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Experimental Determination of the Order Parameter  
Response Function of a Superconductor<sup>†</sup>

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## ABSTRACT

The order parameter response function of a superconductor can be obtained by incorporating it in an asymmetric tunneling junction with a higher  $T_c$  superconductor and measuring an excess current in the I-V characteristic. This current is then proportional to the imaginary part of the space and time Fourier transform of the response function with the frequency set by the bias voltage and the wave vector determined by a magnetic field applied parallel to the plane of the junction. Using this technique, studies of the dynamics of the superconducting order parameter have been undertaken above and below  $T_c$ . Above  $T_c$  the time dependent Ginzburg-Landau equation accurately describes the dynamics. Below  $T_c$  a propagating mode is found which can be associated with fluctuations in the phase of the order parameter. Measurements of the magnetic impurity dependence of the response function have been used to study the "anomalous" term in the Gor'kov-Eliashberg theory which gives the coupling between order parameter fluctuations and fluctuations in the quasiparticle distribution function. Below  $T_c$  and in a current-carrying state, amplitude and phase fluctuations are coupled and the modes soften at the critical current.

Difficulties in determining the order parameter response function of a superconductor stem from the fact that the order parameter is off-diagonal in the number representation and that there is no classical laboratory field which couples to it. The response of a superconductor to electromagnetic fields is for currents to flow. These currents are functions of the order parameter and the response function, which is the conductivity, is a complicated convolution of order parameter response functions.

In recent years it has been found possible to determine  $\chi''(\omega, q)$  using electron tunneling.<sup>1,2,3</sup>  $\chi''(\omega, q)$  is the imaginary part of the space and time Fourier transform of the response function. The superconductor under study is incorporated in an asymmetric tunneling junction with the second electrode being a superconductor with a higher transition temperature.  $\chi''(\omega, q)$  is then proportional to an excess current in the I-V characteristic of the junction with the frequency  $\omega$  set by the bias voltage and the wave vector  $q$  determined by a magnetic field applied parallel to the plane of the junction. The geometry is shown in Fig. 1. Here  $\delta$  is the oxide barrier thickness,  $\lambda_1$  and  $\lambda_2$  are the London penetration depths of the superconductors, labelled 1 and 2;  $T_{c_1}$  and  $T_{c_2}$  are the transition temperatures and  $d_1$  and  $d_2$  are the thicknesses. Usually  $T_{c_1} \approx T \ll T_{c_2}$ ;  $d_1, d_2 \ll \xi(T)$ ;  $\lambda_2 < d_2$  and  $\lambda_1 \gg d_1$ . The physical origin of the connection between the excess current and  $\chi''(\omega, q)$  can be seen by modeling the pair tunneling between the two electrodes using the effective Hamiltonian

$$(1) \quad \mathcal{H}_I = \frac{\bar{c}}{d_1} e^{-i\omega t} \int d^2r e^{iqr} \hat{\Delta}(r) + h.c.$$

where  $\omega = 2eV/\hbar$  and  $q = (2e/\hbar c) H[\lambda_1 + d_2/2]$ . The quantity  $\hat{\Delta}$  is an operator for the order parameter of 1 and is given by  $\hat{\Delta} = g \Psi_\uparrow \Psi_\downarrow$ , where  $g$  is the coupling constant and  $\Psi_\uparrow$  and  $\Psi_\downarrow$  are field operators for the destruction of spin-up and spin-down electrons. The constant  $\bar{C} = \frac{\hbar^2}{e^2} (R_N A)^{-1} \ln \frac{4 T_{c2}}{T_{c1}}$

where  $R_N$  is the normal tunneling resistance and  $A$  is the area of the plane of the junction. This effective Hamiltonian implies a pair transfer current of the form

$$(2) \quad \langle I_1 \rangle = \frac{4e\bar{C}}{\hbar d} \text{Im} \{ e^{-i\omega t} \langle \hat{\Delta}(-q, t) \rangle \}.$$

If  $T > T_{c1}$ , then  $\langle \hat{\Delta}(-q, t) \rangle_0 = 0$  where the average  $\langle \rangle_0$  is with respect to the unperturbed states of metal 1. However, even under these circumstances there is an induced gap in 1 which is the linear response to  $\mathcal{H}_1$ .

$$(3) \quad \langle \hat{\Delta}(-q, t) \rangle = \frac{1}{i\hbar} \int_{-\infty}^t dt' \langle [\hat{\Delta}(-q, t), \mathcal{H}_1(t')] \rangle.$$

Evaluating the commutator, one finds

$$(4) \quad \langle \hat{\Delta}(-q, t) \rangle = C \int_A d^2 r' e^{-iqr'} \int_{-\infty}^{\infty} dt' e^{i\omega t'} \chi(r-r'; t-t'),$$

where the response function is given by

$$(5) \quad \chi(r-r'; t-t') = \frac{i}{\hbar} \Theta(t-t') \langle [\hat{\Delta}^+(r', t'), \Delta(r, t)] \rangle.$$

Combining these results with equation (2) one finds for the excess current due to pair tunneling the result:

$$(6) \quad \langle I_1 \rangle = \frac{4e |\bar{C}|^2 A}{\hbar d} \chi''(\omega, q)$$

For  $T > T_{c_1}$  this is the only contribution from the tunneling of electron pairs. When  $T < T_{c_1}$ , the quantity  $\langle \hat{\Delta} \rangle_0 \neq 0$  and there is in addition the usual Josephson current.

Experiments have been carried out on Pb-Al<sub>2</sub>O<sub>3</sub>-Al junctions and the order parameter susceptibility of Al above  $T_{c_1}$  has been found to be in excellent agreement with that calculated from the time-dependent Ginzburg-Landau equation.<sup>3</sup> Extension of this procedure below  $T_{c_1}$  is complicated by the ac and dc Josephson effects which are linear in the coupling constant of Eq. (1) and are thus much bigger than the current resulting from the induced gap which is proportional to  $|C|^2$ . The magnetic field dependence of the Josephson current can be used to get around this difficulty. One simply applies a field in the plane of a junction large enough to destroy the Josephson current, but below the critical fields of the electrodes. Alternatively, the field may be set at a value which biases the junction at one of the minima of the "diffraction pattern" plot of critical Josephson current vs. applied magnetic field. Then cooling from  $T > T_{c_1}$  to  $T < T_{c_1}$  produces a series of curves of the type shown in Fig. 2. Below  $T_{c_1}$  a shoulder appears and there is a shape change. Actually a second peak at a voltage given by  $eV = \Delta_1(T)$  also appears at  $T_{c_1}$  and moves rapidly out of the voltage field displayed in the figure.

An understanding of the data obtained below  $T_{c_1}$  results if one employs a form of the fluctuation-dissipation theorem which relates  $\chi''(\omega, q)$  to the Fourier transform of the order-parameter correlation function  $S(\omega, q)$ .<sup>4</sup>

In the relevant limit,  $\beta \hbar \omega \ll 1$  and

$$(7) \quad S(\omega, q) = \frac{k_B T}{\pi} \frac{\chi''(\omega, q)}{\omega}$$

Here  $\beta = 1/k_B T$ . Since  $\chi''$  is proportional to the excess current and  $\omega$  is

proportional to the voltage,  $S(\omega, q)$  is proportional to the excess current divided by the dc voltage at which it is measured. In Fig. (4)  $S(\omega, q)$  vs.  $\omega$  is plotted at several temperatures for an aluminum film. A peak at the origin of  $S(\omega, q)$  vs.  $\omega$  results when the modes are diffusive. A peak at finite frequency is the signature of a propagating mode.

The propagating mode in this instance appears to be described by the theory of Schmid and Schön<sup>5</sup> in which the dispersion relation for the mode is linear (see the dashed lines of Fig. (3)). In this theory fluctuations of both the real and imaginary parts of the order parameter are considered:

$$(8) \quad \Delta(rt) = \Delta_0 + \delta\Delta(rt) + i \Delta_0 \delta\varphi(rt)$$

Here  $\Delta_0$  is the equilibrium order parameter,  $\delta\Delta$  is the fluctuation in the amplitude of the order parameter, and  $\delta\varphi$  is the fluctuation in the phase. The real and imaginary parts of the order parameter fluctuation are the "longitudinal" and "transverse" modes, respectively. The "transverse" mode is propagating when the inequality

$$(9) \quad \frac{D}{\lambda^2} \ll \omega \ll \Delta_0 \ll T_c \ll \tau_{\text{imp}}^{-1}$$

is satisfied. Here  $D$  is the diffusion coefficient,  $\lambda$  is the spatial scale of the disturbance,  $\omega$  is its frequency, and  $\tau_{\text{imp}}$  is the impurity scattering lifetime. Equations for the behavior with time of  $\Delta(rt)$  involve its coupling to  $\delta f$ , the fluctuation in the quasiparticle distribution function. This is the so-called "anomalous" term of the Gor'kov-Eliashberg equations.<sup>6</sup> In the linearized theory the real and imaginary parts of the fluctuations of  $\Delta(rt)$  are decoupled. The "longitudinal" mode is always diffusive and is not coupled to charge and current densities. The "transverse" mode involves a variation in the number of quasiparticles and can be excited by quasiparticle

injection. The charge density associated with the propagating transverse mode is small and as  $\vec{k} \cdot \vec{j}_N = -\vec{k} \cdot \vec{j}_S$  the motion of supercurrent is counteracted by the normal current. Damping of the mode is associated with  $j_N$ , this fact explaining why the mode only propagates when  $T \lesssim T_{c_1}$  (near the phase boundary).<sup>5</sup> Propagation occurs only when there is a gap in the single-particle excitation spectrum. Thus in the gapless regime of a superconductor doped with magnetic impurities the mode does not propagate.<sup>7</sup> This effect has been observed in our work on Al-Er films.<sup>8</sup>

A systematic investigation of the magnetic impurity dependence of  $\chi''(\omega, q)$  for  $T < T_{c_1}$  is currently in process. In Fig. 4 we plot the relaxation frequency of the "longitudinal" mode corrected for the effect of finite  $q$  for two different depairing parameters;  $\rho = 0.012$  for which the superconductor has a gap over the temperature range displayed and  $\rho = 0.064$  for which a gapless regime extends 20 mK below  $T_c$ . In the gapless regime the "anomalous" term is switched off by the pair-breaking and the temperature dependence of the relaxation frequency changes from  $(T_c - T)^{1/2}$  to  $(T_c - T)$  as is observed. A more detailed account of this work will be published elsewhere.<sup>8</sup>

In a current-carrying state the "longitudinal" and "transverse" modes are coupled and as the transport current approaches the critical current the modes are predicted to soften.<sup>9</sup> Such a result has been observed and will also be described in detail elsewhere.<sup>8</sup> The softened modes are significant in that they are believed to be the nucleation modes for phase-slip centers in weak links.

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Figure Captions

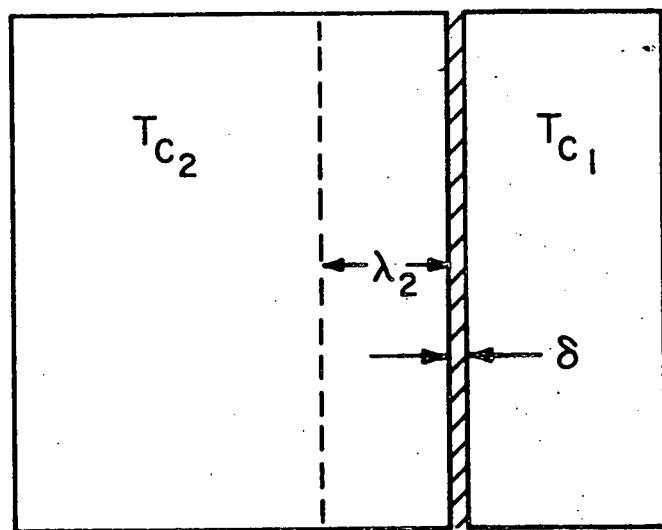
Fig. 1. Schematic of an asymmetric junction.  $d_1$  and  $d_2$  are typically  $10^{-5}$  cm and  $\lambda_2 \sim 5 \times 10^{-6}$  cm. Superconductor 2 is always Pb and superconductor 1 is Al or an alloy of Al and Er.

Fig. 2.  $\chi''(\omega, q)$  vs.  $\omega$  on cooling from  $T > T_{c1}$  to  $T < T_{c1}$  for an Al film.

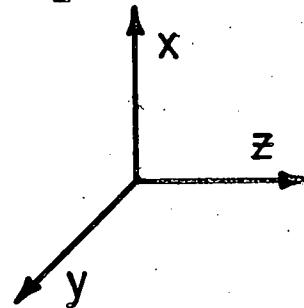
Fig. 3. Structure factors at several temperatures in an Al film. This illustrates the appearance and disappearance of the propagating mode as a function of temperature.

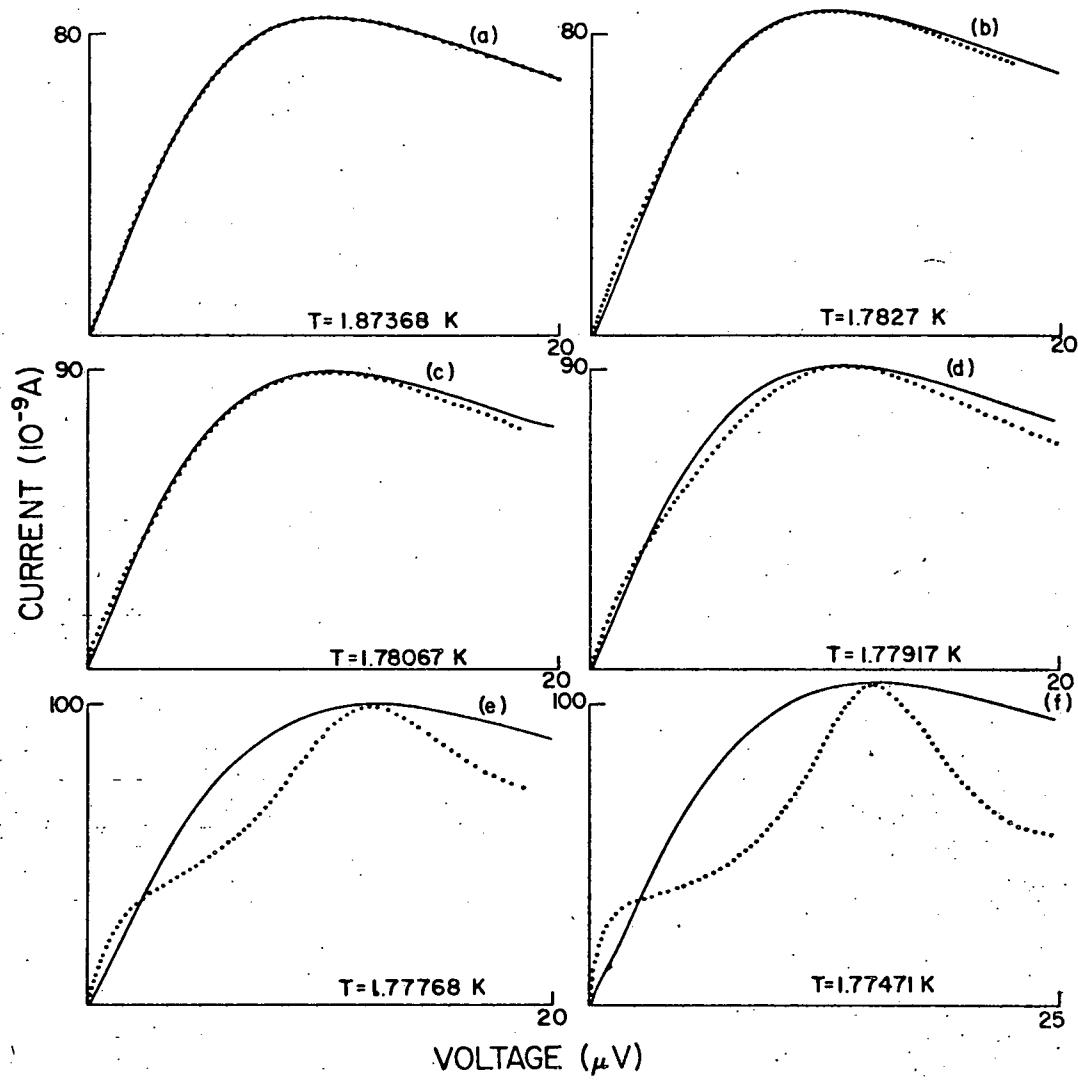
Fig. 4. Longitudinal mode relaxation frequency for  $\rho = 0.012$  and  $0.064$ , corresponding to two different concentrations of the paramagnetic impurity Er.

SUPERCONDUCTOR 2    SUPERCONDUCTOR 1



$d_2$                                      $d_1$





$S(\omega, q)$  (arbitrary units)  $\uparrow$

T

$7.59 \times 10^{10}$

$\omega \rightarrow$

1.75317

1.76022

1.74626

1.77471

1.78217

1.78977

1.83005

