

Superconducting Order-Parameter Fluctuations below T_c †

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Measurements of the frequency- and wave-vector-dependent pair-field susceptibility have been carried out below the transition temperature of a superconductor using a tunneling technique. The results suggest that the dynamical equation of the order parameter below T_c is something other than a diffusive time-dependent Ginzburg-Landau equation.

In this Letter we report the first measurements of the frequency- and wave-vector-dependent pair-field susceptibility^{1,2} of a superconductor below its transition temperature. Previous measurements³ have been confined to the case where the average pair field of one superconductor with transition temperature T_c' was used to determine the pair-field susceptibility of a second metal with transition temperature T_c in the temperature range $T_c < T < T_c'$. The two metals formed the electrodes of a thin-film tunnel junction, with an excess current in the dc I - V characteristic due to pair tunneling giving a direct measure of the imaginary part of the frequency- and wave-vector-dependent pair-field susceptibility of the normal electrode. The frequency and wave-vector dependence of this susceptibility were determined by measuring the voltage and magnetic-field dependence of the excess current.

When both metals are superconducting, the phases of the order parameters of the two electrodes are coupled by the tunneling interaction. The physical manifestations of this coupling are the usual ac and dc Josephson effects. A new effect is observed as the temperature is lowered below T_c in the presence of a magnetic field applied in the plane of the junction, sufficient in magnitude to uncouple the order parameters and destroy the Josephson effect, but not greater than the critical field of either metal. Under these conditions, a contribution to the excess current-voltage characteristic is observed to develop in a continuous manner from features observed above T_c which have been previously interpreted as the pair-field susceptibility.^{2,3} We interpret these low-temperature features of excess current as pair currents arising from the coupling between the order parameter in the electrode with transition temperature T_c' and the fluctuating order parameter in the electrode with transition temperature T_c . In analogy with the situation occurring above T_c , we would expect the excess tunneling current to be a measure of

the susceptibility of the low-transition-temperature electrode.

In superconductors above T_c fluctuations of the order parameter are governed by a diffusive time-dependent Ginzburg-Landau equation,^{4,5} which has been studied in previous measurements of the pair-field susceptibility.³ These order-parameter fluctuations are also responsible for the precursive behavior in the electrical conductivity,⁶ magnetic susceptibility,⁷ and single-particle density of states⁸ which have also been studied extensively in recent years. Below T_c there have been few measurements which have quantitatively probed the dynamical behavior of the order parameter.⁹ Knowledge of the dynamics is important in the analysis of problems of technological interest such as transport in type-II superconductors and the normal-superconductor phase boundary. At the present time a simple time-dependent Ginzburg-Landau equation does not exist for the regime below T_c .¹⁰ Measurements of the pair-field susceptibility below T_c should assist in the determination of a macroscopic time-dependent equation.

We have measured I - V characteristics of carefully masked Al-Al₂O₃-Pb junctions biased from a constant-current source. The aluminum films in these junctions are disordered and have transition temperatures and critical fields substantially in excess of bulk values.¹¹ Measurements were carried out at temperatures such that both the coherence length and penetration depth of aluminum were larger than the thickness of the aluminum electrode of the junction. Consequently, the order parameter and the magnetic field are constant across the thickness. Details of sample preparation, characterization, and testing have been described in detail elsewhere.³ A major departure from previous practice was the determination of the background current from the leading term of the expression for the quasiparticle current derived from the BCS theory in the case of a normal-insulator-superconductor junction.¹²

For every temperature and magnetic field, a two-parameter fit of the experimental I - V characteristic by the theory was carried out over a range of voltages for which the pair current due to fluctuations was small. The coefficients were then used to compute the quasiparticle background current at the very low voltages which were of interest. This procedure was also used to approximate the background even when both halves of the junction were superconducting. Near T_c , where the aluminum gap is small and thermal smearing is large, the difference between results of a fit based on this procedure and a more complete calculation with two full energy gaps is negligible in the region of interest. Care must be exercised in the interpretation of the excess current obtained in this manner since the elementary theory used to calculate the background does not include higher-order single-particle processes or electron-pair interference effects. Thus some features of the excess current-voltage characteristic may arise from processes not removed by the subtraction, which are not related to the fluctuating-pair current.

In the present measurements a minicomputer was used for data acquisition and on-line analysis. Temperatures were regulated to better than 1.5×10^{-5} K and were measured using a calibrated germanium thermometer. Magnetic fields were provided by a small superconducting Helmholtz coil outside the vacuum jacket.¹³ Magneto-resistive corrections to the thermometry were experimentally insignificant for the range of fields and temperatures explored.¹⁴

In Fig. 1 we show typical excess I - V characteristics at several temperatures in a field of 100 Oe. Data were taken at temperatures and fields at which there were no negative dynamical resistances in the I - V characteristic. When $T_c < T < T_c'$, which is the case in Fig. 1(a), the pair current is given by the results of Ref. 2:

$$I_1(V, H) = (4e|\bar{C}|^2 A/d\hbar)\chi''(\omega, q), \quad (1)$$

where χ'' is the imaginary part of the susceptibility χ which is given by

$$\chi^{-1}(\omega, q) = N(0)\epsilon[1 - i\omega/\Gamma_0 + \xi^2(T)q^2]. \quad (2)$$

The relaxation frequency $\Gamma_0 = 8\pi^{-1}k_B T_c \epsilon/\hbar$, the coherence length $\xi(T) = \xi(0)\epsilon^{-1/2}$, $\epsilon = (T - T_c)/T_c$, $q = (2e/\hbar)H(d/2 + \lambda')$, and $2eV = \hbar\omega$. Here d is the thickness of the normal film, λ' is the penetration depth of the superconducting electrode, q is the wave vector, and V and H are the dc bias and magnetic field, respectively. $N(0)$ is the single-

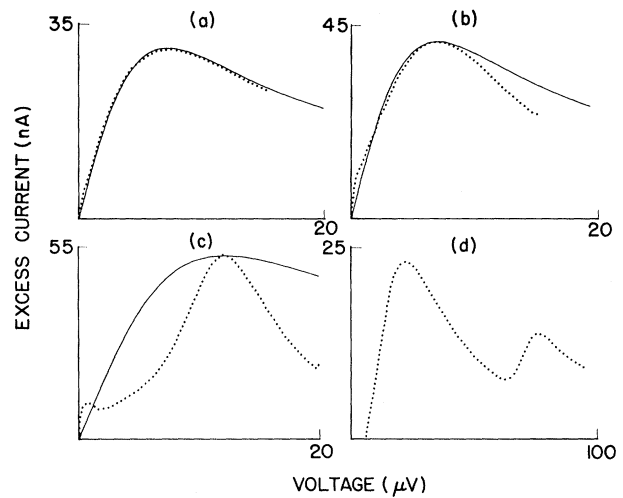


FIG. 1. Excess-current-voltage characteristics. Dots, experimental points; solid lines, calculated by fitting Eqs. (1) and (2) to the peak voltage and current: (a) $T = 1.95465$ K, $H = 100$ Oe ($T > T_c$); (b) $T = 1.93341$ K, $H = 100$ Oe; (c) $T = 1.92719$ K, $H = 100$ Oe; (d) $T = 1.89329$ K, $H = 100$ Oe.

electron density of states, A is the area of the junction, and \bar{C} is a coupling constant which depends upon tunneling parameters. Previously reported measurements are in quantitative agreement with Eqs. (1) and (2) in zero field and in qualitative agreement in the presence of a field. Corrections to the theory associated with both the depression of T_c by the magnetic field and the variation of the order parameter across the thickness of the film have been carried out by Lee and Shenoy.¹⁵ A detailed comparison of our results with their calculations is not essential to the present discussion. However, it is necessary for quantitative interpretation of data which will be presented later.

The solid lines in Figs. 1(a)–1(c) are fits of the peak current and peak voltage by Eqs. (1) and (2). It is important to note that the data in Figs. 1(b), 1(c), and 1(d) were obtained below T_c and are not in agreement with the form of the theory which holds above T_c . In some cases [Fig. 1(d)] there is even a secondary peak. At fixed temperature below T_c the effect of increasing the magnetic field is to reduce the height and the width of the main peak. At fixed magnetic field below T_c the effect of reducing the temperature is to reduce the height of the main peak and increase its width. The shapes of the experimental curves below T_c suggest that the usual diffusive Ginzburg-Landau equation be replaced by an equation which has

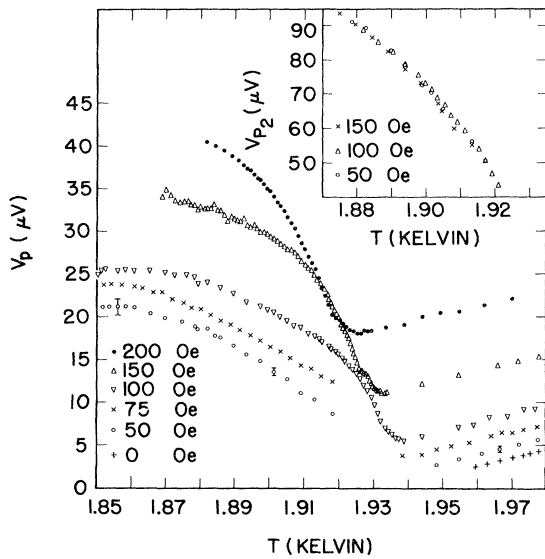


FIG. 2. Peak voltages of the main peaks of curves like those in Fig. 1 plotted against temperature in several magnetic fields. For small fields near T_c the dynamical resistance of the full I - V characteristic becomes negative, leading to switching and making measurements of the characteristic using a constant-current source difficult to interpret. Data obtained in low fields near T_c which exhibit switching are not shown. The inset is a plot of the peak voltage V_{P_2} versus temperature at several values of the magnetic field of the second peak shown in Fig. 1(c).

damped propagating solutions.

Attention should be paid to the second peak appearing in the excess-current-voltage characteristic shown in Fig. 1(d). This peak is currently not understood. Its temperature and magnetic-field dependence, which are given in the inset of Fig. 2, are not consistent with any feature of tunneling junctions of which we are aware. We also have no explanation for the shoulder appearing at low voltages in Figs. 1(b) and 1(c).

In Fig. 2 we plot the peak in the excess-current-voltage characteristic as a function of temperature in several magnetic fields. The onset of the superconducting state in aluminum is marked by a change in the sign of the dependence of the peak voltage on temperature. This is a consequence of the fact that ϵ , the quantity characterizing fluctuations near T_c , is an increasing function of temperature when $T > T_c$ and a decreasing function when $T < T_c$. Above T_c and in a magnetic field, the peak voltage is a measure of the pair relaxation frequency $\Gamma_q = \Gamma_0 [1 + q^2 \xi^2(T)]$ as it is the product of $\hbar/2e$ and Γ_q . If the susceptibility were determined from a Ginzburg-Landau equa-

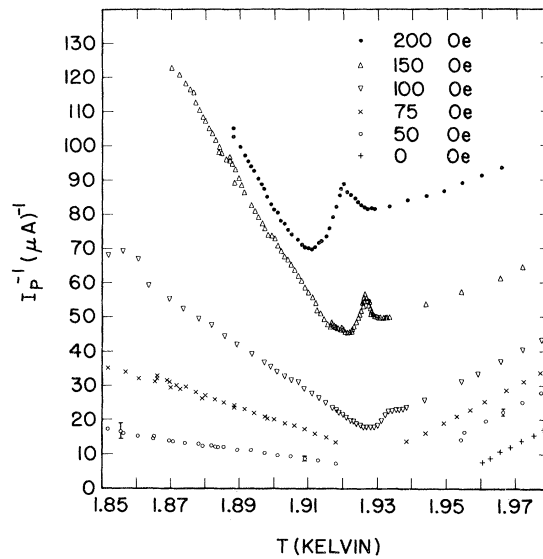


FIG. 3. Inverse peak current versus temperature in several magnetic fields.

tion with propagating solutions, then the peak voltage would be a measure of the resonant frequency of the propagating mode and the width of the resonance would be a measure of the relaxation frequency. Understanding the actual situation below T_c will require a detailed theory.

In Fig. 3 we plot the inverse of the peak current I_P^{-1} as a function of temperature in several magnetic fields. The change in sign of the slope going through the transition region is evident. As the peak current is a measure of the strength of the fluctuations, the unusual nonmonotonic behavior of I_P^{-1} in the vicinity of T_c implies a suppression of fluctuations. As the anomalous behavior occurs over a temperature range within which the inverse correlation length $\xi^{-1}(T)$ is smaller than the wave vector, these effects may be a manifestation of critical behavior.¹⁶

In summary, we have determined the pair-field susceptibility of a superconductor below T_c at finite wave vector. The results suggest that the dynamics of the order parameter are quite complicated. In the absence of a detailed theory, the measurements of the peak voltage of the main peak are hard to interpret as they can either represent the temperature dependence of the relaxation frequency or the resonant frequency of a propagating mode. They may also be a complicated combination of the two. A propagating order-parameter mode could be an excitation in the electron-pair fluid similar to the second-sound mode in superfluid helium-4. Such modes

in superconductors have long been thought to be overdamped because the electron-lattice interaction readily transfers energy and momentum from the electron-pair fluid to the lattice.¹⁷

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¹³The component of the magnetic field perpendicular to the plane of the junction was estimated to be less than 0.7% of the applied field. The contribution to the tunneling current from vortex states produced by the perpendicular component of the magnetic field was estimated, using the data of Band and Donaldson for aluminum-lead junctions, and found to be 4 orders of magnitude smaller than the observed currents [see W. T. Band and G. B. Donaldson, *Phys. Rev. Lett.* **31**, 20 (1973)].

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Dynamic Instability of Vortices in Superconductors*

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We study the nonlinear response of vortices in thin-film superconductors to strong electric fields. In the region of small magnetic fields we find unstable regions of negative differential conductivity. The voltage along the film is therefore predicted to jump from a lower flux-flow value to a higher normal-state value when the transport current is increased beyond the maximum value the flux-flow state can support.

In previous publications¹⁻⁴ we have investigated the dissipation rates and changes in structure of vortices in a superconductor when forced into motion by the application of an electric field \vec{E} . These investigations were confined to the linear response in weak electric fields. In the present paper we extend our calculations to investigate nonlinear effects.

For the present we confine our attention to films whose thickness d is much less than the penetration depth λ for magnetic fields. For thicker films the magnetic field generated by the transport current becomes important and, even

in the absence of an externally applied magnetic field, causes the superconducting structure to break up into an inhomogeneous intermediate-state structure before becoming normal. Thin films, on the other hand, remain homogeneous in this case up to the critical current, where a sudden transition to the normal state occurs with the order parameter jumping discontinuously from a finite value to zero.⁵ The main result of our present work is the determination that this discontinuous behavior persists when an external magnetic field B is applied perpendicularly to the film, creating a resistive vortex state, ex-