Magnetic Field Enhanced Order Parameter Amplitude Fluctuations in Ultrathin Films near the Superconductor-Insulator Transition

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A magnetic field tuned superconductor-insulator transition (SIT) in ultrathin PbBi/Ge films has been observed. Electron tunneling measurements in a magnetic field near the SIT show the presence of a large number of quasiparticle states near the Fermi energy. A simple analysis of these data shows that the average number of Cooper pairs in a coherence volume is on the order of 1 at the SIT. Combined with transport measurements, the data imply that strong fluctuations in the amplitude of the superconducting order parameter occur at this field tuned SIT.

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A great deal of work has been performed in an effort to understand the low temperature phases and phase transitions in two-dimensional electronic systems [1–10]. It is generally accepted that disorder of any strength prevents the formation of simple metallic phases. As a result, interesting phase transitions such as the superconductor-insulator transition (SIT) can occur. The types of insulating phases that can be formed when an ultrathin film passes through a SIT have come under close scrutiny lately. Increasing the normal state sheet resistance and, thus, the amount of disorder in an ultrathin film [1] or increasing the magnetic field applied to a superconductor [2,11] are both detrimental to superconductivity and can drive a SIT. Many experiments suggest that magnetic fields create phase fluctuations that destroy long range phase coherence and, thus, cause a SIT in films with $R \simeq R_Q = h/4e^2$. The insulating phase in this case consists of localized Cooper pairs. Most notably, scaling analyses of the resistive transitions $R(T)$ of InO_x films support this latter picture [2,3]. In contrast, experiments on homogeneous thin films strongly suggest that the disorder tuned SIT is continuous and occurs near R_Q where disorder drives fluctuations in the amplitude of the superconducting order parameter that eventually drive the amplitude to zero [4–6]. The resulting insulating phase consists of localized single electrons.

In this Letter, we present transport and tunneling measurements in quench condensed ultrathin $Pb_{0.9}Bi_{0.1}$ films with normal state sheet resistances $R_N = R(8\text{ K}) \sim R_Q$ and show that strong amplitude fluctuations occur near the magnetic field tuned SIT in these systems. The tunneling measurements for each film at the critical field for the transition, H^c, where $R(T)$ is nearly temperature independent, indicate a large number of quasiparticle states are present near the Fermi energy. This result implies that H^c is close to H_{c2}, the upper critical field where all superconductivity is quenched. In zero magnetic field, the $R(T)$ have widths comparable to the mean field transition temperature in accord with expectations based on the Ginzburg criterion [11]. This behavior indicates that the transitions are dominated by fluctuations. The application of magnetic fields further increases the size of this fluctuation dominated region. We use tunneling measurements of the quasiparticle density of states to show that the characteristic energy required for amplitude fluctuations is comparable to the binding energy of a single Cooper pair in all fields. This energy is so small that thermal and quantum fluctuations in the amplitude of the superconducting order parameter dominate the magnetic field tuned SIT in these films.

The PbBi films were thermally evaporated onto a 6 Å Ge underlayer that had been previously deposited onto a fire polished glass substrate. All experiments indicate that they are homogeneous over subnanometer length scales [4,5]. The substrate was maintained at $T \approx 8$ K for these depositions. Four terminal transport and tunneling measurements were performed *in situ* with the magnetic field oriented perpendicular to the film plane and parallel to the electron tunneling direction. Mn doped Al strips prepared at room temperature served as normal counterelectrodes for the tunnel junctions as described previously [6]. The tunneling conductance $G_j(V)$ was obtained by numerical differentiation of the current-voltage characteristics of the junction.

Similar films have been employed in investigations of the disorder tuned SIT [4,5]. In that transition, below a critical thickness and, correspondingly, above a critical sheet resistance, $R_N \simeq 8\text{ k}\Omega$, all signs of superconductivity disappear, and the film conductance decreases with decreasing temperature logarithmically as appropriate for single electron weak localization and Coulomb interaction effects [12]. Tunneling experiments suggest that amplitude fluctuations dominate this transition [6]. Here we investigate the magnetic field response of superconducting films near this disorder tuned SIT.

Data were obtained on five films with $R_N > 2.5\text{ k}\Omega$ and thicknesses $t < 6$ Å in two separate experimental runs. The $R(T)$ of four of them in magnetic fields are shown in Fig. 1. The $R(T)$ in zero magnetic field are broad. To quantify this, we define the fractional width of the transitions as the temperature interval over which R drops from 90% to 10% of its normal state value

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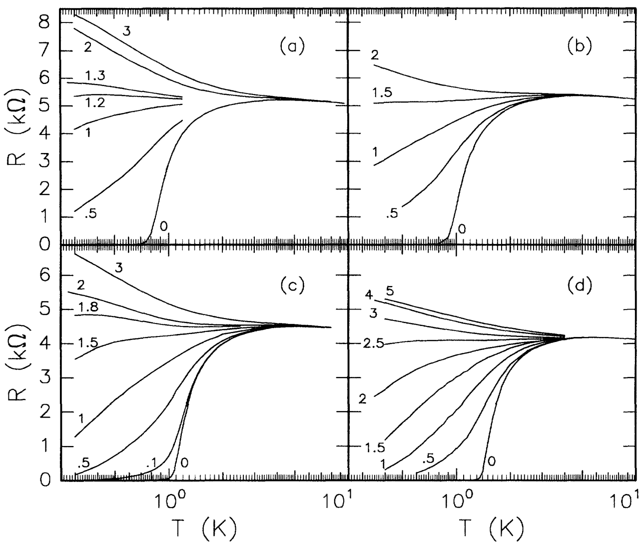

FIG. 1. Semilogarithmic plots of $R(T)$ in magnetic fields for four films. The applied magnetic field is shown in tesla. T_{c0} is (a) 0.96, (b) 1.12, (c) 1.28, and (d) 1.64 K.

divided by T_{c0}, the temperature at which the resistance has dropped to 50% of its normal state value. We use the $R(T)$ obtained at the highest magnetic fields, where most or all of the superconducting effects have been quenched, for the normal state resistance. This procedure is only approximate because the normal state has positive magnetoresistance [12]. $\Delta T/T_{c0}$ ranges from 1.6 (a) to 0.9 (d) for the data in Fig. 1.

Applying a magnetic field drives these ultrathin films through a SIT that is similar to that exhibited by a number of other homogeneous superconductors [10]. At relatively low fields the tail of the transition grows, and at higher fields the $R(T)$ become rounder near T_{c0} and the tails continue to increase in size. For all films shown there is a well-defined field H^c at which $R(T)$ becomes nearly temperature independent from above T_{c0} to below. Concentrating on the data in Fig. 1(d) at $H = H^c \simeq 2.5$ T, $R(T)$ varies by less than 6% over a range of temperature of about $1.5T_{c0}$. Above H^c, $dR(T)/dT < 0$ consistent with the behavior of an incipient insulator. It is possible that this insulating behavior could give way to superconducting behavior at low enough temperatures.

Despite the breadth of the transitions, the superconducting density of states $N_s(E)$ still maintains a BCS-like form from which we can measure an average energy gap Δ_0 [6]. We obtain N_s from *in situ* measurements of the conductance as a function of voltage $G_j(V)$ of Al/Al-oxide/PbBi film tunnel junctions. We normalize $G_j(V)$ by the conductance measured in magnetic fields high enough that $G_j(V)$ is field independent [13]. This procedure eliminates energy dependent normal state effects and tunnel barrier factors. To high accuracy the normalized conductance is given by [14]

$$G_N = \int_{-\infty}^{\infty} N_s(E) \frac{\partial f(E + eV)}{\partial V} dE, \qquad (1)$$

where E is the quasiparticle energy measured relative to E_F and f is the Fermi function. At low temperatures, $G_N(V) \propto N_s(eV)$.

We compare the $G_N(V)$ of four films at a reduced temperature $T/T_{c0} \leq 0.3$ in Fig. 2. All exhibit a peak corresponding to the singularity in the BCS density of states and a zero bias conductance well below 1. The peaks are smaller and the zero bias conductance larger, however, than expected for a BCS superconductor. The quasiparticle density of states appears to have a broadened BCS form and the broadening increases as T_{c0} decreases. Nevertheless, the resemblance of the data to a BCS form enables us to accurately estimate Δ_0, the voltage at which G_N crosses 1 [15]. Δ_0 increases with T_{c0} as expected [6].

Figure 3 exhibits the evolution of the tunneling junction conductance through the field tuned SIT of the film in Fig. 1(d). The data for the other three films of Fig. 1 are similar. Qualitatively, increases in the magnetic field decreases the size of the peaks in $G_N(V)$ and increases the zero bias conductance. It is important to note that $G_N(0) \simeq 0.8$ at H^c indicating the presence of a large number of quasiparticle states near E_F.

We begin our discussion with the data in zero field. The broad transitions are expected for high R_N films. Aslamasov and Larkin showed that Gaussian fluctuations of the amplitude of the superconducting order parameter above T_{c0} create a universal fluctuation paraconductivity [11]:

$$\Delta G = \frac{e^2}{16\hbar} \frac{T_{c0}}{T - T_{c0}}. \qquad (2)$$

Equation (2) predicts that the resistance of a 6 kΩ film drops from $0.9R_N$ to $0.8R_N$ from $2T_{c0}$ to $1.5T_{c0}$

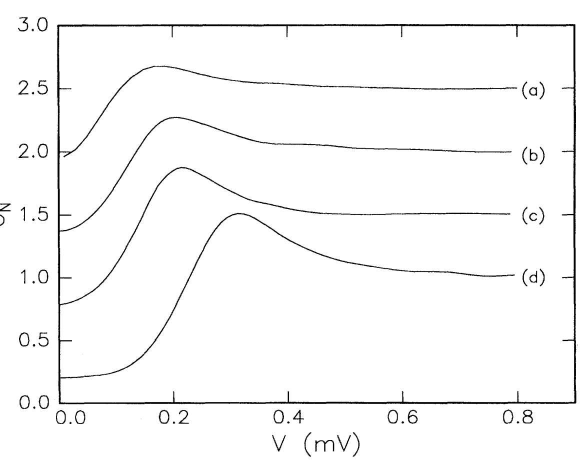

FIG. 2. Normalized junction conductances for those films shown in Fig. 1. T_{c0} and reduced temperature T/T_{c0} are (a) 0.96 K, 0.31, (b) 1.12 K, 0.31, (c) 1.28 K, 0.23, and (d) 1.64 K, 0.22, respectively. The curves have been shifted by multiples of 0.5 for clarity.

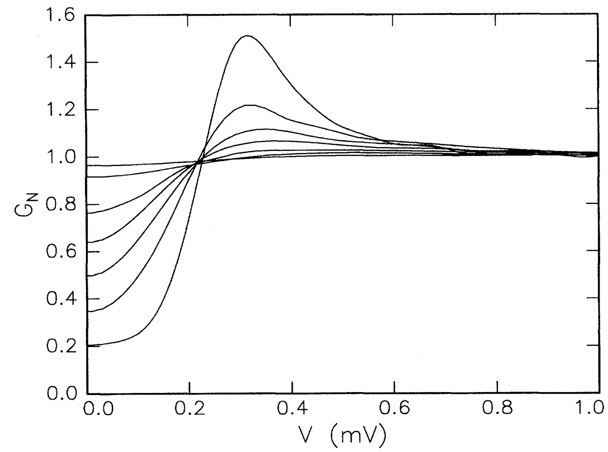

FIG. 3. Normalized junction conductance of one PbBi/Ge film with $T_{c0} = 1.64$ K at $T = 360$ mK. The magnetic fields are from bottom up at $V = 0$: 0, 0.5, 1, 1.5, 2, 3, and 4 T.

suggesting that $\Delta T \simeq T_{c0}$ in rough agreement with the data in Fig. 1(a). Furthermore, the expected size of the critical region of these transitions is large according to the Ginzburg criterion [16]:

$$\delta T = k_B T_{c0}/\Delta C \xi^2 = 0.54(R_N/R_Q)T_{c0} \qquad (3)$$

in which ΔC is the heat capacity jump at T_{c0} in two dimensions and ξ is the zero temperature coherence length. To get the second equality we assumed the standard dirty limit form for the coherence length and used bulk parameters for Pb and a Drude model (see Table I). $\delta T/T_{c0} \geq 0.4$ for the films in Fig. 1. This number is enormous compared to bulk conventional superconductors, for which $\delta T/T_{c0} \simeq 10^{-14}$ [16]. We conclude that fluctuation effects dominate these transitions.

The broadened form of the tunneling density of states suggests that fluctuations in the amplitude of the order parameter exist even well below T_{c0}. To demonstrate this possibility, we estimate the zero temperature condensation energy in a coherence volume $V_{\text{coh}} = \pi \xi^2 t$. For a BCS superconductor, this is $U_{\text{con}} = \frac{1}{2}N(0)\Delta_0^2 V_{\text{coh}}$ [11]. We use the measured Δ_0, the calculated ξ (see Table I), and multiply by a factor $1 - G_N(0)$ to take into account the extra states in the gap. $U_{\text{con}} \simeq 3\Delta_0$ for the film in Fig. 1(d) and values for the other films are shown in the table. This analysis indicates that the average number of Cooper pairs in a coherence volume is comparable to 1. This number is so small that it is likely to fluctuate from coherence volume to coherence volume due to variations in the local density of states or strong Coulomb interaction effects [13]. Since the size of the local energy gap depends on the local Cooper pair density, these fluctuations lead to a distribution of energy gaps in the film. Such a distribution appears in the conductance of a large area tunnel junction as a broadened density of states.

Fluctuation effects in the $R(T)$ become even more important in magnetic field. At temperatures well below where the order parameter amplitude forms and the resistance has dropped substantially below its normal state value (more than a factor of 10) phase fluctuations from vortex motion are generally responsible [2]. Such a picture can account for the tails of the $R(T)$ in fields well below H_{c2}. Close to and above the Cooper pair formation temperature, the influence of H on the fluctuation paraconductivity (amplitude fluctuations) leads to the rounding of the $R(T)$. Such effects have been discussed quite recently by Ullah and Dorsey [8] in relation to high T_c superconductors and superconductor-insulator multilayer systems [8,9]. Physically, a magnetic field restricts the size of V_{coh} to $t\Phi_0/H$ [8] or less and induces pair-breaking effects which reduce Δ_0 [11]. The former makes the fluctuations effectively zero dimensional instead of two dimensional and both reduce U_{con}, the pertinent energy scale for amplitude fluctuations [17]. The two effects increase the temperature range over which fluctuation paraconductivity effects affect the $R(T)$. Theory based on these considerations by Ullah and Dorsey, however, fails to fit the transitions in Fig. 1. This failure probably results from the fact that the critical region is too large even for their modified Hartree approximation.

The $R(T)$ data suggest that at H^c the fluctuation paraconductivity effects become critical, extending to an arbitrarily low temperature. The tunneling data support this notion. At H^c the number of states near E_F is approximately 80% of its normal state value at a temperature of a factor of 8 below T_{c0} implying that the order parameter amplitude is small. More quantitatively, we can estimate the number of Cooper pairs in a coherence volume at H^c. For the data in Fig. 3 $U_{\text{con}}(H^c) = 0.8\Delta_0$ which is much less than the energy necessary to break a single Cooper pair. A large fraction of the film must not contain any Cooper pairs. We presume that the regions with Cooper pairs are superconducting and those without are "normal"

TABLE I. Parameters for five films. $\xi = 0.855(\hbar v_F \ell/\Delta_0)^{1/2}$ where ℓ is the Drude mean free path.

Film	R_N (kΩ)	t (Å)	T_{c0} (K)	Δ_0 (meV)	H^c (T)	ξ (Å)	U_{con}/Δ_0
Not shown	6.35	4.16	0.50	0.045	0.5	625	?
Fig. 1(a)	5.10	4.45	0.96	0.112	1.2	427	1.6
Fig. 1(c)	4.37	4.69	1.28	0.155	1.6	383	2.4
Fig. 1(b)	5.20	5.22	1.12	0.140	1.5	350	1.8
Fig. 1(d)	4.18	5.65	1.64	0.220	2.5	299	2.9

and resistive. This scenario coincides with a simple picture of fluctuation paraconductivity and provides a natural explanation for the coexistence of superconducting correlation effects in the tunneling data and finite resistance in the transport. Further evidence supporting this picture is provided by the fact that H^c scaled linearly with Δ_0 (see Table I). H_{c2} for a dirty superconductor also depends linearly on Δ_0. Whether the configuration of normal and superconducting regions is static or dynamic cannot be determined from our data. In particular, at the lowest temperatures, the normal regions could be associated with the cores of vortices which could move and contribute dissipation. Nevertheless, it is clear that the field tuned SIT occurs very near the upper critical field of these ultrathin homogeneous films.

It is important to differentiate the present results from previous experiments on magnetic field tuned SI transitions. That work focused on the effects of fluctuations in the phase of the superconducting order parameter on the transition in, for example, relatively thick InO_x (100 Å) [3] and granular Al films [7]. The qualitative evolution in magnetic field of the $R(T)$, in the InO_x system, in particular, differs substantially from that in Fig. 1. In increasing magnetic field, the tail of $R(T)$ grows while the sharp drop in resistance that occurs near T_{c0} remains. At a critical magnetic field, the tail is flat at a resistance near R_Q that is independent of R_N. At higher fields the tail rises so that $R(T)$ exhibits quasireentrant behavior. Near the critical field, the data associated with the tail fit the predictions of a scaling theory in which only the boson degrees of freedom (i.e., Cooper pairs and vortices) are relevant [2]. According to that theory, in the superconducting phase the Cooper pairs form a superfluid and in the insulating phase the vortices form a superfluid [2]. Cooper pairs must be well formed in the region of this transition. In contrast, the temperature independent behavior in $R(T)$ exhibited in Fig. 1 extends from $T \geq T_{c0}$ where fluctuation paraconductivity effects dominate to $T \ll T_{c0}$. There is no sign of quasireentrance and no sign of a sharp drop in resistance that could be associated with the formation of Cooper pairs. Also, the resistance at which the $R(T)$ become temperature independent strongly correlates with R_N. These qualitative differences can be traced to the relative importance of amplitude and phase fluctuations to a given system.

The above notwithstanding, other systems, such as $Nd_{2-x}Ce_xCuO_4$, $YBa_2Cu_3O_{6.38}$, and $Mo_{79}Ge_{21}$ [10], show behavior that is qualitatively more similar to the ultrathin PbBi films than the InO_x data suggesting that amplitude fluctuations are important in their magnetic field tuned SI transitions. The $R(T)$ from those films and ours, however, can be scaled using exponents that are very similar to those used to scale the InO_x data where phase fluctuations dominate. This is surprising because our results suggest that the order parameter amplitude is not well formed in the region where the scaling works. Since no scaling theories exist for the amplitude fluctuation dominated case, it is not possible to determine whether these results are contradictory.

In summary, we have presented transport and tunneling data on ultrathin superconducting films near their SIT in magnetic field. The responses of resistive transitions to magnetic field in our films differ from those of InO_x near the SIT where phase fluctuations dominate. Tunneling measurements reveal the presence of a large number of quasiparticle states near the Fermi energy at the SIT. Accordingly, the SIT occurs near H_{c2} where the amplitude of the superconducting order parameter is small and likely to fluctuate.

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