Low-Temperature Insulating Phases of Uniformly Disordered Two-Dimensional Superconductors

M. A. Paalanen, A. F. Hebard, and R. R. Ruel
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 22 May 1992)

Temperature- and magnetic-field-dependent measurements of the longitudinal R_{xx} and transverse R_{xy} resistance of amorphous indium oxide thin-film superconductors reveal the presence of distinct insulating phases for sufficiently high disorder and/or magnetic field. For field-swept transitions at fixed disorder and low temperatures there is a critical field where R_{xx} diverges and the superconductor is transformed into a Bose-glass insulator with localized Cooper pairs. At higher fields there is a second critical field where R_{xy} appears to diverge, the pairs unbind, and localized single electrons characterizing a Fermi-glass insulator dominate.

PACS numbers: 74.65.+n, 73.60.Hy, 74.70.Mq, 74.75.+t

Numerous studies, both theoretical and experimental, have been concerned with the competition between disorder and superconductivity [1]. Disorder induces localized eigenstates and enhanced Coulomb repulsions which jointly act to reduce the transition temperature and ultimately quench all remnants of superconductivity. This competition between disorder and superconductivity is particularly interesting in two dimensions (2D) where the delicate and marginally attractive pairing interaction is derived from electronic states which are localized for arbitrary weak disorder. Experimentally, systematics of the destruction of superconductivity with increasing disorder in thin films is studied by varying the thickness [2-7], composition [3,8], or resistivity for fixed thickness [9]. Convergence between experimental results and theoretical description has been somewhat hampered by the wide variation in microstructural and compositional properties of these thin-film systems. Recent scaling theories [10-12] of the superconducting-insulating (S-I) transition have avoided these details by taking a broad view and formulating a description which is conveniently insensitive to microscopic material properties. In this description the paired electrons and vortices are treated as quantum-mechanical objects which obey Bose statistics and hence can be treated within a purely "bosonic" formulation. There is experimental evidence for the predicted S-I transitions, driven either by increasing disorder in zero magnetic field [6,7,9] or by increasing field for fixed disorder [13,14]. In these experiments the putative S-I transition takes place at $T=0$ sheet resistances which vary within a factor of 2 of the predicted universal value, $h/4e^2=6450\ \Omega/\Box$. Measurements of the field-tuned S-I transition of InO_x films [13], however, confirm detailed predictions [11] of the theory such as the unity value of the dynamical exponent and the scaling collapse of the temperature- and field-dependent resistance data onto a single curve.

An implicit assumption of the scaling treatment is that Cooper pairs exist and are finite in extent at the S-I transition. Thus the magnitude Ψ_0 of the order parameter is finite on both sides of the S-I transition and the true superconducting phase transition, where long-range order is established and the dc resistance is zero, is dominated by fluctuations in the phase ϕ of the order parameter. These statements are unarguably true for granular films [4] where Ψ_0 remains relatively unchanged on each grain as disorder or intergranular resistance increases and suppresses the Josephson phase coherence between neighboring grains. The situation is somewhat different for amorphous or homogeneously disordered films since Ψ_0, as measured by the energy gap [15] or mean-field transition temperature T_{c0} [9], is strongly reduced from its clean-limit value. In those systems where the microscopic disorder is sufficiently uniform, a Kosterlitz-Thouless vortex-antivortex transition [16] at $T=T_c$ is observed [8,9], and it is clear that vortices (bound as pairs below T_c and individually mobile above T_c) and hence Cooper pairs exist on both sides of the transition. There is at present no compelling experimental evidence for the *simultaneous* disappearance in 2D of both the phase and magnitude of the order parameter at the superconducting phase boundary.

In this Letter we present evidence for the existence of Cooper pairs on the insulating side of the two-dimensional S-I transition and show that there are two distinct and separate insulating phases, both of which can be accessed by magnetic-field tuning through and beyond the S-I phase boundary. A schematic of the relevant 2D superconducting and insulating phases is shown in the $T=0$ plot in Fig. 1(a) in which the theoretically introduced [10-12] parameter for disorder, Δ, and perpendicular magnetic field B are plotted on mutually perpendicular axes. For sufficiently low fields and Δ less than a critical value Δ_c, the superconducting vortex-glass state exists. In this state the Cooper pairs are condensed into a macroscopic extended quantum state which coexists with quantum vortices localized (and hence immobile) in random potential-energy minima associated with the disorder. The S-I transition occurs at the solid-line phase boundary where the longitudinal sheet resistance R_{xx} has a universal value R_{xx}^* near the quantum value, $h/4e^2=6450\ \Omega/\Box$. In the adjacent Bose-insulating phase the dual role of the vortices and pairs is interchanged: The vortices, now mobile, are Bose condensed into a macroscopic quantum

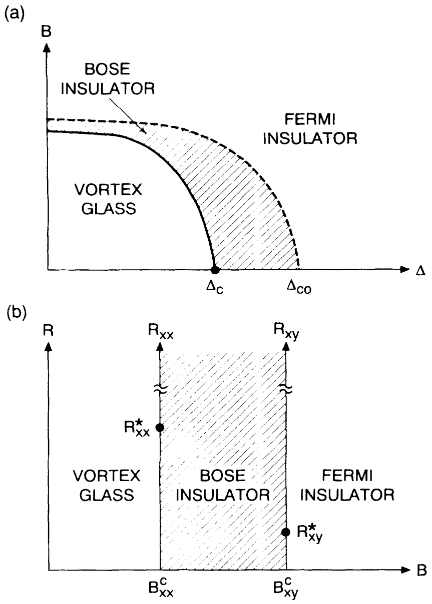

FIG. 1. Schematic showing the relationship between the vortex-glass, Bose-insulator, and Fermi-insulator phases of a 2D superconductor in a perpendicular magnetic field B. (a) The separate phases as a function of field B and disorder Δ, with critical disorder Δ_c marking the $B=0$ superconducting-insulating transition and Δ_{c0} marking the $B=0$ disappearance of localized pairs. (b) The divergences of R_{xx} and R_{xy} through critical resistances R_{xx}^* and R_{xy}^* and at critical fields B_{xx}^c and B_{xy}^c, respectively.

phase and the Cooper pairs become simultaneously localized. Experimental evidence for this picture has been provided in studies of the temperature and magnetic-field dependence of R_{xx} in amorphous InO_x films [13] and oxygen-deficient $YBa_2Cu_3O_x$ crystals [14]. The "bosonic" description of this S-I transition does not take into account the role of unpaired electrons which would be expected to become important when either B or Δ are high enough to ensure the dissolution of the localized pairs. Our measurements, described below, of the field-tuned transverse (Hall) sheet resistance R_{xy} show that there is a surprisingly well-defined and pronounced transition [dashed line in Fig. 1(a)] which terminates for $B=0$ at a higher critical disorder, $\Delta=\Delta_{c0}$, and which can be interpreted as the boundary between a Bose-insulator state containing localized paired electrons and a Fermi-insulator state containing localized single electrons. Moreover, a critical *nonuniversal* sheet resistance R_{xy}^* which increases with increasing disorder and which separates the insulating phases is experimentally determined.

Amorphous composite indium-oxide (InO_x) films, 100 Å thick and 200 μm wide, were prepared with different resistivities on glass or oxidized silicon substrates. Three pairs of equally spaced voltage taps, shown in the Fig. 2

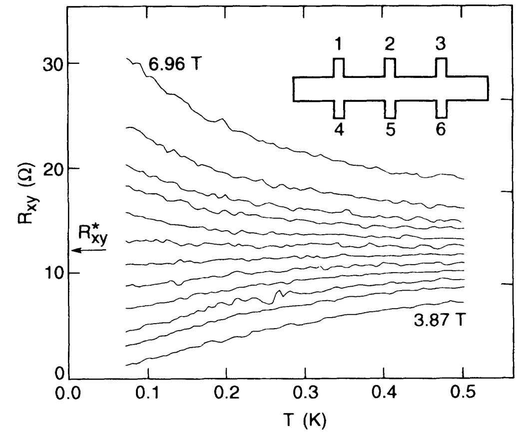

FIG. 2. Temperature dependence of R_{xy} of a 100-Å-thick InO_x film at magnetic fields spanning the range 3.87 T $< B <$ 6.96 T. Inset: A plan view of the voltage lead arrangement used to make the R_{xx} and R_{xy} measurements.

plan-view inset, allowed two independent voltage measurements (V_{12} or V_{45}, V_{23} or V_{56}) of R_{xx} and three independent voltage measurements (V_{14}, V_{25}, V_{36}) of R_{xy}. The agreement of measured normal-state values of R_{xx} on adjacent segments of the same film to within approximately 0.1% indicates excellent uniformity of film properties. The Hall data were taken for both field polarities, and misalignment voltages typically turned out to be no more than a factor of 2 higher than the Hall voltages.

The temperature dependence of R_{xy} for one of our films at field intervals spanning the range 3.87 to 6.96 T is shown in Fig. 2. The critical resistance R_{xy}^*, indicated by the horizontal arrow, defines a critical field B_{xy}^c below which R_{xy} is assumed to approach zero at $T\to 0$ (super-

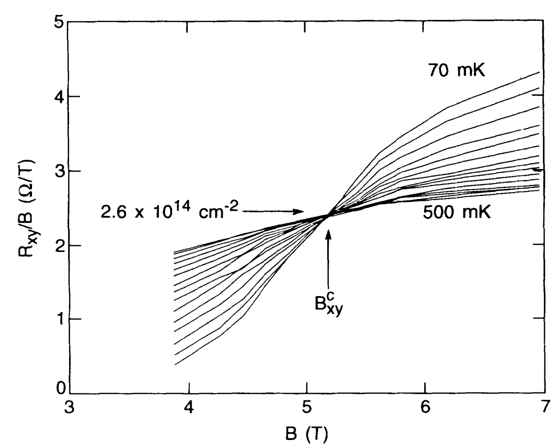

FIG. 3. Magnetic-field dependence of R_{xy}/B interpolated from the data of Fig. 2 at equally spaced temperature intervals spanning the range 70 mK $< T <$ 500 mK. An areal electron density of 2.6×10^{14} cm^{-2} (horizontal arrow) is calculated from $R_{xy}^*=12.4$ Ω and $B_{xy}^c=5.1$ T (vertical arrow) measured for this film.

conducting state) and above which R_{xy} is assumed to approach infinity as $T \to 0$ (insulating state). Except for the significantly reduced resistance scales, these data are remarkably similar to R_{xx} data used to demonstrate the S-I transition in previous work [13]. Figure 3 shows the R_{xy} data of Fig. 2 divided by B and plotted against B for interpolations at equally spaced temperatures spanning the range from 70 to 500 mK. The twisting of the isotherms into a "knot" at a well-defined critical field B_{xy}^c is equally pronounced for the five films investigated and is independent of whether R_{xy} or R_{xy}/B is plotted on the ordinate. Furthermore, the values for R_{xy}^* and B_{xy}^c determined from *isomagnetic* lines such as shown in Fig. 2 are in good agreement with the values determined from *isothermal* lines such as shown in Fig. 3. These data thus imply a temperature-independent R_{xy}^* and B_{xy}^c which parametrize a $T=0$ transition from a state with R_{xy} small and probably zero to a state with R_{xy} large and probably infinite.

Interestingly, the field-induced divergences of R_{xx} and R_{xy} do not occur at the same fields. This is shown in Fig. 4 for a second film where the 40-, 100-, and 200-mK isotherms of R_{xx} (left-hand ordinate) and of R_{xy} (right-hand ordinate) are plotted. The critical field B_{xy}^c is noticeably larger than B_{xx}^c. Permutation of the voltage leads shown in the inset of Fig. 1 allowed a consistency check of the two independent R_{xx} measurements and the three independent R_{xy} measurements, thus removing the possibility of spatial gradients in film properties giving rise to the observed differences.

The systematic dependence of the critical fields, B_{xx}^c and B_{xy}^c, and resistances, R_{xx}^* and R_{xy}^*, on disorder is shown in Fig. 5 for five films. The abscissa variable B_{xy}^c is a reciprocal measure of disorder Δ; that is, higher B_{xy}^c represents a cleaner film with lower resistivity (less disorder) and a higher T_c. Figure 5(a) shows that B_{xx}^c becomes significantly smaller than B_{xy}^c as disorder increases. Figure 5(b) shows that R_{xy}^* is not constant and independent of Δ as is R_{xx}^* but rather increases with increasing Δ. The threefold increase of R_{xy}^* should be contrasted with the relatively constant value of $R_{xx}^* = 6245 \pm 420$ $\Omega/\square \approx h/4e^2$ measured for these same five films.

A plausible scenario implied by the data of Figs. 2–5 for the $T=0$ phases of a 2D superconductor is summarized in the R vs B schematic of Fig. 1(b). At low fields, $B < B_{xx}^c$, the film is in the superconducting vortex-glass state where both R_{xx} and R_{xy} are zero. The field $B = B_{xx}^c$ marks the S-I boundary where R_{xx} diverges, with critical behavior occurring at R_{xx}^*. At higher fields, in the range $B_{xx}^c < B < B_{xy}^c$, the Bose-insulator phase containing localized charge pairs is present and is characterized by $R_{xx} \to \infty$ and R_{xy} small and possibly zero. Experimentally (cf. Fig. 4) we cannot tell if R_{xy} is exactly zero in the zero-temperature limit or whether it is small and perhaps linear in B. Theoretical arguments [17] invoking particle-hole symmetry suggest that $R_{xy}=0$ exactly at the S-I boundary, a behavior consistent with the break point near $B = B_{xx}^c$ in the 40-mK isotherm for R_{xy} in Fig.

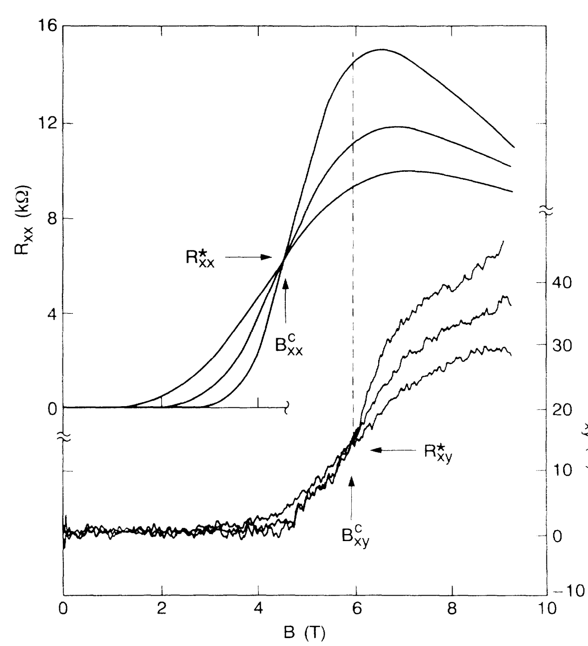

FIG. 4. Field dependence of R_{xx} (left-hand axis) and R_{xy} (right-hand axis) for the same film showing the separation between B_{xx}^c and B_{xy}^c.

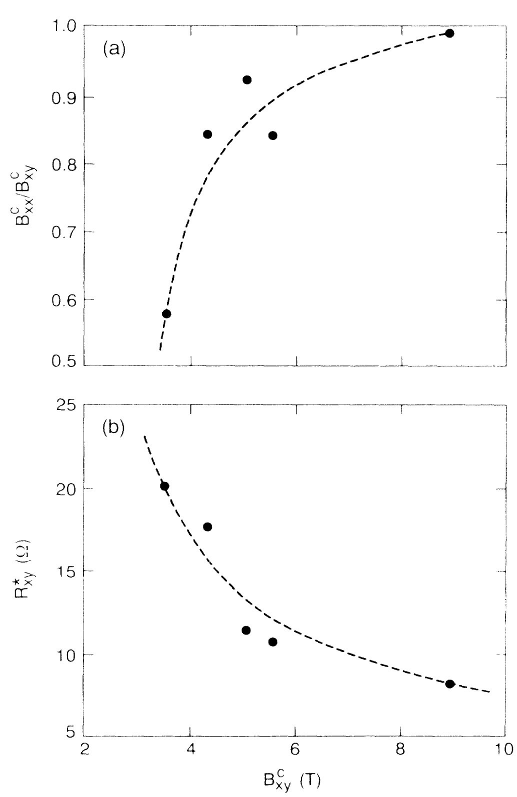

FIG. 5. Dependence of (a) B_{xx}^c/B_{xy}^c and (b) R_{xy}^* on B_{xy}^c for the five films studied. Disorder decreases with increasing B_{xy}^c.

4. At $B=B_{xy}^c$ our data clearly show that R_{xy} appears to diverge, through a nonuniversal "critical" point where $R_{xy}=R_{xy}^*$, to a phase, which we identify as the Fermi insulator, where both R_{xx} and R_{xy} are large and Cooper pairs no longer exist. The assumption that R_{xy} should diverge for localized electrons is consistent with recent theory [18].

Three additional comments should help clarify this interpretation. First, from Fig. 3 we infer that R_{xy}^* is a measure of normal-state properties. This is seen by plotting the field dependence of R_{xy}/B rather than R_{xy} and noting that the highest-temperature isotherm at 500 mK is almost horizontal, thus implying that the electron areal charge density evaluated at the critical point, $N_c=B_{xy}^c/eR_{xy}^*$, is close to that of the normal state. Using the values $R_{xy}^*=12.4$ Ω and $B_{xy}^c=5.2$ T, we obtain the result $N_c=2.6\times10^{14}$ cm^{-2}, which for a 100-Å-thick film implies a volume density of 1.6×10^{20} cm^{-3}. This value for the carrier density in the normal state together with the trend of decreasing carrier density with increasing disorder agrees with the results of Hall and electric-field mobility measurements [19] on similar films. Second, the maximum in R_{xx} and the subsequent decrease in R_{xx} as B increases further (cf. Fig. 4) is consistent with the notion that an insulator with localized charge pairs should have a higher resistance than an insulator with localized single electrons. A similar statement has been made with respect to the unbinding of charged pairs in 2D tunnel-junction arrays [20]. The observed simultaneous increase in R_{xy} and decrease in R_{xx} is unlikely to be associated with the formation of a Coulomb gap. Third, the reduction of B_{xx}^c/B_{xy}^c with increasing disorder [cf. Fig. 5(b)] is analogous to the reduction of T_c/T_{c0} with increasing normal-state sheet resistance [16]. The analogy here is that T_c at zero magnetic field and B_{xx}^c at zero temperature mark distinct and separate superconducting phase boundaries where fluctuations in ϕ dominate, whereas T_{c0} at zero magnetic field and B_{xy}^c at zero temperature mark distinct and separate crossover regimes where fluctuations in Ψ_0 dominate.

The surprising result of the transverse-resistance data presented here is that the crossover at $B=B_{xy}^c$ is so distinct. Thus, in addition to the superconducting-insulating phase boundary with universal properties [solid line of Fig. 1(a)], there appears to be a second phase boundary with nonuniversal properties (dashed line) which separates two insulating phases and presumably marks the sudden disappearance of Cooper pairs. In the absence of theory for a true phase transition, we refer to the boundary between these insulators as a *region of pronounced crossover* rather than a phase boundary. Such a crossover might be characterized in the limit of $R_{xx}\to\infty$ by a power-law relation such as $R_{xy}=(R_{xx})^{\eta(B)}$, where η is a field-dependent exponent which continuously varies from negative values where $R_{xy}\to0$, through zero where R_{xy} is a constant, to positive values where $R_{xy}\to\infty$. As a final remark, we note the similarity of the $T=0$ Hall insulator [21], which is characterized by $R_{xx}\to\infty$ and $R_{xy}\approx B/Ne$, and the $T=0$ Bose insulator identified here, which is characterized by $R_{xx}\to\infty$ and $R_{xy}^*=B_{xy}^c/N_ce$ at $B=B_{xy}^c$. The Fermi insulator, having both R_{xx} and R_{xy} infinite, is distinctly different.

We are grateful to B. Altschuler, M. P. A. Fisher, S. M. Girvin, D. A. Huse, S. A. Kivelson, and B. G. Kotliar for interesting discussions and helpful comments.

[1] T. V. Ramakrishnan, Phys. Scr. **T27**, 24 (1989), and references therein.
[2] M. Strongin, R. S. Thompson, O. F. Kammerer, and J. E. Crow, Phys. Rev. B **1**, 1078 (1970).
[3] J. M. Graybeal and M. A. Beasley, Phys. Rev. B **29**, 4167 (1984).
[4] H. M. Jaeger, D. B. Haviland, A. M. Goldman, and B. G. Orr, Phys. Rev. B **34**, 4920 (1986), and references therein.
[5] R. C. Dynes, A. E. White, J. M. Graybeal, and J. P. Garno, Phys. Rev. Lett. **57**, 2195 (1986).
[6] S. J. Lee and J. B. Ketterson, Phys. Rev. Lett. **64**, 3078 (1990).
[7] D. B. Haviland, Y. Liu, and A. M. Goldman, Phys. Rev. Lett. **62**, 2180 (1989).
[8] K. Epstein, A. M. Goldman, and A. M. Kadin, Phys. Rev. Lett. **47**, 534 (1981).
[9] A. F. Hebard and M. A. Paalanen, Phys. Rev. Lett. **54**, 2155 (1985), and references therein.
[10] M. P. A Fisher, G. Grinstein, and S. M. Girvin, Phys. Rev. Lett. **64**, 587 (1990).
[11] M. P. A. Fisher, Phys. Rev. Lett. **65**, 923 (1990).
[12] M.-C. Cha, M. P. A. Fisher, S. M. Girvin, M. Wallin, and A. P. Young, Phys. Rev. B **44**, 6883 (1991).
[13] A. F. Hebard and M. A. Paalanen, Phys. Rev. Lett. **65**, 927 (1990).
[14] G. T. Seidler, T. F. Rosenbaum, and B. W. Veal, Phys. Rev. B **45**, 10162 (1992).
[15] J. M. Valles, R. C. Dynes, and J. P. Garno, Phys. Rev. B **40**, 6680 (1989).
[16] J. M. Kosterlitz and D. J. Thouless, J. Phys. C **6**, 1181 (1973); M. R. Beasley, J. E. Mooij, and T. P. Orlando, Phys. Rev. Lett. **42**, 1165 (1979); B. I. Halperin and D. R. Nelson, J. Low Temp. Phys. **36**, 599 (1979).
[17] M. P. A. Fisher (to be published).
[18] X.-F. Wang, Z. Wang, G. Kotliar, and C. Castellani, Phys. Rev. Lett. **68**, 2504 (1992).
[19] A. T. Fiory and A. F. Hebard, Phys. Rev. Lett. **52**, 2057 (1984).
[20] J. E. Mooij, B. J. van Wees, L. J. Geerlings, M. Peters, R. Fazio, and G. Schön, Phys. Rev. Lett. **65**, 645 (1990).
[21] S. Kivelson, D.-H. Lee, and S.-C. Zhang, Phys. Rev. B **46**, 2223 (1992).