PHYSICAL REVIEW B VOLUME 30, NUMBER 7 1 OCTOBER 1984

Pair-breaking model for disorder in two-dimensional superconductors

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(Received 20 April 1984)

Measurements of resistive transitions, critical fields, and magnetoconductance as a function of increasing normal-state sheet resistance R_N reveal the systematics of a pronounced change from superconducting to insulating behavior in thin In/InO_x composite films. With increasing R_N, a rapid suppression of the transition temperature together with a *decreasing* critical-field slope parameter are observed. These results are modeled using a pair-breaking formalism in which the pair-breaking rate is found to be proportional to R_N and independent of temperature.

Experimental evidence[1-4] strongly supports the idea that superconductivity in thin films is rapidly quenched when the normal-state sheet resistance R_N reaches a value on the order of 10^4 $\Omega/\square$. The destructive effect of increasing R_N on the superconducting state has been treated theoretically from a microscopic point of view[5-8] and found to be the result of disorder and interaction effects which combine to reduce the electron density of states and enhance the repulsive Coulomb interaction. In this Rapid Communication we report and interpret measurements of resistive transitions, critical fields, and magnetoconductance of thin In/InO_x composite films which reveal the systematics of how superconductivity in two dimensions (2D) is affected by disorder. A simple pair-breaking model is developed to explain the disorder-induced trends in the mean-field transition temperature T_{c0} and the effective electron diffusivity D determined from critical field H_{c2} measurements. Additivity of pair breakers is assumed and the disorder-induced pair-breaking rate which best describes the data is found to be proportional to R_N and independent of temperature.

As these In/InO_x composites have previously been shown to have an amorphous metallic component,[9] we can ignore the macroscopic charging effects, usually modeled for granular materials with an intergrain capacitance parameter,[10] and rely instead on a more microscopic interpretation in which R_N reflects the reduced density of states and enhanced Coulomb repulsion due to disorder. More importantly, previous measurements[11,12] on similar composites have verified the Kosterlitz-Thouless picture of vortex unbinding out to length scales approaching 100 μm. It is, therefore, reasonable to presume that this excellent homogeneity is sufficiently preserved for the films with slightly higher R_N and identical geometry reported here. As a clarifying remark, we would like to emphasize that the recent work of Ovadyahu[13] on three-dimensional indium oxide films treats material which is distinctly different: it is polycrystalline and it does not superconduct.

Figure 1 is a zero magnetic field plot of the resistive transitions of five different films each 500 μm long and 100 μm wide. All of these films have the same 100-Å thickness and the differences in R_N arise primarily from differences in composition. These data clearly manifest the delicate balance between "superconducting" [films (a)–(c)] and "insulating" [films (d)–(e)] behavior. For film (c), the measured sheet resistance at 8 K is 5650 $\Omega/\square$, only 19% lower than the 6970 $\Omega/\square$ of film (d). Significantly, it is within this very narrow range of resistance values near $\hbar/e^2 = 4117$ $\Omega/\square$ that superconductivity in the vortex fluctuation regime, where long length scales in these homogeneous films have been shown to be important,[11,12] is rapidly quenched. Previous investigations of high R_N thin-film superconductors have shown that the excess conductivity in the paraconductivity regime is well described by the Aslamazov-Larkin theory.[1,14] This is justified in dirty films because the large pair-breaking parameter, proportional to R_N, tends to suppress the Maki-Thompson contribution.[14] Accordingly, we have followed the procedure of our own earlier work[12] and used the Aslamazov-Larkin theory in two dimensions (2D) to obtain estimates of the mean-field transition temperatures T_{c0} which are shown as vertical arrows in Fig. 1 for films (a)–(c).

The theoretical model used to analyze these data is based upon a universal relation previously used in studies of gap-

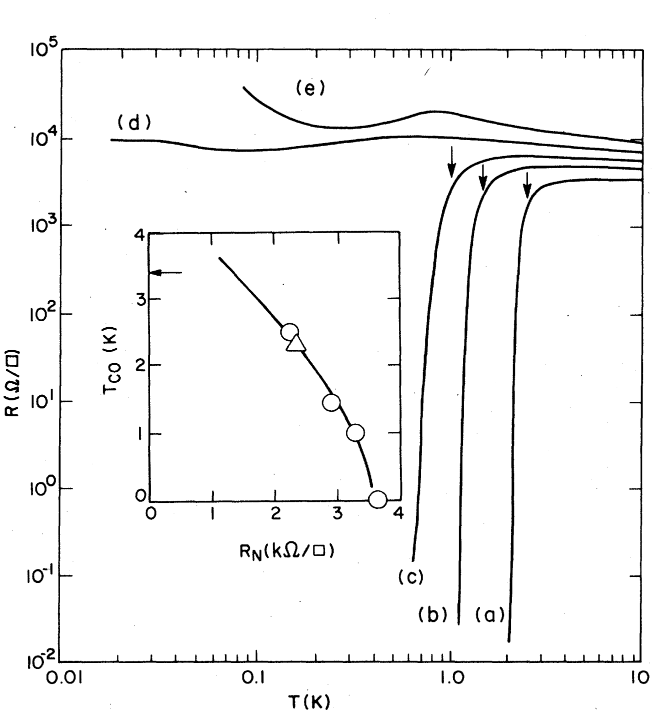

FIG. 1. Logarithmic plot of the resistance transitions of five 100-Å-thick In/InO_x composite films. The transition temperatures for films (a)–(c) are indicated by arrows and the inset is discussed in the text.

less superconductivity[15] and which for our purposes we write in the form

$$\ln\frac{T}{T_{cx}}-\Psi\left(\frac{1}{2}\right)+\Psi\left(\frac{1}{2}+\frac{eDH}{2\pi ck_BT}+\frac{\hbar}{4\pi k_BT\tau_p}\right)=0 \quad . \qquad (1)$$

In this expression Ψ is the digamma function, H the perpendicular magnetic field, τ_p the pair-breaking time associated with disorder, and T_{cx} the unshifted transition temperature which occurs in zero magnetic field and in the absence of disorder ($\tau_p^{-1}\rightarrow 0$). The remaining symbols have their usual meaning. The $T=T_{c0}$ and $H=H_{c2}$ solutions of Eq. (1) delineate the mean-field phase boundary between the superconducting and normal states. It should also be noted that the magnetic-field-induced pair breaker, proportional to H, is assumed to occur in additive combination with the disorder-induced pair breaker, proportional to τ_p^{-1}.

The central assumption of this paper is that the pair-breaking rate can be written in the form

$$\tau_p^{-1}=\frac{8k_BT_{cx}\gamma R_N}{\pi\hbar} \quad , \qquad (2)$$

where γ is a constant with dimensions of inverse sheet resistance to be determined by experiment. Substitution of Eq. (2) into Eq. (1) yields the result

$$\ln\frac{T}{T_{cx}}-\Psi\left(\frac{1}{2}\right)+\Psi\left(\frac{1}{2}+\frac{eDH}{2\pi ck_BT}+\frac{2\gamma T_{cx}R_N}{\pi^2T}\right)=0 \quad , \qquad (3)$$

which we will show below gives an excellent description of the disorder-induced trends in T_{c0} and H_{c2}. We have chosen the constants in Eq. (2), so that for small disorder-induced pair breaking ($\gamma R_N<<1$) in zero field ($H=0$), Eq. (3) reduces to the familiar form

$$\frac{T_{cx}-T_{c0}}{T_{cx}}=\gamma R_N \qquad (4)$$

used by previous investigators in studies of the $H=0$ fluctuation conductivity of thin-film superconductors.[14]

The inset of Fig. 1 is a plot of T_{c0} vs R_N for films (a)–(d). As film (d) does not superconduct we assign it $T_{c0}=0$. The additional film denoted by a triangle was similarly prepared and measured previously.[11] For R_N we use the *room-temperature* values of the sheet resistance as the most appropriate measure of the Boltzmann resistance. This reasoning is supported by our observation that the temperature coefficient of resistance of our films is small and negative near room temperature and remains negative down to $\sim$10 K. Accordingly, electron-phonon processes are not important for electrical transport and the increase in resistance with decreasing temperature for our highly disordered samples is due to localization and interaction corrections to the Boltzmann resistance. The solid line in the Fig. 1 inset results from an $H=0$ least-squares fit to the $T=T_{c0}$ solutions of Eq. (3) where T_{cx} and γ are the fitting parameters. The values $T_{cx}=4.72$ K and $\gamma=1.93\times10^{-4}\ \Omega^{-1}\Box$ are obtained. Although T_{cx} is considerably higher than the 3.4-K transition temperature of bulk indium (horizontal arrow), we note that the solid curve is not inconsistent with transition temperatures $\sim$3.6 K measured for thicker composite films with resistivity approaching a lower limit, dictated by microstructural considerations,[9] of 1000 $\mu\Omega$ cm. The value for γ is consistent with previously published results determined from excess conductivity studies in the paraconductivity regime of high R_N lead[1] and aluminum[14] films.

In the limit of small pair breaking (i.e., $eDH/2\pi ck_BT<<1$ and $2\gamma T_{cx}R_N/\pi^2T<<1$) the implicit solution for the critical field $H=H_{c2}$ in Eq. (3) can be expanded about the disorder-induced critical temperature T_{c0} to give an expression of the form

$$H_{c2}(T)=\frac{4ck_BT_{c0}}{\pi eD}\left[1-\frac{T_{cx}\gamma R_N}{T_{c0}}\right]\left(1-\frac{T}{T_{c0}}\right) \quad . \qquad (5)$$

This expression is equivalent to the well-known dirty-limit result[15] except for the term enclosed in square brackets which has the effect of reducing the critical-field slope parameter $[dH_{c2}(T)/dT]|T_{c0}$ as disorder (γR_N) is increased.

Experimentally, H_{c2} is defined as that field necessary to restore the resistance to the value measured at T_{c0} (cf. arrows in Fig. 1). The results for films (a)–(c), plotted in Fig. 2, show good agreement with the linear T dependence predicted by Eq. (5). The extrapolated $H_{c2}(0)$ for films (a) and (b) is well above the Pauli limit of $18\,400T_{c0}$ Oe.[15] We attribute this behavior as well as the upward curvature in the data at low reduced temperatures to a high spin-orbit scattering rate acting in combination with electron correlation effects which in the presence of a field tend to reduce the Coulomb repulsion.[6,16] In the absence of the bracketed correction term of Eq. (5) one would conclude from the data in Fig. 2 that the electron diffusivity D, proportional to the reciprocal of $[dH_{c2}(T)/dT]|T_{c0}$, *increases* with increasing R_N. This inference is not only counterintuitive, but it is in contradiction with theoretical predictions[6,16] and also does not agree with recent experimental results on thin InGe (Ref. 17), Zn (Ref. 18), and *a*-MoGe (Ref. 19) films. The effect of this novel disorder-induced renormalization of $[dH_{c2}(T)/dT]|T_{c0}$, predicted by Eq. (5) and shown in the data of Fig. 2, is made more explicit in the solid lines of Fig. 2 which represent the numerically determined implicit solutions of Eq. (3) for $H_{c2}(T)$. This family of theoretical

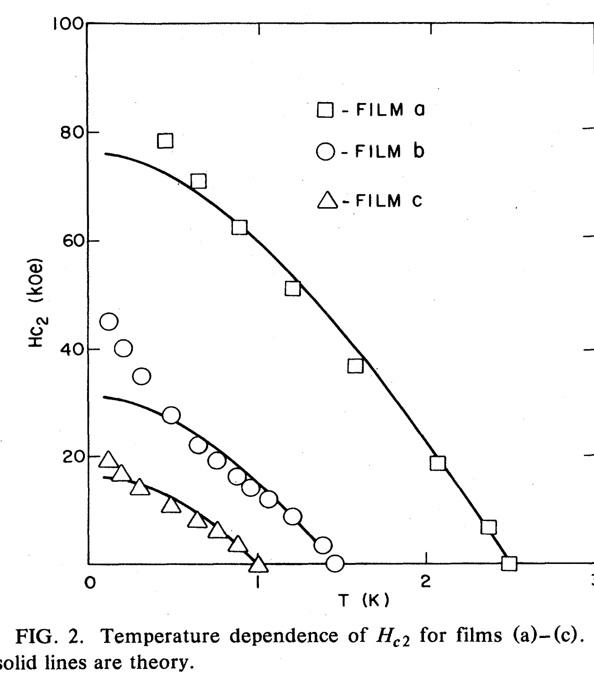

FIG. 2. Temperature dependence of H_{c2} for films (a)–(c). The solid lines are theory.

fits was obtained using the values of γ and T_{cx} determined previously (cf. Fig. 1 inset) and adjusting the remaining parameter D to have the same value $D=0.18$ cm^2/s for each of the three films. This latter assumption implies that the differences in R_N for these films are caused primarily by a disorder-induced reduction of the electron density of states at the Fermi level. We also note that this value for D is not appreciably different than the values obtained from Hall and electric field effect mobility studies made on thicker 3D films of similar composition.[20]

Ebisawa, Maekawa, and Fukuyama[7] have recently utilized a microscopic approach, including localization and interaction effects, to calculate a pair-breaking rate:

$$\tau_p^{-1}=\tau_i^{-1}=\frac{e^2R_Nk_BT}{2\pi\hbar^2}\ln\left(\frac{T_1}{T}\right) , \qquad (6)$$

where τ_i is the 2D electron-electron scattering time.[21] Since T_1, a function of D and the screening constant, is on the order of 10^9 K we can ignore the slow logarithmic variation in T and write [cf. Eq. (2)] $\tau_p^{-1}=8k_BT\gamma_0R_N/\pi\hbar$, where γ_0 is a constant. Substitution of this new rate into Eq. (1) yields a relation similar to Eq. (3) except for the absence of the T_{cx}/T factor in the last term. Interestingly, although the physical basis for Eq. (6) may be more firmly grounded in established theory[21] than Eq. (2), Eq. (1) used with Eq. (6) *does not explain* either the *negative* curvature and the rapid suppression of T_{c0} with increasing R_N (cf. Fig. 1 inset) or the renormalization downwards of $(dH_{c2}/dT)|T_{c0}$ with increasing R_N (cf. Fig. 2). In contrast, the T_{c0} vs R_N dependence of lower resistivity ($\sim 200\ \mu\Omega$ cm) films of Zn (Ref. 18) and a-MoGe (Ref. 19) shows a more gradual decrease with *positive* curvature which is consistent with the use of Eq. (6) in Eq. (1).

Further evidence for the inappropriateness of the inelastic electron-electron pair-breaking mechanism of Eq. (6) with regard to our films can be found in magnetoconductance data from which we can extract a direct measure of τ_i^{-1}. Here, we use 2D theory in which the mitigating effect of magnetic fields on weak localization[22] occur in additive combination with interaction effects calculated from the Maki-Thompson diagram.[23] Our analysis of the data is similar to that used in previous investigations of relatively clean ($R_N < 200\ \Omega/\square$) aluminum films.[24] Figure 3 shows the resulting temperature dependences of τ_i^{-1} for film (a) (circles) and film (d) (triangles). For high enough temperatures (4 K $\lesssim T \lesssim$ 10 K) the effect of superconducting fluctuations on τ_i, to be discussed below, is presumably unimportant, and a comparison of the data in Fig. 3 with theory [Eq. (6)] can be made. In this regard we see that although τ_i^{-1} scales approximately linearly with T as predicted by Eq. (6), we do not see the expected dependence on R_N which should scale the magnitude of τ_i^{-1} for film (d) (triangles) a factor of 1.6 higher than film (a) (circles). Clearly the measured τ_i^{-1} for our highly disordered films does not scale with R_N as would be implied by Eq. (6). We also note that the magnitude of τ_i^{-1} determined experimentally at, say, 5 K (cf. Fig. 3) is a factor of 5 smaller than Eq. (6) predicts.[7,21]

A clue to the possible resolution of this discrepancy may be that the effect of superconducting fluctuations on τ_i has not been included in the theory.[7,21] The plausibility of such a correction, especially near T_{c0} can be made more apparent by comparing the inelastic diffusion length $L_i=(D\tau_i)^{1/2}$

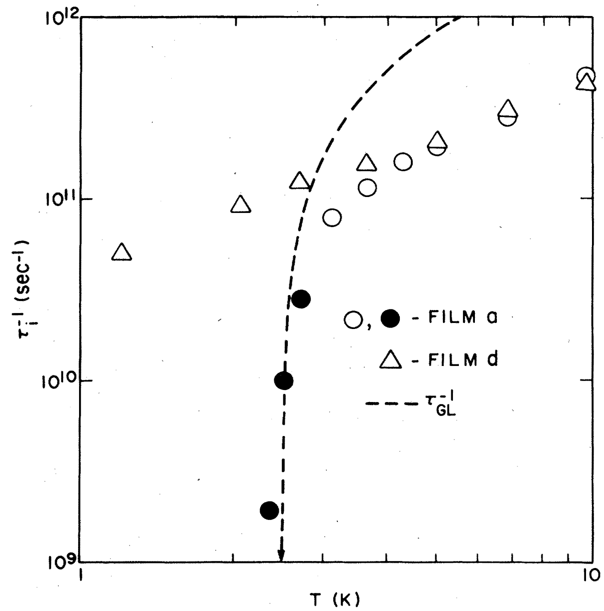

FIG. 3. Temperature dependence of τ_i^{-1} for films (a) (circles) and (d) (triangles). The solid points delineate the Aslamazov-Larkin region discussed in the text.

with the Ginzburg-Landau coherence length $\xi_{GL}(T) = [\pi\hbar D/8k_B(T-T_{c0})]^{1/2}$ which is a measure of the average size of the fluctuating superconducting islands in the paraconductivity regime.[14] In the temperature range near T_{c0} where $\xi_{GL} > L_i$, one might expect the pseudogap associated with the formation of superconducting islands to begin to restrict the phase space available to inelastic scattering events; thus, giving rise to some of the deviation of τ_i^{-1} from power law behavior observed for film (a) at low T in Fig. 3. Close to T_{c0}, however, we expect the Aslamazov-Larkin contribution to dominate the magnetoconductance so that τ_i^{-1} now becomes a measure of the fluctuation rate $\tau_{GL}^{-1}=D\xi_{GL}^{-2}$ of the superconducting islands in the paraconductivity regime which, for film a with $T_{c0}=2.494$ K (cf. arrow on abscissa), has the theoretical dependence indicated by the dashed line. Calculations using a theory[25] which does not include localization corrections show that the Aslamazov-Larkin contribution to the magnetoconductance of film (a) is small at high temperatures and becomes roughly equal to or greater than the measured magnetoconductance at lower temperatures (solid circles).

In conclusion, the pair-breaking formalism developed here gives an excellent account of the observed behavior of T_{c0} and H_{c2} as a function of increasing disorder. There is, however, a clear need for a more complete theory which can account for the unique temperature-independent pair-breaking rate of Eq. (2) which describes so well the disorder-induced trends in T_{c0} and H_{c2} observed here. It may be that the extreme disorder associated with the high resistivities of our films ($\sim 3000\ \mu\Omega$ cm at room temperature), a factor of 10 higher than the resistivities of thin films discussed in previously published work,[17–19] may require a new set of theoretical assumptions. Finally, we note that our explanation for the precipitous quenching of superconductivity observed in these films does not require a crossover from weak to strong localization.[26]

The authors are grateful to E. Abrahams, R. N. Bhatt, A. T. Fiory, J. M. Gordon, P. A. Lee, W. L. McLean, and T. V. Ramakrishnan for useful and stimulating discussions and to P. C. Hohenberg for a critical reading of the manuscript. The technical assistance of H. W. Dail and R. H. Eick is also appreciated.

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