Quasiparticle Trapping from a Single-Crystal Superconductor into a Normal-Metal Film via the Proximity Effect

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(Received 14 August 1989)

We have studied the dynamics of the nonequilibrium state produced in single-crystal superconducting indium by pulsed laser irradiation using tunnel junctions. By a suitable choice of a thin metal film in intimate contact with the indium crystal, we have been able to produce a pronounced change in the time evolution of the excess quasiparticle density which indicates that the quasiparticles are trapped in the film. This effect can take place both when the film is another superconductor of lower energy gap or when it is a normal metal made superconducting by the proximity effect.

PACS numbers: 74.50.+r, 29.40.−n, 73.40.−c

The interaction of a nuclear particle or a photon with a superconductor at low temperatures leads within a few picoseconds to a metastable state consisting of broken Cooper pairs or quasiparticles (electrons) and low-energy phonons. The energy required to break a pair is of the order of 2Δ, where Δ is the superconducting energy gap. For typical superconductors with an energy gap of order 1 meV this implies that 10^3 times more electronic excitations are produced than in more conventional semiconductor detectors. The quasiparticles diffuse in the superconductor with a diffusion coefficient which is determined by the electronic mean free path and are ultimately lost from the system by the loss of recombination phonons. Phonons having energies $\Omega < 2\Delta$ interact rarely with the quasiparticles and propagate ballistically with a velocity determined by the sound velocity. A superconducting tunnel junction (STJ) can be used to detect these excitations. The magnitude of the signal current from a STJ, when biased below the sum of the energy gaps and exposed to a source of excitation, is inversely proportional to the volume of the films of the junction. For a volume of superconductor sufficient to contain the interaction of a γ ray the signals are undetectable. In order to enhance the signal in the case of large crystals of a superconductor, Booth has proposed the idea of quasiparticle trapping where the excess of quasiparticles created in the superconductor by an interaction can be trapped in a smaller volume of a smaller-gap superconductor.[1] The trapping time is proportional to the scattering time τ_s for a quasiparticle of energy E ($\approx \Delta$) to relax to close to the energy gap of the trap Δ_{trap} by phonon emission. For E greater than several times Δ_{trap}, this scattering time varies approximately as $(E/\Delta_{trap})^{-3}$ and scales as the characteristic time τ_0.[2] Booth gives for the trapping time

$$\tau_{trap} = (V_{crystal}/V_{trap})\tau_s , \tag{1}$$

where $V_{crystal}$ and V_{trap} are the volumes of the crystal and trap, respectively. The trap material will also be sensitive to phonons in the energy band $2\Delta_{trap} \leq \Omega < 2\Delta_{crystal}$. We present here the first systematic study of quasiparticle trapping.

In order to demonstrate the quasiparticle trapping mechanism we report here on a series of experiments which investigate the propagation and time evolution of excess quasiparticles in high-purity single-crystal superconducting indium. The propagation of phonons in the same system has been investigated using Al-Al and Al-Pb STJ's. Subsequently we identify the necessary conditions to observe quasiparticle trapping in both Al and Cu films deposited on the indium and demonstrate the dependence of the trapping time on trap thickness. For trapping in Cu films the signal amplitude and decay time are dramatically changed.

Single crystals of In of dimensions $10\times10\times2.5$ mm^3 and mass 1.8 g are grown on sapphire substrates using a vertical Bridgman technique and then chemically cleaned and electropolished. Tunnel junctions are evaporated onto the top surface after ion-beam milling within the same vacuum cycle. If desired, this surface can be ion-beam oxidized to form a tunneling barrier directly on the crystal. To form a trap, either Al or Cu (with a thin, 25-nm, Al surface layer) is deposited onto the cleaned crystal surface and the tunnel barrier is formed by oxidizing the Al surface. For all geometries SiO is used to define junction areas of typically 0.2 mm^2. The probe electrodes are Al or Pb. Junction resistances are in the range 0.1–500 Ω mm^2. The samples are mounted in a helium-3 cryostat which gives base temperatures of 350 mK. Four-terminal electrical connections are made to allow current biasing and voltage measurement. Low-noise amplifiers are used with an integration time between 50 and 200 ns. Signal averaging for 1000 pulses gives an effective noise of less than 20 nV rms. The junctions are current biased in a region of low dynamic resistance to avoid long electronic time constants. Quasiparticles and phonons are created in the In by a pulsed (< 200 ns) solid-state laser which is directed by

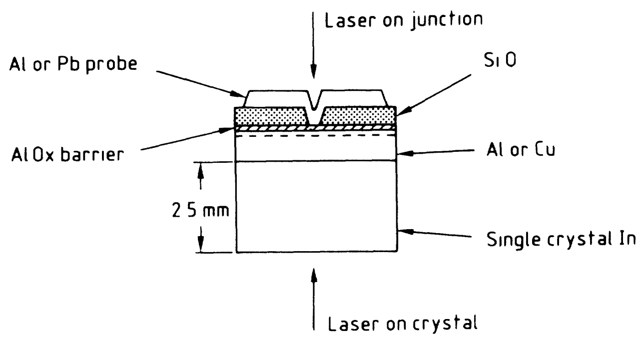

FIG. 1. Schematic of the experimental arrangement (not to scale). For devices using Cu as a trap an additional layer of Al is used to form the tunneling barrier.

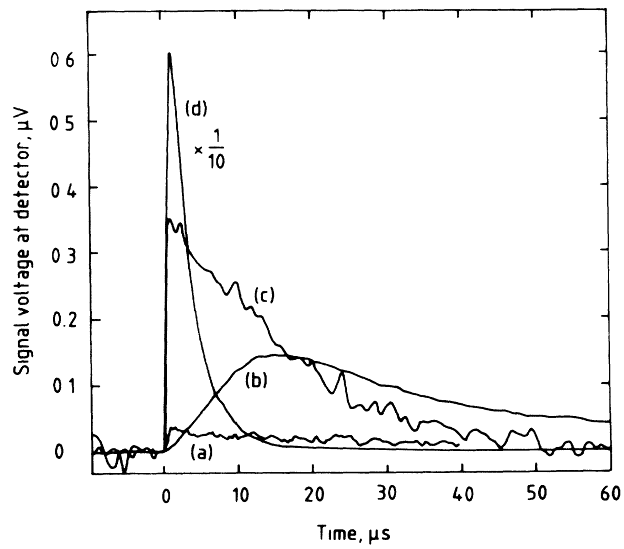

FIG. 2. Voltage pulse following 200-ns laser impulse onto the crystal. Curve *a*, quasiparticle pulse from In(crystal)-oxide-Pb; curve *b*, phonon pulse from In(crystal, not milled)-Al-oxide-Pb; curve *c*, quasiparticle pulse from In(crystal)-Al(trap)-oxide-Al; curve *d*, quasiparticle pulse from In(crystal)-Cu(trap)-(Al)-oxide-Al. This trace is reduced by a factor of 10 compared to *a*–*c*.

an optical fiber onto the surface of the crystal opposite the junction. A second fiber is mounted above the junction to allow measurement of the lifetimes of direct excitations in the STJ films. Figure 1 shows a schematic of the experimental arrangement.

When the crystal surface is oxidized and a larger-gap superconductor such as Pb is used for the probe electrode, the STJ is sensitive only to quasiparticles and to phonons of energy $\Omega \geq 2\Delta_{\text{In}}$. Figure 2, curve *a*, shows the voltage pulse produced by laser excitation for a device using Pb as the probe film at 491 mK. From the arrival time of the pulse[3] we can deduce a value for the diffusion coefficient of the quasiparticles, $D_{\text{qp}}=10$ m^2s^{-1}. This implies an impurity mean free path of 110 ± 10 μm which is in good agreement with estimates using a free-electron model and a residual-resistance ratio of 10000. We interpret the decay time of the pulse as the effective lifetime $\tau_r^* = 37$ μs of the quasiparticle excitation in the In crystal at this temperature.

When an Al-oxide-Al tunnel junction is formed on the crystal without ion milling the crystal surface, we find that there exists a barrier sufficient to strongly impede the diffusion of quasiparticles from the In crystal into the first Al film even though there is no measurable electrical resistance. This conclusion is reinforced by the fact that we obtain very similar results when a layer of SiO is deposited between the In crystal and the junction. Consequently, the junction detects only phonons with energy $\Omega \geq 2\Delta_{\text{Al}}$. Figure 2, curve *b*, shows the pulse observed at 369 mK. The decay time of this phonon pulse and its temperature dependence are the same as those of the quasiparticle pulse of Fig. 2, curve *a*. This implies that the phonons above the detector threshold of 360 μeV are predominantly produced by the same mechanism responsible for the quasiparticle decay. The mechanism appears to be the decay of recombination phonons in the In into two phonons of energy approximately Δ_{In} which subsequently decay with an average lifetime of 8.25 ± 0.25 μs to an energy below the 360-μeV threshold. The pulse shape of Fig. 2, curve *b*, can be fitted using this decay scheme. This lifetime is not too different from the value of about 4 μs calculated using the expression due to Klemens[4] for the anharmonic decay of longitudinal phonons in In at 500 μeV. This interpretation is reinforced by results obtained using Al and Ti superconducting bolometers which indicate that the number of phonons is increasing on these time scales.

When the In crystal is ion milled prior to deposition of the Al, quasiparticles are detected. This is indicated by the arrival time of the leading edge of the voltage pulse of Fig. 2, curve *c*, which is the same as for the quasiparticle pulse of Fig. 2, curve *a*. In all devices with good quasiparticle transmission we observe changes in the current-voltage characteristics of the junctions which indicate that the Al trap has the T_c of the bulk In, and that the value of Δ_{Al} is enhanced at low temperatures. Figure 3 shows the observed temperature dependence of the two peaks in the Al density of states for a thickness of 650 nm Al. The low-temperature energy gap is 230 μeV for this device. For comparison, the dashed curve shows the usual BCS temperature dependence of the Al energy gap. The solid lines are fits to the data using the McMillan model of the proximity effect.[5] In this model the interface between the two metals is considered as a barrier through which the electrons, which experience the long-range pairing interaction in the superconducting state, tunnel with a probability σ and thus modify the energy gap and density of states in the Al film. We conclude that *only* when these modifications are observed can quasiparticles be transmitted and trapped. The fit of

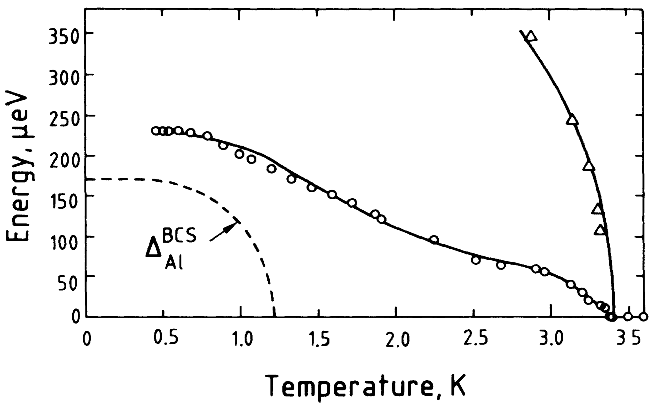

FIG. 3. Temperature variation of the energy of the two peaks in the Al density of states for 650-nm Al film on the In crystal surface. The solid lines are from a fit using McMillan's model of the proximity effect. The dashed curve shows the usual BCS temperature dependence of the Al energy gap.

Fig. 3 gives a value $\sigma=0.13$. The reduction in τ_{qp}, the quasiparticle-number decay time, is evident by comparing the decay times of Fig. 2, curves *a* and *c*. Taking

$$\tau_{qp}^{-1}=\tau_r^{*\,-1}+\tau_{trap}^{-1}, \qquad (2)$$

where $\tau_r^{*\,-1}$ is the quasiparticle decay rate at the appropriate temperature in the crystal without trapping, we deduce a value for τ_{trap} of 39 ± 3 μs. Using Eq. (1) the scattering time τ_s is found to be 10 ± 1.2 ns and a value of τ_0 for the Al film can then be calculated using the expression of Kaplan *et al.*[2] for τ_s/τ_0. We find $\tau_0=0.11\pm0.013$ μs which is very close to the value measured by Chi and Clarke for clean Al films.[6]

Because of the proximity effect, we are not restricted to a very limited number of superconductors for the trap material, but can use a normal metal with a very small induced energy gap. We have tried several metals but have studied only Cu extensively.

For a typical device with a Cu thickness of 250 nm and Al as a probe, the Cu has a low-temperature energy gap of 24 μeV, and a fit using McMillan's model gives a transmission probability $\sigma=0.048$ for the In-Cu interface. The voltage pulse, shown in Fig. 2, curve *d*, has an arrival time characteristic of quasiparticle diffusion, but the decay time is much shorter than that of any of the previous pulses. Using Pb as a probe electrode, the temperature dependence of the amplitude of the signal from the Cu is determined by the recombination time in the Cu. Results are shown in Fig. 4(a), where the solid curve comes from the recombination time calculated using Eq. (8) of Ref. 2 and the measured Cu energy gap. The decay time of the pulses from direct laser irradiation of the STJ has this same temperature dependence. The calculation assumes that the phonon trapping factor which enhances the apparent quasiparticle lifetime in a superconducting film is negligible. This assumption is

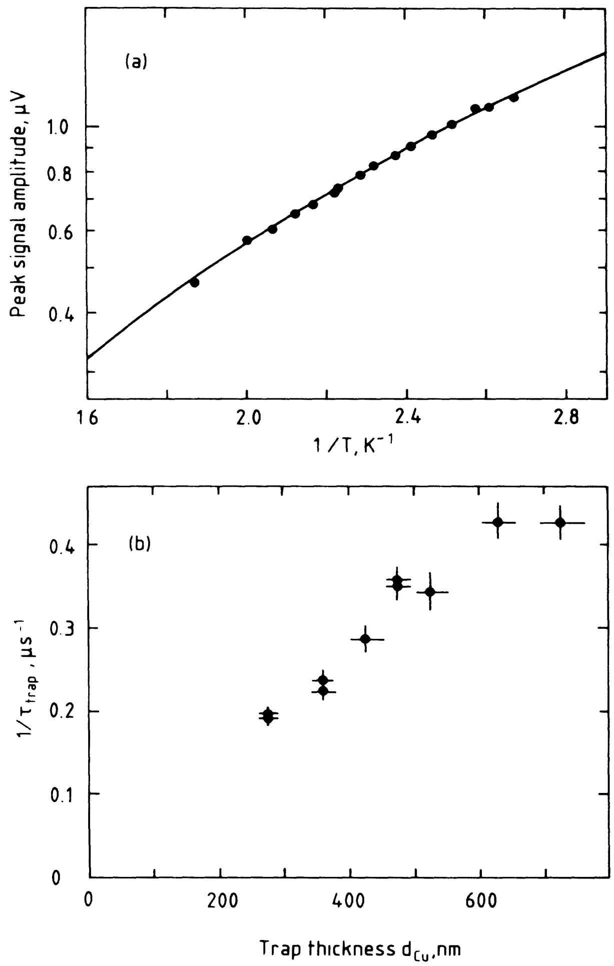

FIG. 4. (a) Temperature dependence of the peak signal from a device using Cu as a trap. The line is a fit using the calculated quasiparticle recombination lifetime in the proximity layer. (b) Plot of trapping rate vs trap thickness for a number of devices with Cu traps.

consistent with an estimate by Long[7] of the phonon mean free path in Cu films (1.2 μm at $\Omega=400$ μeV) and also with the null observation of phonon pulses in this device. We have studied the phonon spectrum emitted in trapping using Al-Pb STJ's, electrically isolated from the trap, as energy-selective phonon detectors.[8] These results demonstrate the production of phonons of energy $\Delta_{In}-\Delta_{trap}$ and reinforce our conclusion that the pulse of Fig. 2, curve *d*, is due to detection of quasiparticles which diffuse in the In crystal and are trapped by the Cu.

Using a suitable diffusion function[9] to model the quasiparticle propagation, the pulses can be fitted and a value for τ_{qp} obtained. Using Eq. (2) we deduce a trapping time τ_{trap} of 5.1 ± 0.2 μs for the pulse of Fig. 2, curve *d*. This is several times shorter than values obtained with Al films of similar thicknesses. We also note that the amplitude of the voltage pulse of Fig. 2, curve *d*, measured at the detector is much larger than that of any of the previous pulses.

We have determined $\tau_{\rm trap}$ for several devices with Cu thicknesses $d_{\rm Cu}$ in the range $250 \le d_{\rm Cu} \le 700$ nm and with low-temperature energy gaps ranging up to 50 μeV. Figure 4(b) shows $1/\tau_{\rm trap}$ plotted as a function of film thickness. The results are consistent with a straight line through the origin, justifying the use of Eq. (1), and give a value for τ_s of 0.62 ± 0.06 ns. Using the expression of Kaplan *et al.* for τ_s/τ_0 with $T_c = 3.40$ K we obtain a value for τ_0 of 1.06 ± 0.10 ns for Cu films with proximity effects.

In conclusion, we have investigated the time evolution of the quasiparticles and phonons produced by pulsed laser irradiation in bulk superconducting In. The dominant production mechanism for phonons in the range $360 < \Omega < 1050$ μeV is from the decay of recombination phonons of energy $2\Delta_{\rm In}$. The quasiparticle decay time is modified by the presence of a proximity metal layer of lower energy gap when the interface between the In crystal and the metal layer is clean. The McMillan model gives a good account of observed energy gaps in this layer. The trapping rate is proportional to film thickness. Trapping occurs both when the layer is a superconductor of lower energy gap or when it is a normal metal made superconducting by the proximity effect.

We would like to express our sincere thanks to B. M. Hawes and A. Wire for their technical expertise. One of us (D.J.G.) acknowledges the financial support of the Atomic Energy Research Establishment (Harwell) during the earlier part of this work.

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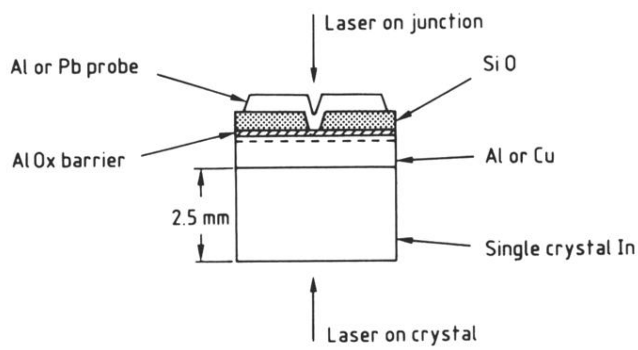

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