

spin-glass. EPR measurements are, in this case, especially useful in investigating the critical dynamics of the spin-glass transition. Anomalies in the EPR spectra have been noted over the years in a variety of alloys,⁵ but were not pursued. It is quite clear that careful remeasurement of the linewidth and g -value anomalies on other well-characterized spin-glass alloys will lead to further clarification of the dynamical nature of the freezing transition.

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¹S. F. Edwards and P. W. Anderson, *J. Phys. F* **5**, 965 (1975); D. Sherrington and S. Kirkpatrick, *Phys. Rev. Lett.* **35**, 1792 (1975).

²A. P. Murani, *J. Appl. Phys.* **49**, 1604 (1978); D. A. Levitt and R. E. Walstedt, *Phys. Rev. Lett.* **38**, 178 (1977).

³S.-k. Ma and J. Rudnick, *Phys. Rev. Lett.* **40**, 589 (1978).

⁴P. W. Anderson and P. R. Weiss, *Rev. Mod. Phys.* **25**, 269 (1953).

⁵A. Nakamura and N. Kinoshita, *J. Phys. Soc. Jpn.* **22**, 335 (1967); A. C. Gossard, T. Y. Kometani, and J. H. Wernick, *J. Appl. Phys.* **39**, 849 (1968); F. W. Kleinhans and P. E. Wigen, in *Magnetism and Magnetic Materials—1971*, AIP Conference Proceedings No. 5, edited by C. D. Graham, Jr., and J. J. Rhyne (American Institute of Physics, New York, 1972), p. 1204.

⁶K. Kawasaki, *Prog. Theor. Phys.* **39**, 285 (1968), and *Phys. Lett.* **26A**, 543 (1968).

⁷R. W. Tustison, *Solid State Commun.* **19**, 1075 (1976).

⁸R. W. Tustison and P. A. Beck, *Solid State Commun.* **20**, 841 (1977).

⁹P. A. Beck, to be published.

¹⁰D. L. Huber, *J. Phys. Chem. Solids* **32**, 2145 (1971).

¹¹K. Kawasaki, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, London, 1976), Vol. 5a, p. 322.

¹²S. Maekawa, *J. Phys. Soc. Jpn.* **33**, 573 (1972).

¹³J. Kötztler and H. von Philipsborn, *Phys. Rev. Lett.* **40**, 790 (1978).

¹⁴A. B. Harris, T. C. Lubensky, and J.-H. Chen, *Phys. Rev. Lett.* **36**, 415 (1976).

¹⁵J. A. Hertz and R. A. Klemm, *Phys. Rev. Lett.* **40**, 1397 (1978).

Direct Measurement of Quasiparticle-Lifetime Broadening in a Strong-Coupled Superconductor

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We have measured the quasiparticle recombination time in the strong-coupled superconductor $\text{Pb}_{0.9}\text{Bi}_{0.1}$ directly by measuring the lifetime-broadened energy gap edge. This is done by measuring the I - V characteristics of a superconducting tunnel junction of the type $\text{Pb}_{0.9}\text{Bi}_{0.1}$ -insulator- $\text{Pb}_{0.9}\text{Bi}_{0.1}$. Agreement with the calculated value is excellent.

Nonequilibrium properties in superconductors have been the subject of substantial interest recently.¹ The response of a superconductor when driven away from equilibrium is very much dependent upon the various electron and phonon relaxation times. Central to the relaxation phenomenon is the recombination time τ_R which is the time taken for two quasiparticles at or near the energy gap edge Δ to recombine into the superfluid condensate. In this Letter we demonstrate that τ_R can be measured in a strong-coupled superconductor directly by measuring the lifetime-broadened energy gap width in a tunneling experiment. Measurements for the alloy $\text{Pb}_{0.9}\text{Bi}_{0.1}$ are presented and good agreement with calculations is achieved.

Previous measurements of the intrinsic quasiparticle recombination time in a superconductor have been plagued by thin-film phonon bottlenecking problems or have been rather complicated, requiring model analysis of data.^{1,2} Very recently, a determination of τ_R in single-crystal Pb was made by measuring the thermal diffusivity and extracting a quasiparticle scattering rate which could be related to the recombination time.² In thermal equilibrium well away from the superconducting critical temperature T_c the recombination rate is given by¹

$$\frac{1}{\tau_R} = \left(\frac{T}{\Delta}\right)^{1/2} \frac{1}{\tau_0} e^{-\Delta/kT}, \quad (1)$$

where τ_0 is related to the electron-phonon coup-

ling strength, and for Pb excellent agreement with the calculations of Kaplan *et al.*³ was obtained.² The strong temperature dependence comes from an occupation term due to the fact that two particles are required in the recombination process. The short lifetimes at temperatures $T/T_c > 0.5$ correspond to a linewidth which is a measurable fraction of Δ and hence one should expect in strong-electron-phonon-coupled superconductors a temperature-dependent smearing of the energy gap edge.

To measure this lifetime smearing, we have fabricated a symmetric superconducting tunnel junction of the form $\text{Pb}_{0.9}\text{Bi}_{0.1}$ -insulator- $\text{Pb}_{0.9}\text{Bi}_{0.1}$. The PbBi alloy was chosen for two reasons. Firstly, tunneling measurements have been performed on $\text{Pb}_{0.9}\text{Bi}_{0.1}$ and the superconducting properties of PbBi alloys [including the electron-phonon coupling function $\alpha^2(\omega)F(\omega)$] are known.⁴ In addition, using the $\alpha^2(\omega)F(\omega)$ from these measurements, Kaplan *et al.*³ have calculated a value for τ_R to be even shorter than in the case of Pb. Secondly, the intrinsic gap widths (due to anisotropy effects) are substantially reduced in alloys with short mean-free paths.⁵ The energy gap width at low temperatures ($T/T_c < 0.3$) in alloys is very small and thus any enhancement due to lifetime effects would be more easily measured. The tunnel junctions studied were evaporated thin films of $\text{Pb}_{0.9}\text{Bi}_{0.1}$ of thickness $\approx 2000 \text{ \AA}$ and had normal-state resistances ranging from 0.1 to 100 Ω . The area of the junctions was 0.25 mm^2 . The oxide barrier was produced by use of previously reported techniques⁶ and only junctions of high quality (displaying very small leakage currents) were measured. The experiment consisted of measurements of the current-voltage (I - V) characteristics of a tunnel junction at various temperatures between 1.0 K and T_c (7.65 K).

In Fig. 1 we show a typical set of I - V characteristics for various temperatures between 1.0 and 7.0 K and we note several features. The temperature dependence of the energy gap 2Δ corresponding to the point of maximum slope in the I - V characteristic is easily observed in this data. For large smearing, it is no longer true that the peak in dI/dV corresponds to 2Δ , but for broadening of the magnitude observed here, it is correct to equate the two. Also, the presence of thermally activated quasiparticles seen as current flow at energies (or voltages) less than 2Δ at the higher temperatures and the freeze-out of these excitations at low temperatures is

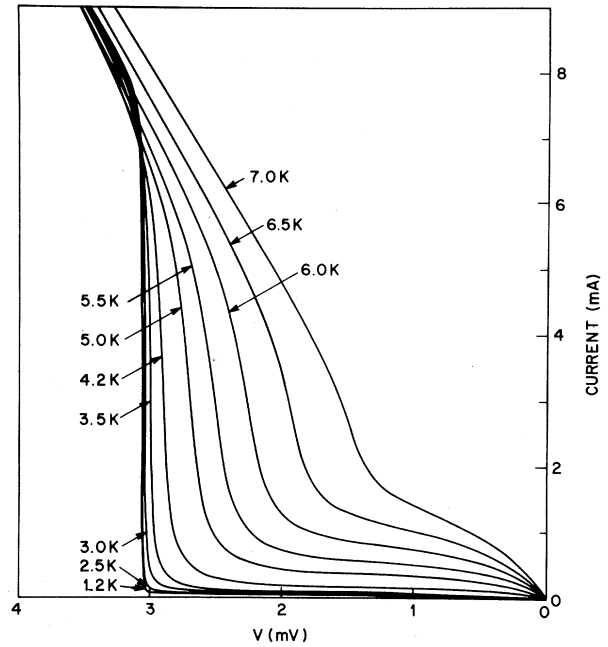


FIG. 1. I - V characteristic for a $\text{Pb}_{0.9}\text{Bi}_{0.1}$ tunnel junction for various temperatures. A small magnetic field (\sim a few gauss) has been applied parallel to the films to suppress the dc Josephson current. Note the increase in the gap smearing with increasing temperature.

apparent. Finally, the result germane to this Letter, the broadening of the gap edge, is also very clear as the temperature is raised. At temperatures below 3.0 K, the current rise at 2Δ is rather sharp while with increasing temperature it clearly broadens. The I - V characteristic of a superconductor-insulator-superconductor tunnel junction can be written

$$I = C_N \int_{-\infty}^{\infty} \rho(E) \rho(E+V) [f(E) - f(E+V)] dE, \quad (2)$$

where within BCS theory the excitation spectrum $\rho(E)$ is given by

$$\rho(E) = \text{Re} [|E| / (E^2 - \Delta^2)^{1/2}]$$

and C_N is the normal-state conductance of the junction. At all temperatures below T_c this expression results in a finite discontinuity at 2Δ in the I - V characteristic due to the discontinuity in $\rho(E)$. Figure 1 demonstrates clearly that this discontinuity is not observed and, in fact, there is a temperature dependence to the slope of the rise at 2Δ . For the reasons outlined earlier we interpret this broadening as due to finite-lifetime effects of the quasiparticles at the gap edge, and we analyze the data of Fig. 1 in more detail by introducing lifetime effects into the density of ex-

citations. We do this by adding an imaginary part to the energy, writing

$$\rho(E, \Gamma) = (E - i\Gamma) / [(E - i\Gamma)^2 - \Delta^2]^{1/2},$$

and substituting $\rho(E, \Gamma)$ for $\rho(E)$ in (2). The real part then gives us the measured I - V characteristic with the broadening due to the Γ term. Because we are interested in a small energy region of the gap edge Δ , we feel justified in choosing the simplest possible description, that of an energy-independent imaginary part. Also, since we are interested in behavior very close to the gap Δ , we assume Δ to be a real constant. For energies away from the gap edge, the energy dependence of the self-energy³ must be included.

In order to analyze the data in more detail we have differentiated the I - V characteristics of Fig. 1 and adjusted Γ for the various temperatures until a best fit to dI/dV vs V is achieved. Δ and T are measured quantities which enter (2) and so the only adjustable parameter is Γ . The

results of such line-shape fits are shown in Fig. 2 for various temperatures and it can be seen that an excellent fit can be achieved. Several junctions of the same alloy have been studied in this fashion and the results are very reproducible. It is even more clear from Fig. 2 that as a function of temperature, the linewidth broadens, and the values for Γ obtained for the fit shown are listed on Fig. 2. The quality of the fit is substantially reduced for a 5% variation of Γ from the values listed.

It is interesting to note that below 2.5 K the line does not further narrow, suggesting an intrinsic width corresponding to a Γ of 0.01 meV. This intrinsic width is also reproducible from junction to junction and has several possible sources. Remnant anisotropy effects could possibly explain the low-temperature width.⁵ Concentration variation in the evaporated films could also be the source of the width but the reproducibility of such an effect suggests that this is not

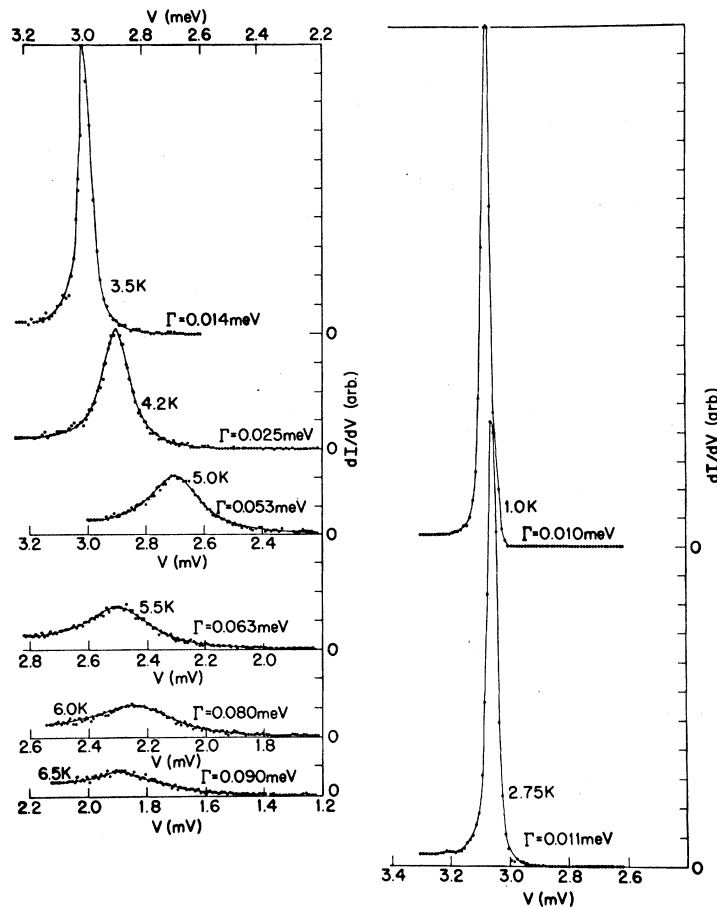


FIG. 2. dI/dV vs V determined from the data using Fig. 1. The solid curves are fits to the data using Eq. (2) with Γ an adjustable parameter.

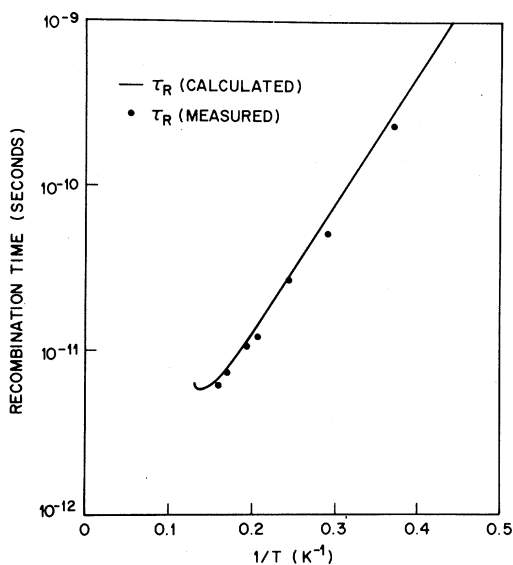


FIG. 3. Measured quasiparticle recombination time compared with the calculated value from Ref. 3.

likely. Extrinsic noise could also tend to cause such a width but again the reproducibility seems to exclude this. Another possibility for such a width is that of statistical concentration fluctuations.⁷ In this alloy, if we assume a coherence length of ~ 100 Å and a statistical fluctuation of Bi concentration in this volume, knowing the Δ /concentration from previous tunneling results⁴ we can estimate the gap variation due to these fluctuations and this turns out to be 0.004 meV, smaller but of the same order of magnitude as our measured value of 0.01 meV. At any rate, this background width is small and easily subtracted from the temperature-dependent component which we analyze in terms of τ_R .

In Fig. 3 we plot the measured τ_R vs $1/T$ and compare with the temperature dependence calculated from the work of Kaplan *et al.*¹³ for $\text{Pb}_{0.9}\text{Bi}_{0.1}$. It is emphasized that there are no adjustable parameters in this comparison as τ_0 is calculated from Eliashberg theory using the meas-

ured values of the electron-phonon coupling strength λ for this alloy.⁴ The agreement between the measured value and that calculated is seen to be excellent and is good evidence that the gap smearing observed is indeed due to lifetime broadening as a result of the recombination process. From Fig. 3 the rapid variation of τ with temperature [Eq. (1)] is also evident. As outlined earlier, the alloy system PbBi was chosen because of the strong-coupling nature of the electron-phonon interaction. For a more weakly coupled superconductor such effects are not so easily observable. An extreme example of this is the case of Al where we have measured the gap broadening and found no temperature dependence between 100 mK and T_c (1.2 K). This is very simply due to the fact that the recombination time in Al is expected to be 10^4 longer than that in PbBi as a result of the weak coupling.

In summary, we have directly measured the quasiparticle recombination time in $\text{Pb}_{0.9}\text{Bi}_{0.1}$ by measuring the lifetime-broadening effects at the gap edge via superconducting tunneling. Excellent agreement between our measured values and those calculated is achieved.

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¹See D. N. Langenberg, in *Proceedings of the Fourteenth International Conference on Low Temperature Physics, Otaveimi, Finland, 1975*, edited by M. Krusius and M. Vuorio (North-Holland, Amsterdam, 1975).

²P. Hu, R. C. Dynes, V. Narayanamurti, H. Smith, and W. F. Brinkman, *Phys. Rev. Lett.* **38**, 361 (1977); V. Narayanamurti, R. C. Dynes, P. Hu, H. Smith, and W. F. Brinkman, *Phys. Rev. B* (to be published).

³S. B. Kaplan, C. C. Chi, D. N. Langenberg, J. J. Chang, J. Jafarey, and D. J. Scalapino, *Phys. Rev. B* **14**, 4854 (1976).

⁴R. C. Dynes and J. M. Rowell, *Phys. Rev. B* **11**, 1884 (1975).

⁵C. K. Campbell, R. C. Dynes, and D. G. Walmsley, *Can. J. Phys.* **44**, 2601 (1966).

⁶J. P. Garno, *J. Appl. Phys.* **48**, 4627 (1978).

⁷We thank J. Mochel for suggesting this possibility.