Experimental Observation of the Relaxation Time of the Order Parameter in Superconductors*

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We have measured directly the relaxation time τ for the superconducting energy gap. We show that near the superconducting transition temperature T_c the relaxation time diverges as predicted by several authors. By also measuring the equilibrium gap Δ we find $\tau \propto \Delta^{-1}$ very near T_c in agreement with the calculations of Schmid and Shön.

One of the most fundamental quantities of the recent activity in nonequilibrium superconductivity[1,2] is the characteristic relaxation time τ of a superconductor to external perturbations which break Cooper pairs, thereby decreasing the order parameter. We have directly measured the relaxation time of the superconducting order parameter (i.e., the energy gap Δ). At low temperatures well-defined quasiparticles recombine to form pairs in accordance with the BCS theory and previous measurements,[3,4] but very near the transition temperature T_c ($T/T_c \geq 0.99$), the relaxation time diverges, which is in agreement with calculations.[5-8] We can understand the divergence in terms of the Ginzburg-Landau theory of second-order phase transitions. The generalized force restoring the order parameter back to its equilibrium value is the derivative of the free energy with respect to the order parameter. This derivative vanishes at T_c leading to a large relaxation time. At low temperatures, although the generalized restoring force is large, the relaxation time is limited by the quasiparticle recombination time which can be large. Our experiment is also unique in that we measure at the same time the equilibrium order parameter. We find that the relaxation time is inversely proportional to the magnitude of the order parameter as predicted by Schmid and Schön.[8]

In the experiment, we study aluminum films, which are part of a tunnel junction used to measure both the equilibrium energy gap and its real time response when a short laser pulse perturbs the aluminum film. Hence we have a direct measure of both the order parameter and the relaxation time.

Aluminum (1000 Å)-tin (2500 Å) tunnel junctions were prepared on glass substrates and immersed directly in liquid helium whose temperature was stabilized automatically to better than 0.5 mK using a germanium resistor sensor and heater. In the I-V characteristic of an asymmetric junction there is structure at $\Delta_{Sn} \pm \Delta_{Al}$ as seen in the inset in Fig. 1. The aluminum film of the tunnel junction was illuminated by a pulsed GaAs injection laser of about 10-nsec risetime, which was also immersed in the liquid helium. The junction was current-biased with a battery supply and the change in its voltage due to the laser pulses was amplified and fed into a sampling scope. Since the aluminum film is opaque, the pulse of light decreases only Δ_{Al}. Therefore when the junction

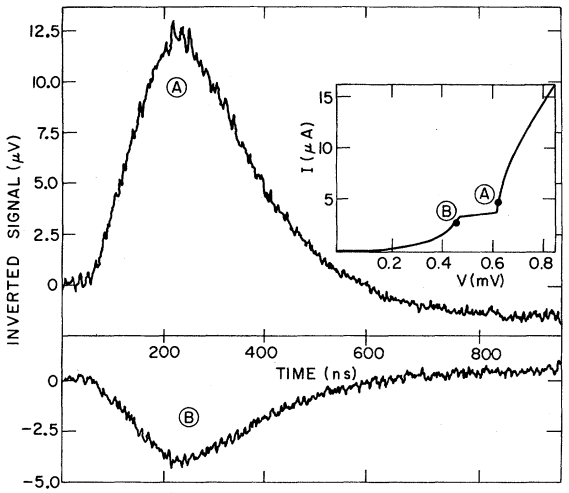

FIG. 1. The change in the junction voltage versus time at bias points A and B (shown in the inset). The laser illumination occurs during the risetime of the pulse.

is biased at point B (Fig. 1) a positive decay signal is seen and when the junction is biased at point A, the signal is negative. Note that these signals are shown inverted in Fig. 1.

In order to reduce direct electromagnetic pickup from the laser (seen as ringing at the beginning and end of the pulse) to about 15 μV peak to peak, the junction and laser were placed in separate Nb shields and connected by a fiber-optic bundle. The amplifier risetime was about 4 nsec and the equivalent noise referred to the input was about 20 μV peak to peak. The noise was adequately averaged by the sampling scope, but the pickup, which was much larger than the signal near T_c (~1 μV), had to be subtracted in the following manner. Data (including pickup) at bias point A were stored in a digital memory. Then the battery connections of the current supply to the junctions were reversed, reaching a point on the characteristic symmetrical to A but with opposite voltage. In this way all the impedances, including the junction differential resistance, were the same so that the pickup should be identical. However, the signal is reversed. When the second set of data is subtracted, the pickup subtracts and the signal adds. The pulses of Fig. 1 were obtained in this way. The signal-to-noise ratio was 5 in the worst case, with negligible evidence of direct pickup. The decay signal shows a slight undershoot caused by the low-frequency threshold of one amplifier (0.1 MHz). An amplifier with a lower threshold frequency did not show the undershoot but had a much poorer signal-to-noise ratio.

The signals were plotted on semilog paper using a baseline which was chosen to give the best exponential decay and is slightly below the undershoot. Provided that the change in Δ was small compared to Δ we found a good fit to a straight line over about three time constants with an average deviation of about 5%. The relaxation time is the slope of such a plot and is shown in Fig. 2 against the reduced temperature T/T_c. The two different samples show essential agreement and a dramatic increase close to T_c. The transition temperatures were determined from an extrapolation of Δ versus T. We have also plotted the steady-state quasiparticle-recombination measurements of one of the authors[3,4] for comparison. In plotting the steady-state data we have scaled the times vertically so as to coincide with the present results.

It is traditional[9] to fit the relaxation times by $(T_c - T)^n$ to compare with theoretical predictions of the constant n. However this is not appropriate in our case since our measured equilibrium energy gap, shown in the inset of Fig. 2, does not follow the BCS temperature dependence[10] (the straight line in the inset) close to T_c where the divergence in τ occurs. Hence we choose to plot τ versus Δ as shown in Fig. 3. Schmid and Schön[8] predicted $\tau \propto \Delta^{-1}$ whereas for a gapless

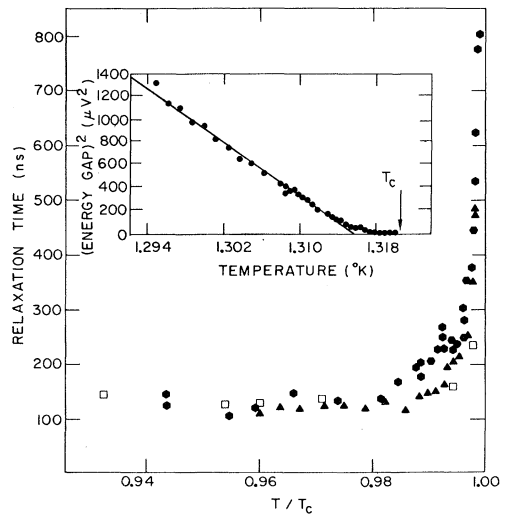

FIG. 2. Relaxation time τ versus the reduced temperature. The solid data points are the relaxation times measured in two different samples. The open squares are the quasiparticle lifetimes which are scaled as explained in the text. The inset shows the temperature dependence of the equilbrium energy gap and the solid line is the BCS prediction.

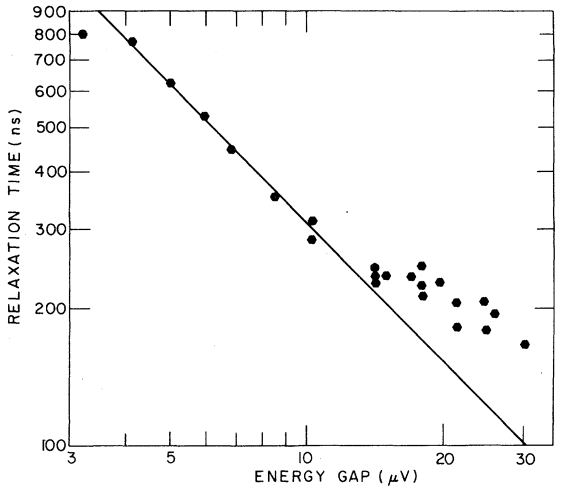

FIG. 3. Measured relaxation time for one sample versus energy gap, showing good agreement with the solid line whose slope is -1. The other sample showed essentially the same behavior.

superconductor[5,11] (fluctuations above T_c or with paramagnetic impurities) the dependence is $\tau \propto \Delta^{-2}$. In Fig. 3 we find good agreement with $\tau \propto \Delta^{-1}$ close to T_c. It should be pointed out that over the same temperature range the best fit of τ against temperature indicates that $\tau \propto (T_c - T)^{-1}$, but we feel that this is only an artifact of the non-BCS temperature dependence of Δ near T_c.

We have considered the possibility that heating or junction electrical time constant can influence the results. It is clear that RC time constant (estimated to be ≤ 8 nsec) or thermal relaxation time (τ_T) (estimated from the calculations of Little[12] to be ≤ 30 nsec) cannot explain the measured divergence, since they are weakly temperature dependent. There is however a possibility that heating is affecting the results, but in the following we give some strong arguments to show this is not so.

At the closest point to T_c ($\sim \frac{1}{2}$ mK below T_c) the signal size is less than $\frac{1}{4}\Delta$ and therefore at this point the temperature cannot change more than $\frac{1}{8}$ mK. There are two possible contributions to the signal we observe at the time the laser pulse is turned off. The first one, S_T, is due to a change in the film temperature which affects Δ and relaxes with the time constant τ_T. Another contribution, S_R, is due to direct pair breaking and decays with the order-parameter relaxation time (τ) which we wish to measure. We make the reasonable assumption that for the highest temperatures $\tau_T \ll \tau$, and recognize the fact that in none of our decays is it possible to fit the data by two exponentials. Hence at high temperatures S_T, the faster decaying component of the signal, must be negligible compared to S_R. We also point out that experimentally the total signal $S_T + S_R$ decreases as T increases. However S_T is expected to increase with T since $d\Delta/dT$ increases with T and dT/dt is approximately independent of T. Hence the ratio S_T/S_R decreases with decreasing T and S_T is negligible for all T. Any other assumption about the relative magnitude of τ_T and τ leads to the same conclusion, so we feel confident that we are measuring τ and not τ_T at all temperatures.

The first reported experiments that showed the divergence of the relaxation time near T_c were the steady-state quasiparticle-lifetime measurements.[3,4] However these measurements were not sufficiently detailed to establish this behavior (see Fig. 2). In these experiments the relaxation time was measured by injecting quasiparticles through a tunnel junction and measuring the perturbed quasiparticle population. In a different experiment, Peters and Meissner[9] measured the response of the superconducting pairs to a high-frequency field and deduced a relaxation time apparently diverging as $(T_c - T)^{-1/2}$. Schmid has treated this problem theoretically.[13] In a third experiment, Clarke[14] has derived the relaxation time of quasiparticles from electron branch to hole branch, from the measured potential difference across a tunnel junction. This branch-imbalance relaxation time showed an energy-gap dependence ($\tau \propto \Delta^{-1}$) which agrees with the calculations by Tinkham and Clarke[15] and Schmid and Schön[8] who show that this corresponds to relaxation of the phase of the order parameter.

In all the above cases, the temperature dependence of the relaxation times are shown to be, or are consistent with, $(T_c - T)^{-1/2}$, which implies Δ^{-1} for a BCS energy gap. What we emphasize is that none of these is a direct measure of the relaxation time, although Peters and Meissner's[9] measurements are based on a rf frequency response. In contrast, the present work is a real-time measurement of the order parameter, i.e., the superconducting energy gap. For our case, Schmid and Schön[8] predict that the relaxation time for the magnitude of the order parameter is $[\pi^3/7\xi(3)](\tau/\Delta)\tau_e$, where τ_e is the electron scattering time for any inelastic collisions. We feel that our results adequately confirm this gap dependence and we determine that τ_e is 7 nsec.

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Weakly Pinned Fröhlich Charge-Density-Wave Condensates: A New, Nonlinear, Current-Carrying Elementary Excitation

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New, nonlinear, charged elementary excitations are predicted to occur for weakly pinned Fröhlich charge-density-wave condensates at low temperatures.

The existence at low temperatures of a pinned Fröhlich charge-density-wave (CDW) condensate[1,2] in the interesting linear-chain conductors tetrathiafulvalene-tetracyanoquinodimethane (TTF-TCNQ) and $K_2Pt(CN)_4Br_{0.3}\cdot 3H_2O$ (KCP) now seems to be fairly well established.[3,4]

On the basis of a phenomenological theory, we have been able to investigate the *nonlinear* phase dynamics of weakly pinned CDW condensates. We wish to report here the following most interesting theoretical finding: As a specific consequence of nonlinearity and the periodicity of the potential $V_p(\varphi)$ by which the condensate is pinned, an altogether new type of mobile, current-carrying, elementary excitation of the condensate occurs. These new, nonlinear, charged excitations arise, formally, from "solitary wave" or elementary-particle-like solutions[5] of a nonlinear wave equation for the local phase $\varphi(x,t)$. We shall refer to them here as "φ particles." Physically, they correspond to propagating *localized* compressions (φ particles) or rarefactions (anti φ particles) in the local condensed electron density $n_s(x,t)$. They separate segments of the condensate having common uniform phase and may therefore be viewed as mobile domain walls. A finite threshold energy, E_0, is required for their creation and they can thus only be thermally excited at finite T. Furthermore, since the charges on the φ and anti φ particles are equal and opposite, the number of φ and anti φ particles present at any given T must necessarily be the same. In this respect the φ particles may be regarded as the inevitable Schottky defects of the perfect "ionic crystal" of which the condensate with completely uniform phase at $T=0$ is representative.

It is interesting that although the condensate is pinned, and therefore unable to contribute a collective Fröhlich conductivity,[1,2] the charged φ particles now render it conducting.

In general, local deformations of an otherwise perfectly uniform Fröhlich CDW condensate may result from local variations in either the condensate's amplitude or phase. The linear dynamics of such deformations (amplitude and phase "phonons") have been discussed in an elegant paper by Lee, Rice, and Anderson[6] (LRA). Sufficiently weak pinning, however, implies a comparatively *much softer* force constant for phase deformations, with the result that at low temperatures spontaneous local deformations in the phase will dominate those occurring in the amplitude.[7] Since the local phase variations involved need not necessarily be small, a nonlinear treatment of their dynamics is called for.

The latter may be studied on the basis of a straightforward generalization of a phenomenological theory of the linear phase dynamics given