Pair-field susceptibility of proximity-effect sandwiches

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Measurements of the pair-field susceptibility of Pb-Ag proximity sandwiches in the normal state are quantitatively consistent with a generalized time-dependent Ginzburg-Landau model in which the finite penetration of the barrier between the normal and superconducting films reduces the relaxation frequency of the superconducting fluctuations in a manner similar to the reduction caused by magnetic impurities.

The pair-field susceptibility of a superconductor $\chi(\omega,q)$ is the space and time Fourier transform of the response function of the order parameter or pair field.[1,2] Experimentally, the imaginary part of the susceptibility $\chi''(\omega,q)$ has been determined in tunneling experiments where it is proportional to an excess current in the I-V characteristic of asymmetric junctions in which the superconductor under study is incorporated as one electrode and the other electrode is a superconductor with a higher transition temperature. The frequency ω and wave vector q are proportional to the dc bias across the junction and to the magnetic field applied in the plane, respectively. Measurements of the pair-field susceptibility have been used to probe fluctuations of the order parameter Δ in the normal state and to investigate its dynamical behavior in the superconducting state.[3–5]

In this comment we report the first measurements of the pair-field susceptibility of Cooper-limit proximity sandwiches.[6] The Cooper limit results when the films of the proximity sandwich are thinner than their respective coherence lengths. We have verified the functional form of the time-dependent Ginzburg-Landau model for such structures and determined the magnitude of the relaxation frequency of superconducting fluctuations. The latter was found to be in quantitative agreement with a theory which provides such a generalized Ginzburg-Landau model[7] and which is based on the tunneling model[8] of the proximity effect. In contrast with other studies of the proximity effect,[9] the relevant parameter of the theory is not a free parameter as it is determined from measurements of the transition temperature of the sandwiches.

The present measurements were restricted to the regime above T_c because of difficulties in unequivocally resolving the features of the excess current below T_c. There were basically two reasons for this. First, because the pair potential of a proximity sandwich is in effect an average of the pair potential of the superconductor over the combined thickness of the superconducting and normal films, the amplitude of the excess current is reduced from the value which would result from tunneling measurements of an isolated superconducting film of the same transition temperature.[6] Second, in contrast with previous work on Al films[3–5] which had elevated critical magnetic fields ($\sim$ tens of kilogauss) Pb-Ag proximity sandwiches exhibited critical fields of the order of 100 G or less over the relevant temperature range below T_c. A critical field of such a small size does not permit application of large enough magnetic fields over the temperature range of interest to quench various features of the coherent Josephson effects which contribute to and confuse the interpretation of excess currents below T_c.

Junctions, which were all 0.3×0.3 mm^2 in area, were fabricated by depositing a layer of Ag onto a glazed alumina substrate in an ultrahigh-vacuum system at a background pressure of 10^{-9} to 10^{-8} Torr. Then, without breaking the vacuum, a Pb film was immediately deposited over the Ag film to form a proximity sandwich. The method of Garno[10] involving a controlled humidity chamber was used to oxidize the Pb film. The oxidized proximity sandwich was then returned to the vacuum system whereupon its edges were masked and a Pb counterelectrode was deposited. The yield of useful junctions formed using this procedure was small.

A total of 16 Pb-Ag proximity sandwiches with Ag film thicknesses of 1250 Å were fabricated with transition temperatures ranging from 1.1 to 1.80 K. The variation of T_c of these sandwiches with Pb thickness was found to be a linear function of the thickness of the Pb film, a result consistent with the films being in the Cooper limit.[11] An additional argument supporting the idea that these sandwiches are in the Cooper limit will be given later. With the standard choices for Θ_D, λ, and μ^* of Pb and Ag, the T_c equation of Silvert for proximity sandwiches with a strong coupling superconducting film appeared to overestimate T_c.[11]

Details of the cryogenic apparatus and the analysis used to extract the excess current due to pair tunneling from the total measured tunneling current have been given elsewhere.[4,5] The excess current-voltage characteristics above T_c were found to be quasi-Lorentzian in shape, consistent with a standard diffusive time-dependent Ginzburg-Landau model. In this instance the voltage V_p at the peak of the excess current is a measure of the order parameter relaxation frequency Γ through the relation $V_p = (\hbar/2e)\Gamma$. A typical excess-current voltage characteristic exhibiting the quasi-Lorentzian character is shown in Fig. 1. Although the measured excess current at the peak is only the order of 0.7 nA, a current of such magnitude is relatively large in comparison with the single-particle tunneling current. For this particular sample the latter was very small, increasing with voltage at the rate of 0.1 nA/μV. It was thus less than 1 nA at voltages less than 10 μV. There were consequently no difficulties in making the subtraction of the single-particle current from the total measured current, which is required in order to obtain the excess current. This analysis could not be carried out as accurately on the sample with the higher leakage current. This is reflected in larger scatter in the data for sample 1 in Fig. 2.

Entin-Wohlman has developed a theory for the excess current in the case of a Cooper-limit proximity sandwich composed of superconducting and magnetic films in intimate electrical contact.[7] The finite penetration probability for electrons traversing the potential barrier between the superconducting and normal films acts to reduce the relaxation frequency due to the fluctuations. In this instance the proximity effect reduces Γ of the sandwich in a manner similar to that of any other pair-breaking parameter ρ such as that caused by magnetic impurities. By setting to zero the pair-breaking parameter associated with spin-flip scattering, the theory yields a prediction relevant to the geometry of the present work.

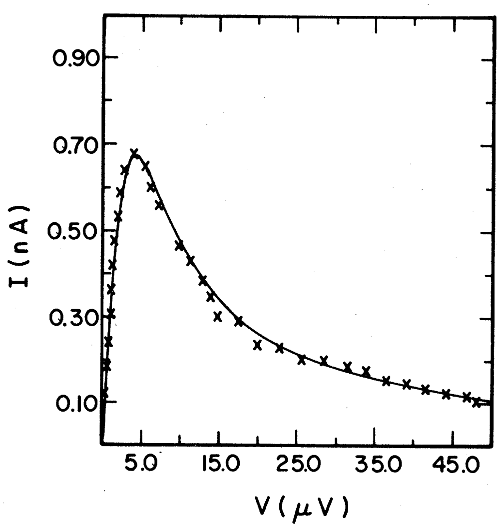

FIG. 1. Typical excess current-voltage characteristics of a Ag/Pb-PbO-Pb junction (sample 2) with the "quasi-Lorentzian" superimposed. This particular curve was obtained at $T = 1.31$ K.

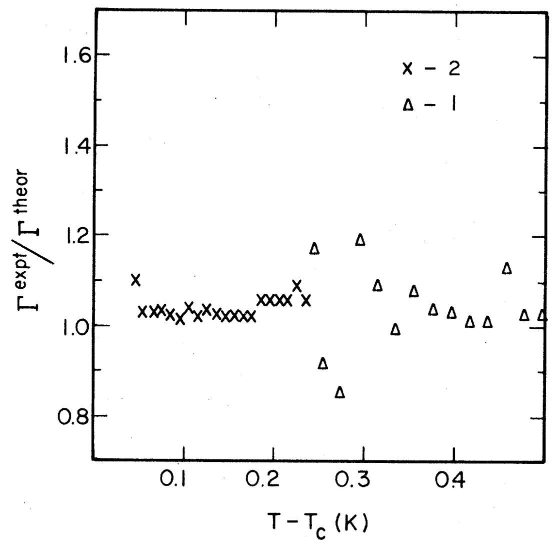

FIG. 2. Plots of $\Gamma^{\rm expt}/\Gamma^{\rm theor}$ for samples 2 and 1. $\Gamma^{\rm theor}$ is calculated from Eq. (3) using $\rho_c^{\rm tu}$ determined by Eq. (1) from the measured transition temperature. $\Gamma^{\rm expt}$ is obtained from the voltages V_p at the peak currents of curves such as that of Fig. 1.

This theory can be completely parametrized using the results of the pioneering work of McMillan[8] in which it is shown that the reduction of the transition temperature T_c of a proximity sandwich from the transition temperature T_{c0} of a solid superconducting film is given by

$$\ln\frac{T_{c0}}{T_c} = \frac{1}{R+1}[\psi(-\tfrac{1}{2}+\rho_c^{\rm tu}) - \psi(-\tfrac{1}{2})] \;, \qquad (1)$$

where $R = d_N N_N / d_S N_S$. Here d_N and d_S are thicknesses of the normal and superconducting films and N_N and N_S are the densities of states at the Fermi level in the N, S sides, respectively. Also, $\psi(x)$ is a digamma function.

The quantity of $\rho_c^{\rm tu}$ is given by

$$\rho_c^{\rm tu} = \hbar(1+R)/2\pi k_B T_c \tau_N \;, \qquad (2)$$

where $\hbar/\tau_N = \pi T^2 A d_S N_S$, A is the area of the films and T^2 is the matrix element squared of the tunneling Hamiltonian between N and S. In McMillan's notation, $\tau_N = L_N / v_F \sigma$, where L_N is an average path length for collisions with the barrier in N, σ is barrier transmission probability, and v_F is the Fermi velocity.

The order-parameter relaxation frequency is then given in Ref. 7 by

$$\Gamma = \Gamma_0 \frac{1 + Rf(\rho_c^{\rm tu})\psi'(\rho_c^{\rm tu}+\tfrac{1}{2})/\psi'(\tfrac{1}{2})}{1 + R\,\psi'(\tfrac{1}{2}+\rho_c^{\rm tu})/\psi'(\tfrac{1}{2})} \;, \qquad (3)$$

where

$$f(\rho_c^{tu}) = \frac{1-\rho_c^{tu}\psi'(\frac{1}{2}+\rho_c^{tu})}{\psi'(\rho_c^{tu}+\frac{1}{2})/\psi'(\frac{1}{2})} \quad . \tag{4}$$

Here ψ' is the trigamma function and Γ_0 is the bare relaxation frequency of the order parameter. Near T_c, Γ_0 is given by

$$\Gamma_0 = \frac{8k_B T_c}{\pi\hbar}\left(\frac{T-T_c}{T_c}\right) \quad . \tag{5}$$

From measurements of T_c and the value of R, Eq. (1) can be used to determine ρ_c^{tu}. Then R and ρ_c^{tu} can be used in Eq. (3) to calculate Γ. The important feature of this analysis is that except for R, one is not required to know in detail any of the parameters of Eq. (2).

In Fig. 2 we plot the ratio of values Γ determined from the peak voltages[5] of curves like those in Fig. 1, to the theoretical value computed from Eq. (3). In Table I the various parameters of the two junctions studied in detail are enumerated. Free-electron parameters given by Kittel were used in Eqs. (1) and (3).[12] In particular, it should be noted that m^*_{Pb} was taken to be 1.97.

The agreement of Γ determined from the peak of excess current voltage curves with that calculated from Eq. (3) using ρ_c^{tu} obtained from Eq. (1) must be viewed with some care as both equations are based on the assumption that λ_N, the electron-phonon coupling constant in the Ag film is zero. Since the calculations of Ref. 7 for $\chi''(\omega,0)$ are carried out under such an assumption it would seem inappropriate to use any of the more general results for T_c of proximity sandwiches[9,11,13] to determine the parameters of the system. The fact that the agreement between experiment and theory is good actually implies that the corrections for finite λ_N are contained in ρ_c^{tu} which is a common parameter of both the T_c equation and $\chi''(\omega,0)$. Since the analysis does not require the determination of ρ_c^{tu} from microscopic parameters, the shortcomings of the model never become apparent and may in some sense be concealed.

The same may be true of modifications due to strong coupling effects in Pb. Some years ago Fulde and Maki showed that the time-dependent Ginzburg-Landau equation in the strong coupling case was of the same form as the weak coupling superconductors, but with altered coefficients which depend on electron-phonon spectral quantities.[14] A similar result has been found for $\chi''(\omega,0)$.[15] The fact that Eqs. (1) and (3), both based on weak coupling theory, can be used to consistently describe the data implies that strong coupling corrections are small or nonexistent, or perhaps are hidden in much the manner that corrections for finite λ_N mav be concealed. Further experimental work, i.e., direct measurements of Γ on Pb films and additional theoretical calculations are needed to resolve this issue. The existence of a strong coupling correction to Γ would have serious implications for the apparent agreement with theory of measurements of fluctuation enhanced conductivity in strong coupling superconductors such as Bi, Ga, and Pb.[16] A strong coupling correction as large as 1.2 to 1.4 would destroy the present good agreement of experiment with theory.[17]

A final point has to do with the spatial variation of the order parameter in proximity sandwiches. Entin-Wohlman has only calculated the $q=0$ value of the susceptibility which turns out to be the usual quasi-Lorentzian with renormalized coefficients. From the measurements, the q dependence of $\chi''(\omega,q)$ for a proximity sandwich is seen to have the same functional form as in the single film case.[2,3] In this instance the relaxation frequency at finite q is given by $\Gamma_q=\Gamma_0[1+q^2\xi^2(T)]$, where $q=(2e/\hbar)(\frac{1}{2}d+\lambda)H$. Here d is the film thickness, λ is the penetration depth of the Pb counterelectrode, and H is the value of the magnetic field applied in the plane of the junction.[2] From measurements of the q dependence of $\chi''(\omega,q)$ we find $\xi(T)=3760\ \text{Å}/\epsilon^{1/2}$ under the assumption that d is the thickness of the Pb-Ag composite. Thus $\xi(T)$ of the composite, over the entire range of temperatures investigated, would appear to be larger than the thickness of either film and larger than the coherence length of Pb which is only 830 Å. This result would imply that the films are in the

TABLE I. Junction properties.

Sample number	Ag thickness (Å)	Pb thickness (Å)	R_N (Ω)	$R_N G_L$ [a]	T_c (K)	ρ_c^{tu} [b]	R	dV_p/dT (μV/mK)	Γ^{theor}/Γ_0 [c]
1	1250± 20	240± 20	0.24	0.053	1.31	0.306	1.441	0.10± 0.02	0.865
2	1250± 20	240± 20	3.8	0.0003	1.27	0.308	1.429	0.096± 0.003	0.865

[a] R_N is the normal tunneling resistance and G_L is the leakage conductance.
[b] ρ_c^{tu} is determined using Eq. (1).
[c] Γ^{theor}/Γ_0 is given by Eq. (3).

Cooper limit.

In making proximity sandwiches consisting of 240-Å-thick Pb layers deposited on top of 1250-Å-thick Ag layers there may be questions as to whether the Pb films are continuous and whether there are substantial thickness variations in the form of balls or crystallites. Such variations could be of the order of 100 Å and would perhaps lead to local variations of T_c and of the order parameter. In the absence of detailed electron microscopy of the surfaces one cannot answer these questions in a definitive manner. Nevertheless some strong statements can be made based on an interpretation of the tunneling data itself.

The small yield of useful junctions indicates that the above considerations are generally very important. However, for the two junctions we have reported here the questions may be irrelevant. First, we were not able to prepare unshorted Ag-Ag_xO_y-Pb junctions using the techniques of Ref. 10. Thus any unshorted, low-leakage junctions must have had base electrodes in which the Pb layer completely covered the underlying Ag layer. Second, the low-temperature I-V characteristics of the junctions exhibited sharp gap structures. In addition, the zero-voltage Josephson tunneling current in the better of the two samples was documented in detail. It exhibited a nearly ideal Fraunhoffer pattern in its dependence on an applied magnetic field. Such a result is consistent with the existence of a high degree of homogeneity of the barrier thickness and a spatially uniform penetration depth. Third, measurements of the temperature dependences of the zero-voltage dc Josephson current below T_c and χ'' above T_c are consistent with the relevant theories, each parametrized by the same transition temperature.[2,18] This was true over a range of temperature starting about 0.3 K below T_c to 0.5 K above. Finally, because the apparent zero-temperature coherence length of the Ag-Pb sandwich is 3760 Å, one would expect substantial averaging of the order parameter and of the order parameter fluctuations below and above T_c, respectively, even if there were substantial variations in the thickness of the Pb film.

It is useful to remark on the small size of the excess current due to pair tunneling above T_c. Usually this current is observed to be one-to-two orders of magnitude smaller[3,4] than the predictions of Ref. 2. If the coupling of the pair field of the high-T_c electrode to the order parameter of the proximity sandwich is the same as the coupling in the case of a single film, than an additional reduction in the magnitude of the excess current would be expected. The reason for this is that $\chi''(\omega)$ of a proximity sandwich is scaled down from its value in an isolated film by the factor

$$\frac{d_S}{d_S+d_N}\,\frac{1+R}{1+R\,\psi'(\rho_c+\frac{1}{2})f(\rho_c)/\psi'(\frac{1}{2})}\ ,$$

which for these samples is 0.37.[7] The peak currents of the proximity sandwiches studied here are about 0.27 of the values obtained in the case of Al-Al_2O_3-Pb junctions of the same geometry and normal tunneling resistance.

In conclusion, we have measured the pair-field susceptibility of Cooper-limit Pb-Ag proximity sandwiches in their normal state. The frequency dependence of the susceptibility is found to be in agreement with the theory and the q dependence is similar to that of an isolated film. The relaxation frequency calculated from the peak voltage is also found to be in agreement with the theory. A full understanding of the implications of this apparent quantitative agreement for both strong coupling modifications of the parameters and a nonzero electron-phonon coupling constant in the normal metal will require further experimental and theoretical work.

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