ise to give detailed information on the conduction-electron spin-flip correlation time in the ground state and will be considered in detail in a subsequent publication.

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[1]J. Kondo, Progr. Theoret. Phys. (Kyoto) 32, 37 (1964).

[2]P. W. Anderson, J. Appl. Phys. 37, 1194 (1966). Y. Nagaoka, Phys. Rev. 138, A1112 (1965); Progr. Theoret. Phys. (Kyoto) 37, 13 (1967). K. Yosida, Progr. Theoret. Phys. (Kyoto) 36, 875 (1966). F. Takano and T. Ogawa, Progr. Theoret. Phys. (Kyoto) 35, 343 (1966). J. R. Schrieffer, J. Appl. Phys. 38, 1143 (1967). J. Kondo, Progr. Theoret. Phys. (Kyoto) 36, 429 (1966).

[3]A. J. Heeger and M. A. Jensen, Phys. Rev. Letters 18, 488 (1967).

[4]G. J. Van den Berg, in Proceedings of the Ninth International Conference on Low Temperature Physics, edited by J. G. Daunt, D. O. Edwards, F. J. Milford, and M. Yaqub (Plenum Press, New York, 1965), p. 933; G. Knapp, J. Appl. Phys. 38, 1267 (1967); M. D. Daybell and W. A. Steyert, Phys. Rev. Letters 18, 398 (1967).

[5]M. A. Ruderman and C. Kittel, Phys. Rev. 96, 99 (1954); T. Kasuya, Progr. Theoret. Phys. (Kyoto) 16, 45 (1956); K. Yosida, Phys. Rev. 106, 893 (1957).

[6]Note that there is a correlated spin density within a coherence length of the impurity but this spin density, being part of the singlet, is flipping quantum mechanically at a high frequency and will not contribute to the Cu nmr linewidth. We also note that the spatial spin correlation found from the variational solution of Ref. 3 is identical with those found by Nagaoka (Ref. 2) and by Takano and Ogawa (Ref. 2).

[7]H. Suhl, Solid State Commun. 4, 487 (1966).

[8]J. Owen, M. Brown, W. D. Knight, and C. Kittel, Phys. Rev. 102, 1501 (1956); T. Sugarawa, J. Phys. Soc. (Japan) 14, 643 (1959).

[9]J. Friedel, Metallic Solid Solutions (W. A. Benjamin, Inc., New York, 1963), Chap. XIX.

[10]Takano and Ogawa, Ref. 2; A. J. Heeger and M. A. Jenson, to be published; K. Yosida and H. Ishii, to be published.

[11]J. P. Franck, F. D. Manchester, and D. L. Martin, Proc. Roy. Soc. (London) A263, 499 (1961).

[12]G. White, Can. J. Phys. 33, 119 (1955).

[13]M. W. Klein and R. Brout, Phys. Rev. 132, 2412 (1963).

[14]J. A. Cureaga, B. Dreyfus, R. Tournier, and L. Weil, in Proceedings of the Tenth International Conference on Low Temperature Physics (to be published).

[15]P. W. Anderson, Phys. Rev. 124, 41 (1961).

[16]It should be noted that the resistivity results are also in agreement with the resonant-scattering theory as worked out by H. Suhl and D. Wong, Physics 3, 17 (1967); H. Suhl, Phys. Rev. Letters 18, 743 (1967).

RELAXATION AND RECOMBINATION TIMES OF QUASIPARTICLES IN SUPERCONDUCTING Al THIN FILMS

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Recently[1] there has been considerable interest in the decay of excited quasiparticles in superconductors. It has been conjectured[1] that an excited quasiparticle of energy E decays via a two-step process. The first consists of a relaxation in time τ_T to energy Δ, where 2Δ is the energy gap of the superconductor. This process occurs primarily by emission of a phonon of energy $E-\Delta$.[2,3] The second step is recombination in time τ_R of two quasiparticles at energy Δ to form a Cooper pair and is accompanied by the emission of a phonon of energy 2Δ.[4,5]

We have measured for the first time τ_T in thin aluminum films at 0.37°K and find that τ_T decreases exponentially as a function of $(E-\Delta)/\Delta$ and is $\sim 0.5\times10^{-8}$ sec at $(E-\Delta)/\Delta = 1.0$. We have also measured τ_R as a function of temperature and find that at 0.37°K, $\tau_R \sim 1.0\times10^{-6}$ sec and that it decreases with increasing temperature up to $T \sim 1.2$°K. At temperatures above 1.2°K,[6] τ_R appears to increase

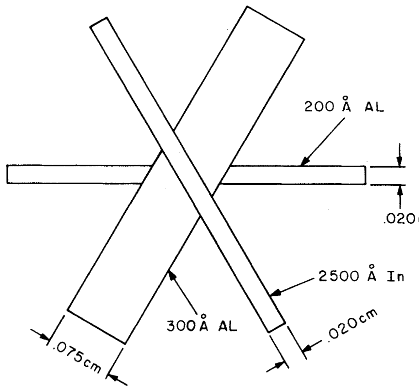

FIG. 1. Sample geometry.

slightly.

The measurements were performed on two superimposed superconducting tunnel diodes ($Al/Al_2O_3/Al/Al_2O_3/In$ in the present case) formed by the successive evaporation of three film stripes as shown in Fig. 1. Care was taken to insure that the top and bottom stripes were not in contact. The $Al/Al_2O_3/Al$ diode (diode 1) is biased such that quasiparticles are injected into the center aluminum stripe and, depending on the relaxation and recombination times, causes an increase in the steady-state quasiparticle distribution in the center film. This new distribution is probed by the second diode ($Al/Al_2O_3/In$) and shows up on its I-V characteristic.[7] In order to obtain a measurable change in the I-V curve due to injection, it is necessary to make the center stripe as thin as possible.

In Fig. 2 we show a series of I-V curves taken on diode 2 at 0.37°K for various voltages on diode 1. Here $\Delta_1 = 0.221$ mV and $\Delta_2 = 0.571$ mV. We note that for $V_1 = 2\Delta_1$ (curves 0 to 10) all the curves are similar to one another. For these biases on diode 1 all the injected quasiparticles are at $E \sim \Delta_1$ and the change in the quasiparticle population is proportional to τ_R. At biases where $V_1 > 2\Delta_1$ we note that in the region $V_2 < \Delta_2 - \Delta_1$ each curve deviates from its predecessor, in contrast to the similarity between the curves obtained at $V_1 = 2\Delta_1$. These deviations occur for the following reason. The injected quasiparticles are now in the energy range $\Delta_1 < E < V_1 - \Delta_1$. Because of the singularities in the quasiparticle density of states in the Al films, the injected quasiparticle distribution will have one singularity at $E = V_1 - \Delta_1$ and another at $E = \Delta_1$. The singularity at $E = V_1 - \Delta_1$ should, according to theory, produce a jump in the current I_2 at $V_2 = V_T = \Delta_1 + \Delta_2 - V_1$. The height of this jump should be proportional to $\tau_T(E)$. Experimentally this jump is rounded off and shows up as a deviation between the successive curves depicted in Fig. 2. The rounding off of the current jump is probably due to thermalization and the finite width of the In gap edge (30 μV).

In order to evaluate τ_R we consider only those curves corresponding to injection at $V_1 = 2\Delta_1$. For a given injection current I_1, the number of excited quasiparticles tunneling into the middle stripe is not proportional to I_1, but rather

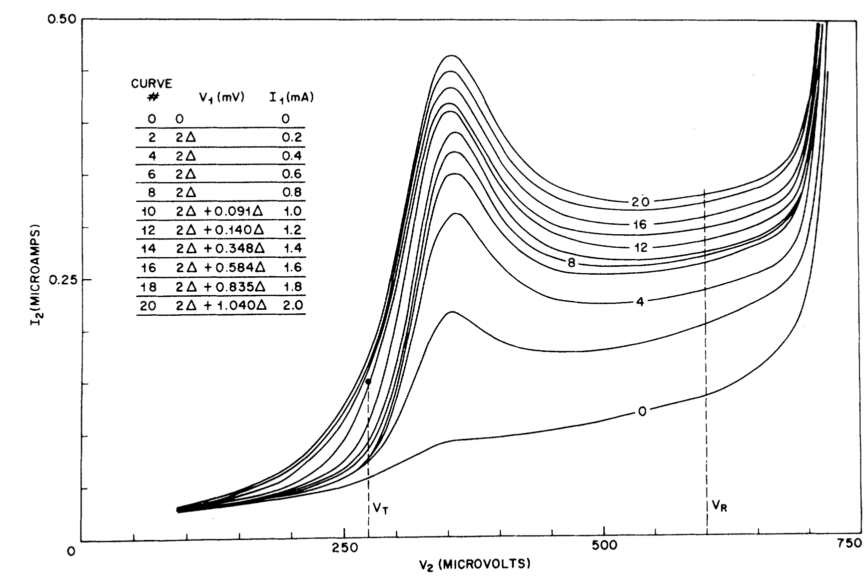

FIG. 2. I-V curve for diode 2 ($Al/Al_2O_3/In$) for different values of injection biases on diode 1 at 0.37°K. V_T is indicated for curve 14.

to $(I_1-I_1^0)$, where I_1^0 is the contribution of thermally excited particles. Referring to the semiconductor model of excitations in a superconductor, one can easily find that the current due to thermally excited particles is composed of two equal parts, one creating and the other destroying an equal number of excited quasiparticles in the middle film. We assume, therefore, that at $V_1=2\Delta_1$ all the injected quasiparticles lie in a narrow energy band at the top of the Al gap. In the steady state the change in the current I_2 at any bias $V_2 \geqslant \Delta_2-\Delta_1$ is proportional to $\tau_R(I_1-I_1^0)$. Because of the instability near the negative resistance region we choose a fixed bias $V_2=0.6$ mV which lies at a stable point in the temperature range of interest. At this bias the quasiparticles near the top of the Al gap tunnel to the In film into a fairly constant density of states whose value is 1.4 times the normal state density. With this in mind it is easy to show that the recombination time is given by

$$\tau_R=\frac{A_1}{A_{12}}\frac{ev(1)\Delta I_2(V_R)}{1.4C(I_1-I_1)}, \tag{1}$$

where $\Delta I_2(V_R)$ is the change of I_2 corresponding to a given injection current I_1; $v(1)$ and A_1 are the volume and area, respectively, of the second stripe common to diode 1; A_{12} is the area of the center stripe common to diodes 1 and 2 (see Fig. 1); and $C=\sigma_2(\infty)/2eN_1(0)$ is a constant for a given diode. Here $\sigma_2(\infty)$ is the conductivity of diode 2 in the high-voltage limit, and $2N_1(0)=3.33\times10^{22}$ $\mathrm{eV^{-1}\,cm^{-3}}$ is the normal density of states for both spins in Al.[8] In our case $C=1.97\times10^{-24}$ $\mathrm{C\,sec^{-1}\,cm^3}$ for the diode used in run 12.

In order to obtain $\tau_T(E)$ it is necessary to compute the jump in current I_2 expected at $V_2=V_T=\Delta_1+\Delta_2-V_1$ and then compare with the experimental jump. From tunneling theory[9] one finds that the jump in current is proportional to

$$W=\lim_{\epsilon\to0}\frac{V_1-\Delta_1}{[(V_1-\Delta_1)^2-\Delta_1^2]^{1/2}}\int_{V_1-\Delta_1-\epsilon}^{V_1-\epsilon}\frac{(E-V_1)(E+V_T)dE}{\{[(E-V_1)^2-\Delta_1^2][(E+V_T)^2-\Delta_2^2]\}^{1/2}}=\frac{V_1-\Delta_1}{[(V_1-\Delta_1)^2-\Delta_1^2]^{1/2}}\frac{\pi}{2}(\Delta_1\Delta_2)^{1/2}.$$

If we denote the measured increment in current at $V_T=\Delta_2+\Delta_1-V_1$ by $\Delta I_2(V_T)$ then

$$\tau_T(V_1-\Delta_1)=\frac{\Delta I_2(V_T)e^3N_1(0)v(1)}{W\sigma_1(\infty)\sigma_2(\infty)}\frac{A_1}{A_{12}}. \tag{2}$$

Since we are interested in the steady-state population at $E=V_1-\Delta_1$ due to the relaxation from $E=V_1-\Delta_1$, any excess population from thermal tails extending up from energies $E<V_1-\Delta_1$ must be taken out. We find that the best way to achieve this is to take $\Delta I_2(V_T)$ as the difference in current at V_T between the given curve and its predecessor, the latter current being nearly equivalent to the thermal tail originating from energies $E<V_1-\Delta_1$.

In Fig. 3(a) τ_R is plotted as a function of temperature. τ_R was determined by plotting $\Delta I_2(V_R)$ vs I_1 at various temperatures and taking the slope of this curve as $(I_1-I_1^0)$ goes to 0. Thus we obtain the dependence of τ_R on temperature in the limit of zero injection. The temperature dependence of τ_R has been calculated by Schrieffer who finds that for $\Delta/kT\gg1$

$$\tau_R\sim(\Delta/kT)^{1/2}\exp(\Delta/kT),$$

which gives a much stronger temperature dependence than that indicated in Fig. 3(a). We find that for $\Delta/kT>3$, $\tau_R\sim\exp(0.3\Delta/kT)$. At present we are unable to account for this discrepancy.

In Fig. 3(b) we plot τ_T vs $(E-\Delta_1)/\Delta_1$ at 0.37°K. We find that $\tau_T(E-\Delta)=1.11\times10^{-7}\exp[-3.34(E-\Delta)/\Delta]$. The fact that τ_T is at least an order of magnitude less than τ_R means that all injected quasiparticles first relax to the top of the gap and then recombine as discussed earlier. In the present experiment it is difficult to measure τ_T as a function of temperature because of the large thermal quasiparticle population at higher temperatures.

Several factors have to be taken into account in determining τ_R and τ_T by the present method. These are (1) heating of the diode by the dc power input, (2) diffusion of the quasiparticles away from the common area of the diode, and (3) loss of excited quasiparticles in the center film due to straight through tunneling[10] where an injected quasiparticle traverses the second film and tunnels directly into the top

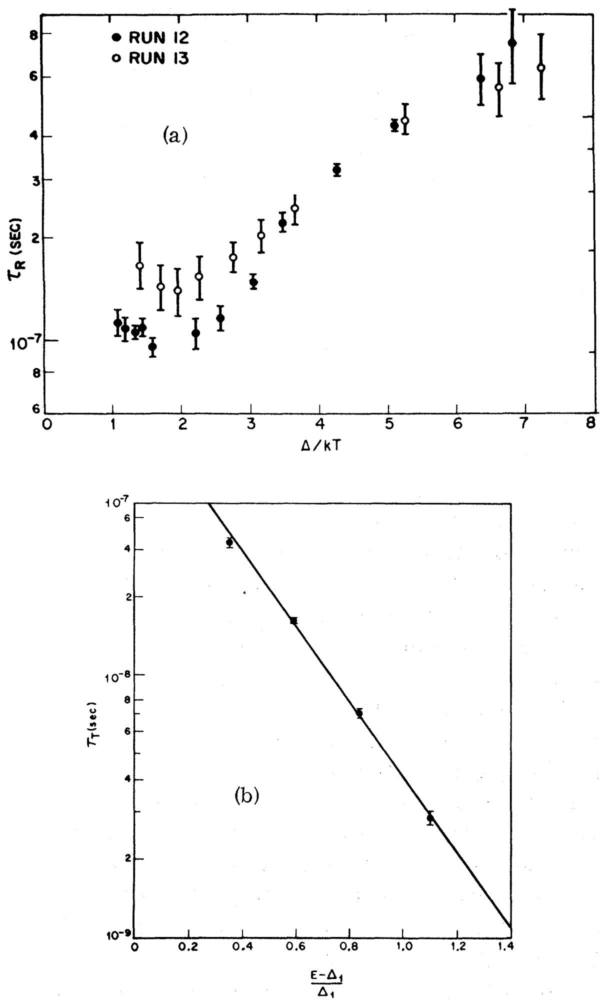

FIG. 3. (a) Plot of τ_R vs Δ/kT. The indicated errors are relative errors only. (b) Plot of τ_T vs $(E-\Delta)/\Delta$ at 0.37°K. The indicated errors are relative only.

film (In). Heating will cause the total number of quasiparticles to rise and will thus cause τ_R and τ_T to appear larger than they are. A series of measurements were made on the rise in temperature of the middle film as a function of injected power by noting the change of the Al energy gap, as measured by diode 1, as a function of input power from diode 2. In this way we were able to estimate the heat rise of the middle film and we find that any error introduced by heating is less than 10% at the injection levels used in determining τ_R and τ_T. Diffusion will reduce the number of quasiparticles present and thus cause τ_R and τ_T to appear smaller than they are. We find the diffusion length of quasiparticles in the middle film to be about 2×10^{-3} cm, much smaller than the width of the stripes. Diffusion errors are thus only about 10%. The straight-through tunneling process will give rise to an excess current I_2' in addition to I_2 and makes τ_R and τ_T appear larger. We find that $I_2'/I_2 < 10^{-5}$. The over-all absolute errors in measuring τ_R and τ_T are about 30% and come mainly from error in measuring film thickness. We also studied the variation of $\Delta I_2(V_R)$ over a wide range of injection current I_1 at various temperatures. The curves obtained have a number of interesting features which will be described in a later publication. Among these features we find the following: (1) $\Delta I_2(V_R) = 0$ for $I_1 - I_1^0 \leq 0$ showing that indeed the current from thermally excited quasiparticles does not produce net excitations in the middle film. (2) For the range of values of I_1 at $V_1 \sim 2\Delta_1$, the behavior of $\Delta I_2(V_R)$ is such as to show clearly the decrease of τ_R with increasing steady state quasiparticle population. This is particularly prominent at low temperatures where the original thermal population is relatively small.

In conclusion, we have developed a method by which we have measured τ_R and τ_T as a function of temperature and energy, respectively. In addition to this we are able to measure the dependence of τ_R on quasiparticle density at a given temperature and the energy profile of the excited quasiparticles. By using these techniques we are better able to understand the quasiparticle relaxation and recombination processes in superconductors. In the future we plan a further study of Al as well as other superconductors using these techniques.

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[1]W. Eisenmenger and A. H. Dayem, Phys. Rev. Letters 18, 125 (1967).

[2]J. Bardeen, G. Rickayzen, and L. Tewordt, Phys. Rev. 113, 982 (1959).

[3]L. Tewordt, Phys. Rev. 127, 371 (1962); 128, 12 (1962).

[4]J. R. Schrieffer and D. M. Ginsberg, Phys. Rev. Letters 8, 204 (1962); D. M. Ginsberg, Phys. Rev. Letters 8, 207 (1962).

[5]It is also possible for the recombination to occur via photon emission. E. Burstein, D. N. Langenberg, and B. N. Taylor [Phys. Rev. Letters 6, 92 (1961)] have

shown that photon emission lifetime ~0.4 sec in Pb which is much smaller than the phonon lifetime $\sim 10^{-7}$ sec (see Ref. 4).

[6]The transition temperature for these films was ~1.45°K. We find that $2\Delta(0)/kT_c \sim 3.5$. See B. Abeles, R. W. Cohen, and G. W. Cullen, Phys. Rev. Letters 17, 631 (1966).

[7]This is the same method used by B. Taylor to measure recombination times in Al. B. Taylor, thesis, University of Pennsylvania, 1962 (unpublished).

[8]T. P. Sheenen, Phys. Rev. 149, 370 (1966).

[9]I. Giaever and K. Megerle, Phys. Rev. 122, 1101 (1961).

[10]B. N. Taylor, Private communication.

HOT-ELECTRON–PHONON INTERACTIONS IN GaP

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In a recent Letter, Williams and Simon[1] claim to have determined by direct measurement the mean free path for energy loss to phonons of energetic electrons in GaP. The mean free path they obtain is, however, of questionable significance since it is based on a greatly oversimplified view of hot-electron transport in this material. This is true also of previous determinations of this mean free path.[2]

It is assumed by Williams and Simon that the energy loss per collision is 0.050 eV, the longitudinal optical (LO) phonon energy at the center of the Brillouin zone.[3] This type of phonon may well be predominant for losses of low-energy electrons. Polar optical scattering is known to be important at low fields in GaP at room temperature.[4,5] It is not, however, the only important scattering process, even in relatively pure material, since the mobility calculated for this mechanism is greater than the observed mobility.[4,5] It has been speculated that acoustic deformation-potential scattering is the other process of importance,[5] but it could well be that that process is scattering among the equivalent (100) valleys. The latter is the predominant scattering process for the (100) minima in silicon. In support of this hypothesis, Epstein[6] notes that the Hall mobility at 300° and above shows a somewhat stronger temperature dependence than could be accounted for by a combination of polar and intravalley acoustic-mode scattering.

The rate of energy loss of an electron of energy $\mathcal{E}$ to polar modes is[7]

$$\left(\frac{d\mathcal{E}}{dt}\right)_{\rm po} = -\frac{2eE_0\hbar\omega_l}{(2m\mathcal{E})^{1/2}}\left[(N_{ql}+1)\cosh^{-1}\left(\frac{\mathcal{E}}{\hbar\omega_l}\right)^{1/2} - N_{ql}\sinh^{-1}\left(\frac{\mathcal{E}}{\hbar\omega_l}\right)^{1/2}\right], \tag{1}$$

where e is the electronic charge; ω_l and N_{ql} the angular frequency and number, respectively, of LO phonons; and E_0 an effective field whose magnitude is a measure of the coupling to the LO phonons. If (1) is plotted versus $\mathcal{E}$, it is found that for the slowest electrons $(d\mathcal{E}/dt)_{\rm po} > 0$, indicating net gain. As $\mathcal{E}$ increases, $(d\mathcal{E}/dt)_{\rm po}$ becomes negative and its magnitude increases. The magnitude of the loss rate reaches a maximum and then decreases as $\mathcal{E}$ increases further, eventually as $\ln\mathcal{E}/\mathcal{E}^{1/2}$. For GaP this maximum is reached at $\mathcal{E} \simeq 4\hbar\omega_l$ or 0.2 eV. The rate of loss of electrons in the ith valley due to intervalley scattering is given by[8]

$$\left(\frac{d\mathcal{E}}{dt}\right)_{\rm iv} = \sum_j \frac{D_{ij}^{\,2}(\overline{m}_j)^{3/2}}{2^{1/2}\pi\hbar^2\rho}\left[(N_{qij}+1)(\mathcal{E}-\hbar\omega_{ij}-\mathcal{E}_{0j})^{1/2} - N_{qij}(\mathcal{E}+\hbar\omega_{ij}-\mathcal{E}_{0j})^{1/2}\right], \tag{2}$$

where the sum is taken over all possible final valleys; D_{ij} and ω_{ij} are the coupling constant and angular frequency, respectively, for the transition $i \rightleftarrows j$; N_{qij} is the number of phonons with angular