit was a modification of one described by Fischer and Marshall.[10] The resolving time of the apparatus was about 2×10^{-8} second. This reduced the accidental rate to well below 10% of the total rate at the shortest counter-to-source distance. Table I shows the results of runs at two counter-to-source distances using this method. Using the same Dewar and electromagnet, the superconductivity of the lead was checked by suspending the disks from a torsion balance and observing the magnetic moment. Below the critical field (which for lead at 4.2°K is 550 gauss), the usual superconducting behavior was observed, i.e., the disks behaved like perfect diamagnets; and upon exceeding the critical field, the disks lost all magnetic properties as they should have if they were no longer superconductors. However, on turning off the electromagnet, fairly large frozen-in magnetic moments were observed, and associated with these moments were fields large enough to exceed the critical field in certain regions of the sample. Thus, even with the field off, not all of the sample was superconducting. It was estimated that probably more than half the volume returned to the superconducting state. However, to ascertain exactly what fraction was superconducting would have been a problem of considerable difficulty and it was felt that a new approach would be more fruitful.

[10] J. Fischer and J. Marshall, Rev. Sci. Instr. **23**, 417 (1952).

Procedure *B*

The second method for changing from the superconducting to the normal state consisted in raising the temperature above the transition temperature by means of a heater. This was done as shown in Fig. 2. A heater in the form of a 50-ohm, 0.1-watt carbon resistor was mounted on a copper strip above the lead disks and source. Below the source and disks which were spectroscopically pure lead 0.006 in. thick,[11] and mounted on the same strip, was another 50-ohm, 0.1-watt carbon resistor which was used to measure the temperature. The mounting strip was attached to a $\frac{1}{8}$-in. thin-wall brass can which was suspended from the top of the Dewars by a stainless steel tube. Helium gas at a pressure of a few millimeters served to establish thermal equilibrium between the lead and the helium bath.

The detailed procedure was to count double coin-

[11] These disks are slightly thinner than the maximum range in lead of the 0.54-Mev positron group from Na^{22} and they are considerably thinner than the maximum range of the 1.83-Mev positron group. However, only about 0.1% of the former are transmitted and the latter group is only 0.06% as strong as the former group. [See P. M. Endt and J. C. Kluyker, Revs. Modern Phys. **26**, 100 (1954).] Thus the background associated with these transmitted positrons makes a negligible contribution compared to the annihilation in the source background.

TABLE I. Ratio of the two-photon coincidence rate in normal lead to the rate in superconducting lead.

	Procedure Count alternately	Distance from crystal to source	Total number of coincidences counted	Ratio
A	with field of 750 gauss on and off	10 in. or 17 in.	36.6×10^6 [a]	1.0011[b] ±0.0016[c]
B	Count alternately with sample at 8.0°K and 4.2°K	24 in.	36.8×10^6	1.0003[d] ±0.0011[c]

[a] About the same number of coincidences were counted to measure the correction for the influence of the magnetic field on the counters.
[b] Corrected for influence of magnetic field on the counters, for annihilation in the source, and for accidentals.
[c] Standard deviation (statistical).
[d] Corrected for annihilation in the source and for accidentals.

cidences for five minutes at 4.2°K, then to raise the temperature to 8.0°K, and as soon as thermal equilibrium had been reached, which was about thirty seconds, to take another five minute count of doubles. The procedure was repeated until all the helium had evaporated (about 4 hours). Table I also shows the results of this experiment. The scintillation counters were 2-in.$\times1\frac{1}{2}$-in. diameter thallium-activated sodium iodide cylinders mounted on DuMont 6292 photomultiplier tubes. A Garwin[12] type coincidence circuit was used. The resolving time of the apparatus was about 2×10^{-7} sec. In this arrangement the accidental counts were about 6% of the total counts.

In order to check on the equipment, we repeated Pond's experiments with aluminum and polystyrene. The ratio of the two-quantum annihilation rate for positrons undergoing annihilation in aluminum compared to polystyrene was found to be 1.0046±0.0011 after correcting for annihilation in the source and backscattering from the polystyrene, and assuming that the collinear two-gamma annihilation probability is the same for positrons being annihilated in the source as for positrons being annihilated in the aluminum. This result is in reasonable agreement with Pond's measured value of 1.0055±0.0008 for this ratio. We were unable to obtain reproducible results without a lead collimator. However, one of the main advantages of the collimator is to make the apparatus less sensitive to slight displacements of the source. In neither procedure *A* nor *B* was there any necessity to change the position of the source, as is the case in the Pond experiment, so it was felt that the collimator was not necessary for the low-temperature experiments.

DISCUSSION

The purpose of our experiment was to search for evidence for the existence of orthopositronium in superconducting lead. The negative result which we obtained by procedure *B* is consistent with no *ortho*-positronium being formed when positrons stop in superconducting lead but by itself does not prove that none is formed.

[12] R. L. Garwin, Rev. Sci. Instr. **21**, 569 (1950).

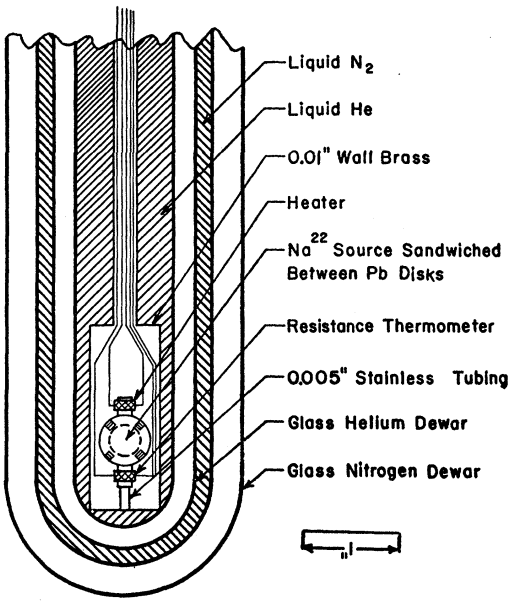

FIG. 2. Apparatus for changing the state of the lead sample from superconducting to normal by varying the temperature.

Our negative result is in agreement with Graham, Paul, and Henshaw,[4] and Green and Madansky,[13] who find no lifetime change, and with Stump[14] who finds no change in the angular correlation from normal to superconducting lead. The sensitivity of the method as a means for detecting *ortho*-positronium is discussed in the appendix.

APPENDIX

If one assumes that (a) positrons stopping in superconducting lead form *ortho*-positronium, but positrons stopping in normal lead do not; (b) annihilation probabilities for other processes such as direct three-quantum annihilation, etc., are unaffected by the state of the lead; (c) the backscattering coefficient is unaffected by the state of the lead; (d) the probability that a positron annihilates by two-quantum emission is the same regardless of whether it annihilates in the source or in normal state lead; and (e) $\lambda_2\gg\lambda_3$, then $f\lambda_3/\lambda_2=(N_2/S_2)-1$, where f is the fraction of the positrons stopping in superconducting lead which form *ortho*-positronium, λ_3 is the three-gamma decay rate of *ortho*-positronium in superconducting lead, λ_2 is the two-gamma decay rate associated with *ortho* to *para* conversion for positrons stopping in superconducting lead, N_2 is the two-gamma coincidence rate due to positrons stopping in normal lead, and S_2 is the two-gamma coincidence rate for positrons stopping in superconducting lead. Thus with a statistical error of 0.0011 as we had in procedure *B*, the quantity $f\lambda_3/\lambda_2$ must be greater than 0.0011 for us to observe an effect. This means, for example, that if 30% of the positrons formed

[13] B. Green and L. Madansky, Phys. Rev. **102**, 1014 (1956).
[14] R. Stump, Phys. Rev. **100**, 1256(A) (1955).

ortho-positronium in superconducting lead[15] and if its three-quantum decay rate were the same in superconducting lead as in vacuum ($1/\lambda_3=1.4\times10^{-7}$ sec), then the *ortho*-positronium must have a mean life ($\tau\approx1/\lambda_2$) greater than 5×10^{-10} sec in order for us to observe an effect.

[15] In most substances which show a long-lifetime component, about 30% of the positrons are responsible for it. [See R. E. Bell and R. L. Graham, Phys. Rev. **90**, 644 (1953).]

ACKNOWLEDGMENTS

It is a real pleasure to acknowledge the contributions of V. L. Telegdi who worked with us in conceiving and designing the experiment.

PHYSICAL REVIEW VOLUME 103, NUMBER 3 AUGUST 1, 1956

Electron-Nuclear Wave Functions in Multiphonon Processes

JORDAN J. MARKHAM
Research Division, Zenith Radio Corporation, Chicago 39, Illinois
(Received December 27, 1955)

For most purposes, the wave functions of a crystal with an electron trapped at an impurity are approximated by the product of two functions, one involving the lattice coordinates and the other the position of the electron. The electronic function may be determined for the equilibrium position of the lattice (static approximation) or one may assume that the electronic wave function continuously adjusts itself to the instantaneous position of the lattice (adiabatic approximation). The author relates the wave functions to Hamiltonian operators which do not have some of the most common operational properties. A comparison of the two approximations is made using the variational principle. The static underestimates the kinetic energy and overestimates the potential energy, while the second does just the reverse. Although the formal treatment is quite general and includes all the effects for harmonic vibrations, the actual terms were evaluated for a simple model. The calculations show that for extremely shallow traps the static approximation may be slightly superior, while for deep traps the adiabatic approach should be used. For traps of depths less than 0.1 ev the methods are essentially equivalent; in any actual calculations, other approximations must be made and they are of greater importance than the slight difference between either of these approaches.

1. INTRODUCTION

A GREAT many papers have appeared concerning electronic transitions which involve more than one phonon. The problem resolves itself into three parts. First, one must know what perturbation causes the transition; this requires a detailed knowledge of the electron-nuclear wave functions used. It is at this phase that one distinguishes between multiphonon and single-phonon processes. Second, we need to know the number of vibrational modes which enter into the transition; this depends on the energy distributions of the electrons and phonons. Finally, one must sum up the various individual probabilities.

In recent years progress has been made in the third phase of the problem[1] by assuming the validity of certain electron-nuclear wave functions. The calculations of Kubo[1] and Vasileff[1] use the so-called Born-Oppenheimer, or adiabatic approximation, which is presumably based on a paper of Born and Oppenheimer.[2] A search of the literature revealed, however, that the exact approximation first appeared in 1940.[3] The word "exact" is of utmost importance since one must know (in principle at least) precisely what wave functions are being used to carry through a multiphonon calculation. On the other hand, O'Rourke,[1] Frenkel,[4] Moglich and Rompe[4]; and Goodman, Lawson, and Schiff[4] used a second approach, originally formulated by Born and Oppenheimer.[5] The most appropriate name for the second approximation seems to be "static."[6-8]

There seems to be some feeling that the adiabatic approximation is "exact" (see Lax), and certainly superior to the static. With the possible exception of the book of Born and Huang,[7] the author has found nothing in the

[1] K. Huang and A. Rhys, Proc. Roy. Soc. (London) **A204**, 406 (1950); R. Kubo, Phys. Rev. **86**, 929 (1952); M. Lax, J. Chem. Phys. **20**, 1752 (1952); R. C. O'Rourke, Phys. Rev. **91**, 265 (1953); H. D. Vasileff, Phys. Rev. **96**, 603 (1954), **97**, 891 (1955). See also the work of S. I. Pekar Uspekhi Fiz. Nauk. **50**, 197 (1953) (English translation by M. D. Friedman of W. Concord, Massashusetts) and references therein.

[2] M. Born and J. R. Oppenheimer, Ann. Physik **84**, 457 (1927).

[3] F. Seitz, *The Modern Theory of Solids* (McGraw-Hill Book Company, Inc., New York, 1940), p. 470.

[4] J. Frenkel, Phys. Rev. **37**, 17 and 1276 (1931); F. Moglich and R. W. Rompe, Z. Physik **115**, 707 (1940); and Goodman, Lawson, and Schiff, Phys. Rev. **71**, 191 (1947).

[5] Strictly speaking, one need not assume that O'Rourke used the static approximation. In the notation to be developed, one need only assume that $\langle\varphi_{n'}(R)|\mathbf{M}|\varphi_n(R)\rangle$ is not a function of the nuclear coordinates. $\mathbf{M}$ is the electric moment operator for the system. This is not quite as restrictive as the static approximation. The above assumption is referred to as the Condon approximation. As will become evident shortly, the static approximation always leads to Condon's; the adiabatic may also in very special cases.

[6] Other names have been used in place of static, namely "harmonic" by Born and Huang[7] and "Hartree" by Lax.[8] One should not in any way relate this problem with the usual many-electron ones, where another Hartree approximation appears (reference 3, p. 234). The term "adiabatic" can have more than one meaning; the one used here agrees with Kubo, Vasileff, and Huang and Rhys. It does not agree, however, with the one used by Born and Huang.

[7] M. Born and K. H. Huang, *Dynamical Theory of Crystal Lattices* (Oxford University Press, Oxford, 1954), p. 166.

[8] M. Lax, *Photoconductivity Conference* (John Wiley and Sons, Inc., New York, 1956).