Unusual insulating phase at low temperature in thin indium films

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We have prepared a series of thin indium films whose disorder is systematically introduced, and measured the temperature-dependent and magnetic-field-dependent Hall resistance R_{xy} as well as the longitudinal resistance R_{xx} at low temperatures. By increasing the field at fixed disorder, we have found, in addition to a usual critical field B_{xxC} where $R_{xx}(T\rightarrow 0)\rightarrow\infty$, another critical field B_{xyC} ($>B_{xxC}$) where $R_{xy}(T\rightarrow 0)$ diverges. With increasing disorder, B_{xxC} decreases faster than B_{xyC}, thus the region $B_{xxC}<B<B_{xyC}$ becomes broader as the critical disorder is approached. We suggest a possibility that the region $B_{xxC}<B<B_{xyC}$ corresponds to the Bose-insulator phase.

I. INTRODUCTION

The superconductor-insulator (*S-I*) transition induced by disorder has been extensively studied in various two-dimensional (2D) systems, such as thin continuous films and granular ones which consist of small grains connected by Josephson junctions. In these systems, the experimental data reveal that global phase coherence disappears when the (longitudinal) sheet resistances R_{xxn} in the normal state exceed the values R_C, which are order of $h/4e^2=6.5$ kΩ.[1–5] Meanwhile, Fisher[6] has considered on the basis of the "bosonic" description the influence of a perpendicular magnetic field B on 2D superconductors and predicted a fundamentally different type of field-induced *S-I* transition. When the field B is increased at low temperatures, field-induced vortices Bose condense at some critical field B_{xxC}; thus the transition from a superconductor with localized vortices ("vortex glass") to an insulator with localized Cooper pairs ("Bose glass") takes place. In his picture, a competition between condensation of Cooper pairs and vortices is essential. Based on these arguments, he has developed a scaling theory for the *S-I* transition. Experimental verifications for the transition have been suggested from the unity value of the critical exponent $z_B\nu_B$ and/or the scaling collapse of the temperature- and field-dependent resistance $R(T,B)$ onto a scaling curve.[7–12]

Recently, Paalanen, Hebard, and Ruel[13] have measured the longitudinal R_{xx} and the transverse R_{xy} resistance of amorphous composite InO_x films at low temperatures in various magnetic fields B and found a distinct insulating phase. When B is tuned through B_{xxC} at $T\rightarrow 0$, a superconducting state where both R_{xx} and R_{xy} are zero is transformed into an insulating state where R_{xx} diverges and R_{xy} is small or zero. With further increase in B, there appears a second critical field B_{xyC} at which both R_{xx} and R_{xy} diverge. They have interpreted the second critical field B_{xyC} as the phase boundary or the region of crossover between a "Bose-glass insulator" containing localized Cooper pairs and a "Fermi-glass insulator" containing localized single electrons. They have studied five films, which are superconducting at $B=0$, with different normal-state resistances and found that B_{xxC}/B_{xyC} decreases from 1 to about 0.6 with increasing disorder. From the results, they have proposed a phase diagram (Fig. 1 in Ref. 13) showing schematically the relationship between the vortex-glass, Bose-insulator, and Fermi-insulator phases of a 2D superconductor in the disorder-*B* plane. The proposed phase diagram is so novel and interesting, however, that experimental work including the data in the highly disordered regime where B_{xxC} vanishes, has not been performed yet.

In this report, we have made similar and more systematic measurements of R_{xx} and R_{xy} for field-swept transitions over a wider range of disorder through the disorder-driven *S-I* transition ($B_{xxC}\rightarrow 0$) using granular films of indium. We have investigated a series of nine films with average grain size $\bar{d}$ of 13 nm, because we know from previous studies[11,12] that films with $\bar{d}<20$ nm can be viewed as a dirty superconductor rather than a Josephson network[4] and well obey the scaling theory by Fisher. The advantage of the present system is that we have only to fabricate one clean film, and disorder can be introduced systematically in it, as described below, by weakening the coupling strength between grains, keeping other parameters constant.

II. EXPERIMENTAL

A granular film is fabricated at room temperature by repeating the cycles of vacuum deposition of a small amount of indium with average thickness (mass thickness) of 4–5 nm and surface oxidation four times. The film thus obtained is composed of nearly two-dimensionally coupled superconducting particles with average grain size $\bar{d}$ of 13 nm. The degree of surface oxidation is controlled so that the sheet resistance in the normal state R_{xxn} ($T=10$ K) is 1.3 kΩ (film 1). This method has been employed in studying the disorder-driven *S-I* transition for two-dimensional superconductors.[4] After the low-temperature measurements are performed, the film (film 1) is uniformly heated to 370 K in air. This heat treatment results in a monotonic increase

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in R_{xxn}. For example, the initial resistance $R_{xxn}=1.3$ kΩ for film 1 is slightly increased to 1.35 kΩ (film 2) after ~1 h heating, and in the final stage of the study we obtain the most resistive film with $R_{xxn}=4.1$ kΩ (film 9) by heating for a total of ~10 h. From transmission electron microscopy (TEM) observation, apparent structural changes have not been observed within the experimental resolution. We accordingly consider that the increase in R_{xxn} by heat treatment is due to the growth of a surface oxide layer and/or to a slight opening between the grains.

Good uniformity of the film is confirmed from the fact that the values of R_{xxn} for neighboring segments of the film differ by only 0.1% or less. A dc electric current is used to measure simultaneously the longitudinal R_{xx} and transverse (Hall) R_{xy} resistances. The Hall voltage is measured using a pair of equally spaced voltage probes which lie between pairs of equally spaced voltage probes for the R_{xx} measurements. The longitudinal component of the voltage arising from slight misalignment of the Hall probes is eliminated by reversing the magnetic field. Each film is directly immersed into liquid ^{3}He or ^{4}He to achieve good thermal contact. In various magnetic fields, the temperature is measured and controlled by means of calibrated carbon resistors and the vapor pressure of liquid ^{3}He or ^{4}He. In this study, the magnetic field B is directed perpendicularly to the film surface.

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the sheet resistance $R_{xx}(T)$ for five selected films in zero magnetic field. Films with R_{xxn} smaller than 3 kΩ (films 1–7) exhibit a superconducting transition, while those with R_{xxn} greater than 3 kΩ (films 8 and 9) behave like an insulator, showing an increase in $R_{xx}(T)$ at low temperatures. Thus the critical sheet resistance R_C turns out to be about 3 kΩ. This value of R_C, together with the tendency for the superconducting transition temperature T_C to decrease with increasing R_{xxn}, is consistent with previous results for dirty superconductors.[4]

When we plot the temperature dependences of R_{xx} and R_{xy}/B (R_{xy} divided by B), for the most conductive film (film 1) in various fields, we notice that the data for R_{xy}/B look remarkably similar to the data for R_{xx}. In both cases, with increasing B the slope of the isomagnetic curve below 1 K changes its sign from positive to negative at around $B=5.3$–5.5 T, suggesting that a field-induced transition from a state where R_{xx} and R_{xy}/B (or R_{xy}) are zero to a state where R_{xx} and R_{xy}/B (or R_{xy}) are infinity takes place at $T=0$. The critical fields B_{xxC} and B_{xyC}, which, respectively, cause R_{xx} and R_{xy}/B (or R_{xy}) at low temperatures to be constant, are estimated to be 5.35 and 5.50 T. In Figs. 2(a) and 2(b), the R_{xx} and R_{xy}/B data at various temperatures are plotted as a function of B. In each figure, one can clearly notice the presence of an intersection point of the isothermal lines at low temperatures. B_{xxC} and B_{xyC} can be determined immediately from the intersections in Figs. 2(a) and 2(b), since they correspond, respectively, to temperature-independent points (B_{xxC},R_{xxC}) and (B_{xyC},R_{xyC}) at low temperatures. Thus determined values of $B_{xxC}=5.35$ T and $B_{xyC}=5.50$ T are in good accord with the values obtained above. We emphasize that B_{xyC} is larger than B_{xxC}.

Qualitatively similar behaviors of R_{xx} and R_{xy}/B to those shown in Figs. 2(a) and 2(b) are observed for dirtier films (films 2–7) with higher R_{xxn} and lower T_C. For these films, B_{xxC} becomes remarkably smaller than B_{xyC} with increasing disorder (R_{xxn}). As shown in Figs. 2(c) and 2(d), the difference between B_{xyC} and B_{xxC} for film 7 ($R_{xxn}=2.4$ kΩ) is 0.45 T, which is certainly larger than that (0.15 T) for film 1 ($R_{xxn}=1.3$ kΩ). For more disordered films 8 ($R_{xxn}=3.2$ kΩ) and 9 ($R_{xxn}=4.1$ kΩ) B_{xxC} is considered to be zero, since $R_{xx}(T)$ does not exhibit the superconducting transition but shows an increase at low temperatures as seen in Fig. 1. Nevertheless, B_{xyC} for film 8 takes a noticeably large value around 3.5 T. For the most resistive film 9 ($R_{xxn}=4.1$ kΩ), we cannot determine the B_{xyC} value without great ambiguities because the R_{xx} below 1 K becomes so large that the measuring current must be kept low enough to prevent Joule heating, leading to less precision in determination of $R_{xy}(T)$.

To see the relation between $R_{xx}(T)$ and $R_{xy}(T)$ at low temperatures more closely, we plot, for various magnetic fields, data for $R_{xy}(T)$ as a function of corresponding data for $R_{xx}(T)$ on a log-log scale with T as a running variable (Fig. 3). It is found from Fig. 3 (upper part) that most of the data points that do not lie in the vicinity of the field-induced S-I transition clearly display the striking power-law relationship expressed as

$$R_{xy}(T)=A[R_{xx}(T)]^{\beta}, \tag{1}$$

where A is a positive constant nearly independent of the magnetic field (but sample dependent). The exponent $\beta(B)$ for films 1 and 7 is plotted in Figs. 4(a) and 4(b), respectively, as a function of magnetic field B. It is obvious from these figures that in each film $\beta(B)$ tends to a value which is close to 2 or more with decreasing B. Similar scaling behaviors of $R_{xy}(T)$ as a function of $R_{xx}(T)$ have

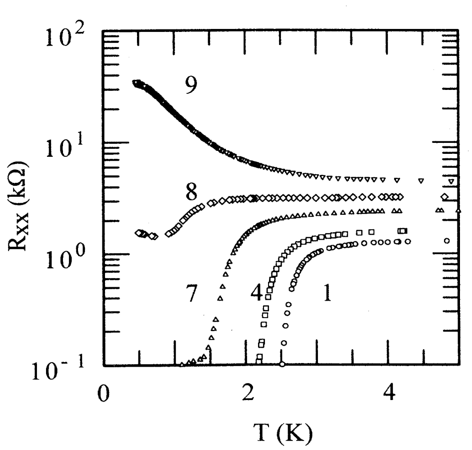

FIG. 1. Temperature dependence of the sheet resistance $R_{xx}(T)$ for five selected films.

been reported in several high-T_C superconductors.[14–16] Thus the present results indicate that the scaling behavior expressed by Eq. (1) is a general feature in the mixed state of the superconductors. As the field B increases and approaches the critical field, $B_{xxC}=5.35$ T for film 1 and 4.35 T for film 7, $\beta(B)$ increases abruptly. Here, we extracted $\beta(B)$ from the data of $R_{xx}(T)$ and $R_{xy}(T)$ at low temperatures. A larger β value implies that $R_{xy}(T)$ falls far faster than $R_{xx}(T)$ as T decreases. Thus the abrupt increase in β at high magnetic fields B ($<B_{xxC}$) signals the transition to the *unusual* insulating state where $R_{xx}(T\rightarrow 0)$ diverges and $R_{xy}(T\rightarrow 0)$ is small or zero. As B passes through B_{xxC} where dR_{xx}/dT change its sign from positive to negative, β varies from $\beta\rightarrow +\infty$ to $\beta\rightarrow -\infty$. With further increase in B, β continuously changes from negative values where $dR_{xy}/dT>0$, through zero where dR_{xy}/dT is zero, to positive values where both $R_{xx}(T\rightarrow 0)$ and $R_{xy}(T\rightarrow 0)$ diverge.

Figure 5(a) shows the R_{xxn} dependence of R_{xyC}. It is clearly seen from the figure that R_{xyC} is not *universal* but proportional to R_{xxn} except for the most resistive film 9. The values of R_{xyC} ($\sim 10\ \Omega$) are found to be significantly smaller than the values of R_{xxC} (~ 1 kΩ). Therefore, in discussing the universal resistances at the *S-I* transition[6] expressed as

$$(R_{xxC})^2+(R_{xyC})^2=(h/4e^2)^2\ , \qquad (2)$$

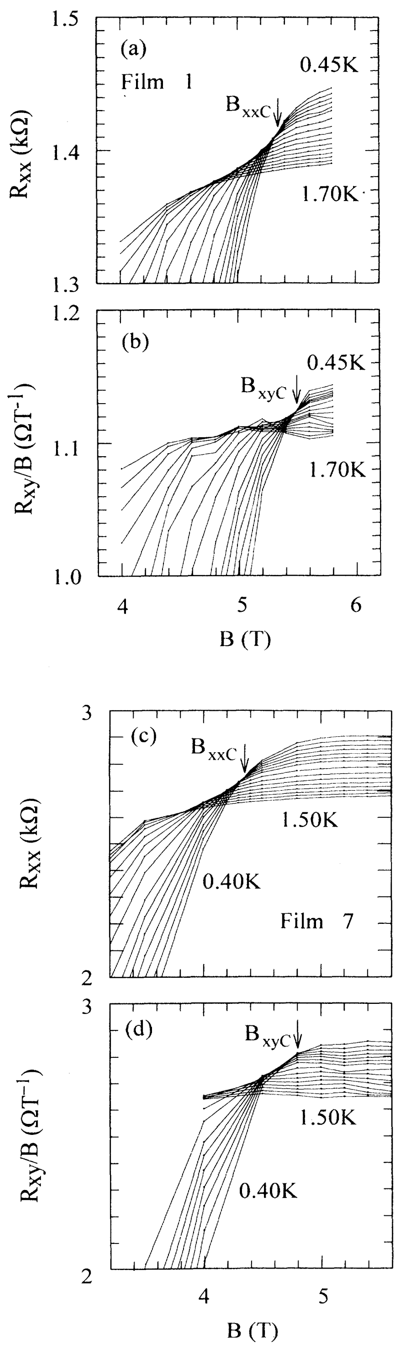

FIG. 2. Magnetic-field dependence of (a) R_{xx} and (b) R_{xy}/B for film 1, and that of (c) R_{xx} and (d) R_{xy}/B for film 7, at various temperatures. Isotherms range from 0.45 to 1.70 K for film 1 and from 0.40 to 1.50 K for film 7 in 0.05 K steps below 0.70 K and in 0.10 K steps above 0.70 K.

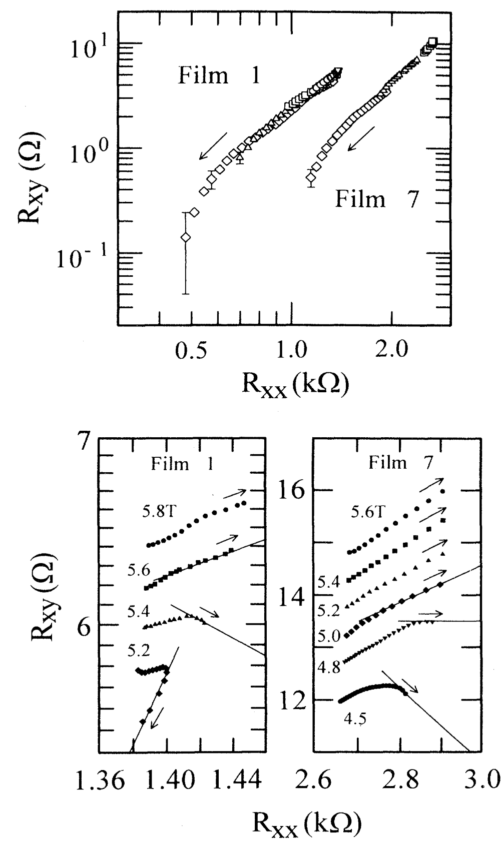

FIG. 3. The log-log plots of R_{xy} vs R_{xx} dependences at $B<B_{xxC}$ (top) and $B\gtrsim B_{xxC}$ (bottom) with T as a running variable. Arrows indicate the direction of $T\rightarrow 0$. Top: Symbols correspond to the various fields: ◇, 4.0 T; △, 4.4 T; □, 4.6 T; ○, 4.8 T; ▽, 5.0 T (film 1 with $R_{xxn}=1.3$ kΩ); ◇, 3.0 T; △, 3.5 T; □, 4.0 T (film 7 with $R_{xxn}=2.4$ kΩ). Bottom: In the vicinity of the critical field, B_{xxC} or B_{xyC}, only the data at low temperatures are used to extract the β values in Eq. (1) as indicated by the solid lines.

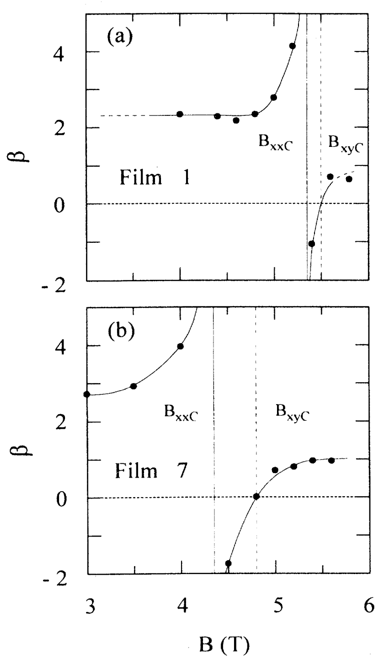

FIG. 4. Magnetic-field dependence of the exponent $\beta(B)$ in Eq. (1) for (a) film 1 and (b) film 7. Vertical solid and broken lines represent B_{xxC} and B_{xyC}, respectively. The region where $\beta(B)$ is negative ($B_{xxC} < B < B_{xyC}$) corresponds to the distinct insulating phase.

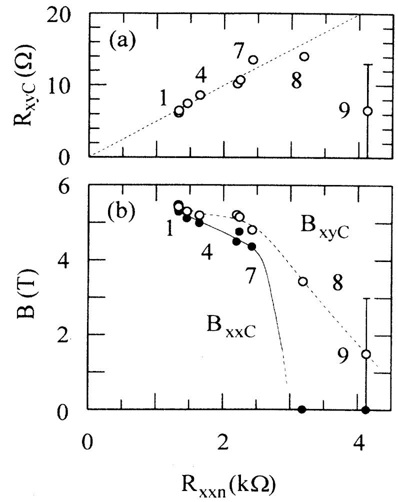

FIG. 5. (a) R_{xyC} is plotted against R_{xxn}. (b) Dependence of the critical fields B_{xxC} (solid circles) and B_{xyC} (open circles) on R_{xxn} for the nine films studied. The region $B_{xxC} < B < B_{xyC}$ corresponds to the unusual insulating phase.

we have only to consider R_{xxC}. Though we cannot exactly verify the universality of R_{xxC} from the present results, it is noteworthy that the values of R_{xxC} are close to $R_C \sim 3$ kΩ and stay in a limited range between 1.4 and 2.7 kΩ, which are in good accordance with our previous results for a more uniform film of indium prepared by quench condensation ($R_{xxC} \approx 2.2$ kΩ) and similar granular films of indium with larger grain sizes $\bar{d}$ of 22 and 28 nm ($R_{xxC} \approx 1.2$ and 1.8 kΩ, respectively).

In Fig. 5(b), B_{xxC} and B_{xyC} are plotted as a function of R_{xxn} for nine films. In addition to the usual *S-I* phase boundary (B_{xxC}) determined from the R_{xx} data, another phase boundary (B_{xyC}) determined from the R_{xy} data is present. As the authors in Ref. 13 pointed out, it is difficult to state conclusively at present whether it is the phase boundary of a true phase transition or merely a region of pronounced crossover. With increasing R_{xxn}, B_{xxC} decreases faster than B_{xyC}, and hence the region $B_{xxC} < B < B_{xyC}$ becomes broader. The existence of the unusual insulating phase is certain; however, we know currently little about it. We cannot state unambiguously from the present data if R_{xy} is exactly zero or finite at $T \rightarrow 0$ and, furthermore, the physical meaning of the phase is not very clear.

As a possible explanation, we identify this insulating phase as the $T=0$ Bose-glass insulator. The following evidence may support this interpretation. (1) We have shown from previous studies[11,12] that, as long as our system is viewed as a dirty superconductor ($\bar{d} < 20$ nm), it obeys Fisher's scaling theory which assumes the presence of the Bose-glass insulator. (2) The present results that we have shown here for nine films over a wide range of disorder are consistent with the previous results[13] for amorphous InO_x films having less disorder. It is surprising that the experimentally obtained phase diagram shown in Fig. 5(b) remarkably resembles the one [Fig. 1(a) in Ref. 13], which has been schematically illustrated based on data for InO_x films in a limited range of disorder. The remarkable similarity of the critical behaviors of the R_{xx} and R_{xy} data close to the *S-I* transition observed in different 2D systems favors the idea that the 2D field-induced *S-I* transition is universal and presumably dominated by the competition between the condensation of Cooper pairs and vortices. In order to make a definitive statement, it is essential to confirm experimentally the finite amplitude of the order parameter in the Bose-insulator phase.[17] Thus measurements of electron tunneling are now in progress.

To summarize, we have made systematic measurements of R_{xx} and R_{xy} for field-swept transitions over a wide range of disorder through the disorder-driven *S-I* transition using thin granular films of indium. We have found an unusual insulating phase where R_{xx} diverges but R_{xy} is small or zero at low temperatures. We suggest the possibility that this phase is the Bose-glass insulator.

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