Possibility of the vortex-antivortex transition temperature of a thin-film superconductor being renormalized by disorder

A. F. Hebard and G. Kotliar
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 19 September 1988)

The universal relation between the Kosterlitz-Thouless transition temperature T_c and the superfluid sheet density of thin-film superconductors with mean-field transition temperature T_{c0} results in a monotonically decreasing dependence of the ratio T_c/T_{c0} on the normal-state sheet resistance R_n. Ambiguity in the experimental definition of R_n in highly disordered thin-film superconductors is addressed by reexamining previously published data on amorphous composite In/InO_x films. Arguments are presented in favor of using the zero-temperature value of R_n, a quantity obtained by extrapolation. The dependence of T_c/T_{c0} on R_n that results from such a choice is in agreement with theory for dirty superconductors and thus suggests that additional corrections to T_c in the presence of extreme disorder are not required.

I. INTRODUCTION

It is well known that the resistive transition of a thin-film superconductor can be significantly broadened if the film is made thin enough or the sheet resistance high enough. The original proposed by Beasley, Mooij, and Orlando[1] (BMO) that such a broadening might be intrinsic and due to a two-dimensional phase transition of the Berezinskii-Kosterlitz-Thouless type[2,3] generated considerable experimental activity. This activity, which has been reviewed in a number of places,[4–6] has utilized a variety of measurement techniques in the study of spatially periodic Josephson-coupled arrays and thin-film superconductors having granular or amorphous microstructure. The majority of these experiments confirm that the basic ideas in the theory describe the behavior of thin-film superconductors. There is, however, a conspicuous absence of precision measurements of the constants and the functional dependences predicted by the theory.[6]

Simply stated, the theory predicts that fluctuations in the phase rather than the magnitude of the superfluid order parameter dominate in two dimensions. These fluctuations give rise to vortex-antivortex pairs which, at the longest length scales, unbind at the phase transition temperature T_c to produce flux-flow dissipation at zero frequency (dc). Accordingly, the temperature T_c where the resistance is zero is less than the mean-field transition temperature T_{c0}, a temperature where the resistance would have been equal to zero in the absence of vortex excitations. The central result of the BMO analysis is the prediction of a smoothly varying monotonically decreasing dependence of the ratio T_c/T_{c0} on the normal-state sheet resistance R_n, thus providing a plausible explanation for the broad resistive transitions always observed in thin films with high R_n. Experimental results showing detailed agreement with these predictions have been obtained for quenched condensed HgXe films[7] where the resistance at 4.2 K was chosen to be R_n. The rationale presented by the authors for this choice for R_n was that there was very little change in the resistance up to the higher temperatures of 10 K. This is a valid procedure for films with a metallic conductivity in the Boltzmann regime.

While R_n is weakly temperature dependent and well defined in the Boltzmann regime, where the product of the Fermi wave vector k_F and the electronic mean-free path l is greater than unity (i.e., $k_F l \gg 1$), it becomes strongly temperature dependent close to the metal-insulator transition or in the strongly localized two-dimensional regime. Amorphous-composite In/InO_x has a very large intrinsic resistivity, and the temperature dependence of the conductivity reveals it to be in the scaling region on the conducting side of the metal-insulator transition.[8] Because of the strong temperature dependence, it is not clear, *a priori*, at which temperature or scale one has to evaluate the normal-state transport coefficients which enter into the formulas of the classical theory of dirty superconductors.

This question has been addressed theoretically by Kapitulnik and Kotliar[9] and by Bulaevskii and Sadovskii.[10] The conclusion of their work is that the superconductivity introduces an energy cutoff in the problem with a magnitude on the order of T_{c0} or the energy gap Δ. The existence of this cutoff allows a finite superfluid stiffness in three-dimensional (3D) systems close to the metal-insulator transition and in two-dimensional (2D) systems which according to the scaling theory of localization have a zero-temperature conductivity $\sigma(0)$ equal to zero.

Because of the ambiguity in the experimental definition of R_n, great care must be exercised in interpreting the dependence of T_c/T_{c0} on R_n. Consider, for example, the simple thermodynamic argument reviewed in Ref. 5. In this argument, an estimate of T_c is obtained by calculating the thermodynamic free energy, $F=U-TS$, for a free vortex in a system of characteristic size R. As both the self-energy U and the entropy S have the same functional dependence on R there is a crossover temperature, identified as T_c, above which F becomes negative and it is energetically favorable for free vortices to exist. Clearly the temperature-dependent quantities, including R_n, which characterize the superconducting state must be evaluated at $T=T_c$. In films which are sufficiently thin or dirty, so that R_n is large, T_c can be significantly less than T_{c0}, thereby extending the temperature range over which an extrapolation of the normal-state properties must be made.

In this paper, we use previously published data on thin-film amorphous-composite In/InO_x films[11] to show that the low-temperature value of R_n, a quantity obtained by extrapolation gives good agreement with the BMO dependence for R_n as high as $\sim 50\ k\Omega/\square$. The result has two important implications. (1) In the limit of extreme disorder where $T_c/T_{c0}\rightarrow 0$ the BMO dependence is appropriate, and all the disorder-induced corrections to T_c enter in the strong renormalization of the sheet resistance. (2) The dependence of T_c/T_{c0} on R_n has important consequences in that superconductivity will disappear when, with decreasing temperature, R_n starts to cross over into the 2D regime. In the following sections, we will first review the theoretical arguments used to obtain the BMO dependence of T_c/T_{c0} on R_n (Sec. II) and then present experimental data showing that the use of a zero-temperature estimate of R_n gives good agreement with the BMO dependence (Sec. III). The necessity of using an extrapolative procedure to determine normal-state properties will be shown to be a natural consequence of electronic transport in a unique superconducting system which is situated close to the metal-insulator transition and which obeys scaling theory with temperature as the scaling parameter.

II. A PERSPECTIVE ON THE DEPENDENCE OF T_c/T_{c0} ON R_n

The starting point for the Berezinskii-Kosterlitz-Thouless theory[2,3] as applied to charged superfluids is the universal relation

$$k_B T_c = \pi\hbar^2 n_s^*(T_c)/2m^* \tag{1}$$

between T_c, the renormalized areal superfluid density n^* and the mass of the elementary superfluid particle m^*, equal to twice the mass of the electron. Equation (1) can be cast into a more convenient form by using the London equation definition of the two-dimensional magnetic screening length

$$\Lambda = 2\lambda^2/d = \Lambda = m^*c^2/2\pi e^{*2} n_s^* \tag{2}$$

for a film with thickness d and bulk screening length λ to obtain the relation

$$\epsilon(T_c)k_B T_c = \left[\frac{\Phi_0}{4\pi}\right]^2 \frac{1}{\Lambda(T_c)} , \tag{3}$$

where $\Phi_0 = hc/2e$ is the quantum of flux. The effect of renormalization near T_c has been taken into account by the inclusion of the vortex dielectric constant ϵ_v, a quantity predicted by theory[12] and determined by experiment[7,13] to be close to unity.

The experimental challenge in ascertaining the validity of Eq. (3) consists of two parts. First, a self-consistent procedure for determining T_c must be obtained and second, the value of Λ must be measured at this temperature. Measurements of the screening currents induced by an ac magnetic field in amorphous composite In/InO_x films have shown an observed discontinuity in the temperature dependence of Λ^{-1} to be consistent with Eq. (3).[13] In all experiments to date, however, there is some uncertainty in the determination of T_c owing to the frequency- or current-dependent length scales set by the measurement technique and which arise because of the scale-dependent vortex interactions.

Using the dirty-limit formula which relates Λ to the normal-state sheet resistance R_n, BMO showed that Eq. (3) can be written in the form

$$\frac{T_c}{T_{c0}} f^{-1}\left[\frac{T_c}{T_{c0}}\right] = 2.18\frac{\hbar}{e^2\epsilon_v R_n} , \tag{4}$$

where the temperature-dependent factor f has the form

$$f\left[\frac{T}{T_{c0}}\right] = \frac{\Delta(T)}{\Delta(0)}\tanh\left[\frac{0.88\Delta(T)T_{c0}}{\Delta(0)T}\right] . \tag{5}$$

Equation (4) is slightly different from the original BMO expression as it includes the quantity ϵ_v, a near-unity multiplicative factor appearing in combination with R_n, to account for the distinction between renormalized and unrenormalized quantities.[7] The solid line of Fig. 1 shows the $\epsilon_v=1$ solution of Eq. (4). It is important to note the smooth dependence of T_c/T_{c0} on R_n which in the limit $T_c/T_{c0}\rightarrow 0$ has the dependence

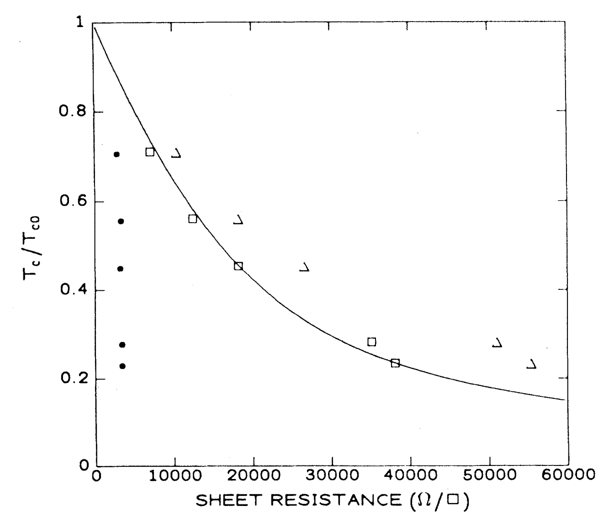

FIG. 1. Plot of T_c/T_{c0} as a function of R_n for five 100-Å-thick In/InO_x films showing better agreement with theory (solid line) when $R_n(0)$ (open symbols) rather than $R_n(295)$ (closed circles) is used. The squares denote extrapolations based on the temperature-dependent conductivity of a 600-Å-thick annealed film, and the triangles result from a similar analysis of as-deposited films.

$$T_c/T_{c0}=2.18\hbar/\epsilon_v e^2 R_n \ . \quad (6)$$

Also, for a given value of R_n, there is increased temperature broadening of the transition if $\epsilon_v>1$. Interestingly, Eq. (4) can also be derived for a weakly coupled Josephson square array in which the critical current I_c between gains in the array is related to the tunnel junction resistance R_n by the expression

$$I_c=\frac{\pi\Delta(0)}{2eR_n}f\left[\frac{T}{T_{c0}}\right], \quad (7)$$

where $f(T/T_{c0})$ is the same temperature-dependent factor appearing in Eq. (4).[14]

In 3D the scaling relation form for $\sigma(T)$ mentioned in Sec. I takes the form

$$\sigma(T)=\frac{e^2}{\hbar}\left[\frac{1}{\xi}+\frac{A}{l_{\text{in}}(T)}\right]=\sigma(0)+\alpha T^{1/4} \quad (8)$$

with A an unknown constant. The zero-temperature conductivity $\sigma(0)=e^2/\hbar\xi$ is proportional to the reciprocal of the localization length ξ and the inelastic electron scattering length l_{in} has a power-law temperature dependence with nonuniversal exponent. The constant α has been determined from an observed $T^{1/4}$ dependence of $\sigma(T)$ for a single 600-Å-thick film at nine different stages of annealing[8] to have a value $\alpha=41.4\pm1.8\,\Omega^{-1}\,\text{cm}^{-1}\,\text{K}^{-1/4}$. Equation (8) is valid as long as the films are in the three-dimensional regime where either $l_{\text{in}}(T)<d$ or $\xi<d$. Assuming that $\xi>d$, when $l_{\text{in}}(T)\cong d$ one expects on theoretical grounds,[15] as suggested experimentally[16] for crystalline In_2O_3, that $R(T)$ will grow exponentially with decreasing temperature. A possible explanation for the rapid disappearance of T_c relies on Eq. (6) and the fact that at some crossover temperature scale T_x there is a crossover sheet resistance R_x where $R_x=R(T_x)$. For $T<T_x$, R increases rapidly with a concomitant quenching of superconductivity at $R_n(T_c)=R(T_x)$.

III. RESISTANCE BROADENING IN In/InO$_x$ FILMS

With the simplifying assumption that $\epsilon_v=1$, experimental verification of Eq. (4) requires the determination of three parameters: T_c, T_{c0}, and R_n. In Ref. 11 detailed measurements of T_c, T_{c0}, and the room-temperature sheet resistance $R(295)$ were presented for five different 100-Å-thick films. Nonlinear current-voltage measurements combined with magnetoresistance data were used to determine T_c and Aslamazov-Larkin theory fits to the zero-field resistive transitions were used to determine T_{c0}. The resulting dependence of the ratio T_c/T_{c0} on the measured room-temperature sheet resistance is shown as solid circles in Fig. 1. Significantly, however, the data falls well below the expected theoretical dependence of Eq. (4) ($\epsilon_v=1$) shown by the solid line.

As T_c is on the order of 20% of T_{c0} for the most disordered film of Fig. 1, it is reasonable to expect that the discrepancy might be due to an incorrect choice of R_n as discussed above. To test the expectation that $R(0)$ is a better measure of the normal-state properties at $T=T_c$, we utilize previously published electric-field-effect data in which the disorder parameter $k_F l$ of a 600-Å-thick film was varied by thermal annealing.[8] After each stage of annealing, extrapolation of the temperature-dependent conductivity $\sigma(T)$ was used to determine the zero-temperature conductivity $\sigma(0)$. The measured dependences of $\sigma(0)/\sigma(295)$ and T_{c0} on $(k_F l)^{-2}$ shown in Ref. 8 are replotted in Fig. 2 as a dependence of $\sigma(0)/\sigma(295)$ on T_{c0}. The data clearly show a trend in which $\sigma(0)$ and T_{c0} simultaneously approach zero. Said in another way, superconductivity in the In/InO$_x$ system disappears at or very near the metal-insulating transition where $\sigma(0)=0$.

At present, we are unware of a theoretical justification for the linear dependence of $\sigma(0)/\sigma(295)$ on T_{c0} shown in Fig. 2. This dependence, however, is confirmed by a similar plot, shown in Fig. 3, for 23 different as-deposited films with thicknesses indicated in the legend. These data were obtained by using the room-temperature $\sigma(295)$ and liquid-nitrogen-temperature $\sigma(77)$ values of $\sigma(T)$ to calculate [cf. Eq. (8)] values for $\sigma(0)$ and α for each film. The mean value of α is found to be $54.9\pm6.1\ \text{cm}^{-1}\,\text{K}^{-1/4}$, approximately 30% larger than the value found for the annealed film.

A direct consequence of the scaling theory prediction of Eq. (8) is a linear relationship between $\sigma(0)$ and $\sigma(295)$. The linear dependence, shown in Fig. 4 for the as-deposited films, confirms this behavior and illustrates graphically the location of the critical room-temperature conductivity σ_c which delineates the boundary between metallic and insulating behavior. The $\sigma(0)=0$ intercept of Fig. 4 gives $\sigma_c=188.5\ \Omega^{-1}\,\text{cm}^{-1}$ which is very close to the value $\sigma_c=172.9\ \Omega^{-1}\,\text{cm}^{-1}$ obtained from a similar plot (not shown) for the annealed-film data. The overlap of the data of Fig. 4 for films of different thickness also illustrates that the inelastic scattering processes, as mani-

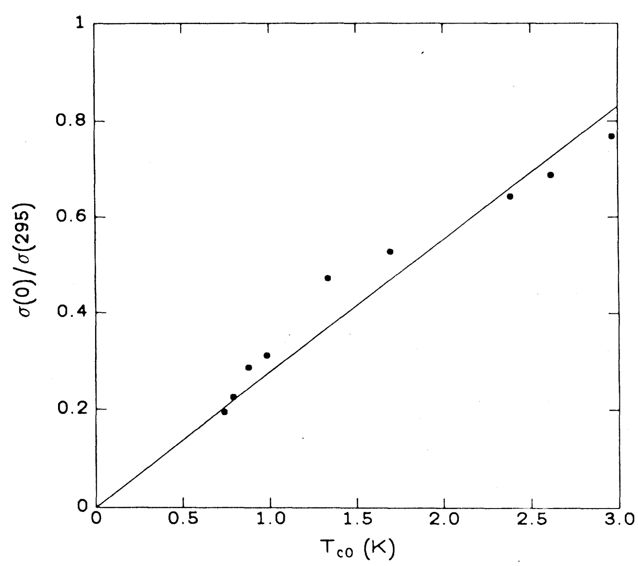

FIG. 2. Plot of $\sigma(0)/\sigma(295)$ vs T_{c0} for a single 600-Å-thick film annealed to selected stages of disorder. The solid line is a regression fit discussed in the text.

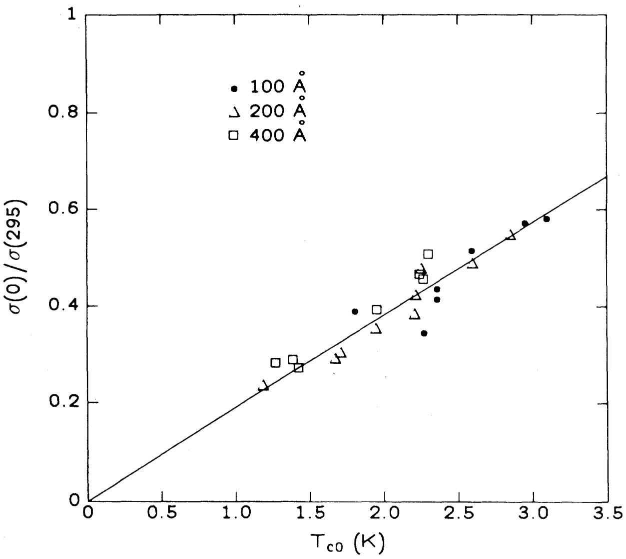

FIG. 3. Plot of $\sigma(0)/\sigma(295)$ vs T_{c0} for as-deposited films with 100 Å (●), 200 Å (△), and 300 Å (□) thicknesses. The solid line is a regression fit discussed in the text.

fested in the constant α of Eq. (8), are independent of film thickness for films as thin as 100 Å. Together with the notion that inelastic processes are responsible for the pair breaking which suppresses T_{c0}, this observation is consistent with previous work[17] demonstrating that for amorphous composite In/InO_x films with $d \geq 100$ Å the disorder-induced suppression of T_{c0} results from a three-dimensional mechanism depending on film *resistivity*

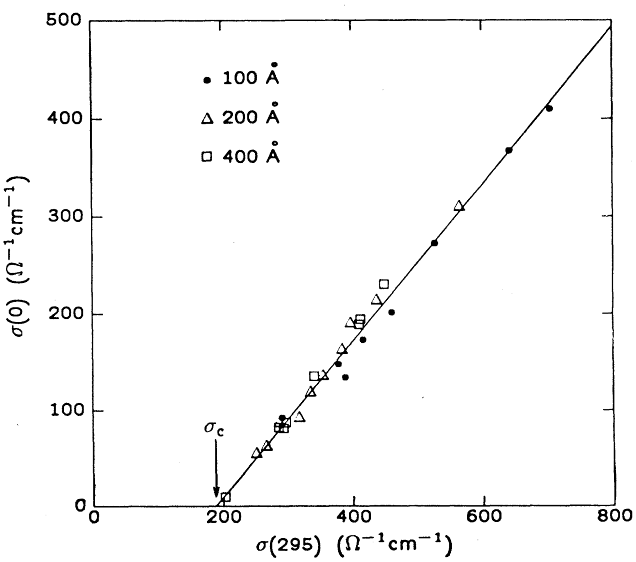

FIG. 4. Plot of $\sigma(0)$ vs $\sigma(295)$ for as-deposited films with 100 Å (●), 200 Å (△), and 300 Å (□) thicknesses. The solid line regression fit intercepts the $\sigma(0)=0$ ordinate at a critical conductivity σ_c depicted by the arrow.

rather than film *sheet resistance* as it would for two-dimensional Josephson-coupled arrays.

Since $\sigma(T)$ is expected to have the functional dependence of Eq. (8), at the present level of accuracy it does not make much difference whether we extrapolate to T_{c0}, T_c, or to $T=0$, as long as one remains in the three-dimensional regime. If one is in the two-dimensional regime, our procedure underestimates the zero-temperature resistance.

A simplifying assumption which enables us to utilize the data shown in Figs. 2 and 3 to estimate $\sigma(0)$ for the five films under consideration in Fig. 1 is to use a linear regression fit with an intercept constrained to pass through the origin. This linear-relationship, shown by the solid lines in Figs. 1 and 2, has the form $\sigma(0)/\sigma(295)=0.277T_{c0}$ for the annealed film and $\sigma(0)/\sigma(295)=0.191T_{c0}$ for the as-deposited films, hence enabling a calculation of $\sigma(0)$ as a function of T_{c0} and $\sigma(295)$. The difference, mentioned previously, between the values of α for the annealed film and the as-deposited films is reflected in the 30% smaller slope of the solid-line regression fit of Fig. 3 when compared to the fit of Fig. 2. Calculation of the zero-temperature normal-state sheet resistance $R_n(0)=1/\sigma(0)d$ for the five 100-Å-thick films shown in Fig. 1 using interpolations on these data is justified only if T_{c0} is independent of d as has been shown experimentally.[17]

The open symbols of Fig. 1 show the dependence of T_c/T_{c0} on $R_n(0)$, where $R_n(0)$ is calculated for the annealed film (squares) by using interpolation on the linear dependence of Fig. 2 and for the as-deposited films (triangles) by using interpolation on the linear dependence of Fig. 3. The agreement with the $\epsilon_v=1$ solution to Eq. (4) is satisfactory. This agreement implies that, at least for homogeneous amorphous films, the predicted relationship between T_c/T_{c0} and R_n holds for T_c as low as $0.2T_{c0}$. Accordingly, additional corrections to T_c which are not contained in $R_n(T_c)$ in the presence of extreme disorder are unnecessary.

IV. CONCLUSIONS

For In/InO_x films with $d=100$ Å the disorder-induced suppression of T_{c0} is clearly 3D, with a dependence on film resistivity rather than sheet resistance.[17] The temperature T_c at which all resistance disappears marks a 2D phase transition mediated by the presence of 2D vortex excitations. We have shown in this study that the BMO equation, which simply relates the ratio T_c/T_{c0} to R_n, is satisfied providing R_n is evaluated near T_c. Use of "normal-state" values above T_{c0} have been shown to be inappropriate because of the strong temperature-dependent normal-state properties dictated by the proximity of In/InO_x to the metal-insulator transition. There is consequently no need to include additional corrections to the BMO formula in the presence of extreme disorder.

Estimates of $R_n(T_c)$ have been made within the context of a scaling theory description of the behavior of $\sigma(T)$. This analysis has shown a remarkable (and unexplained) linear dependence of $\sigma(0)/\sigma(295)$ on T_{c0}. In addition, the linear scaling theory dependence of $\sigma(0)$ on

$\sigma(295)$ gives a value $\sigma_c = 180\ \Omega^{-1}\,\mathrm{cm}^{-1}$ for the critical room-temperature conductivity which delineates the boundary of the metal-insulating transition. The simultaneous disappearance of superconductivity (cf. Figs. 2 and 3) and metallic behavior (cf. Fig. 4) at this boundary has to our knowledge only been demonstrated for the In/InO_x system. Interestingly, for $d = 100$ Å, the critical value for $R_n(295)$ which defines the crossover from superconducting to insulating behavior is $1/\sigma_c d = 5560\Omega/\Box$, a value accidentally close to $\hbar/e^2$ noted previously.[11] For In/InO_x films, superconductivity is thus *rapidly* quenched near a critical high-temperature $(T > T_{c0})$ value of normal-state *resistivity* rather than *sheet resistance* as is found for Josephson-coupled granular Sn films.[18] When measured with respect to $R_n(T_c)$, however, T_{c0}, T_c, and T_c/T_{c0} show a smooth monotonically decreasing dependence with no evidence of a threshold near $\hbar/e^2$.

As a final remark, we note that flux-flow measurements made at $T = T_c$ give an enhanced vortex mobility which increases dramatically as T_c decreases.[11] Interpreted within the context of the Bardeen-Stephen model, this enhanced mobility is evidence for a normal-state core which is rapidly taking on the characteristics of an insulator as the film becomes more disordered. This behavior reflects proximity to insulating behavior as the disorder increases and the temperature is reduced.

ACKNOWLEDGMENT

The authors would like to acknowledge useful discussions with A. T. Fiory and M. A. Paalanen.

[1] M. R. Beasley, J. E. Mooij, and T. P. Orlando, Phys. Rev. Lett. **42**, 1165 (1979).
[2] V. L. Berezinskii, Zh. Eksp. Teor. Fiz. **61**, 1144 (1971) [Sov. Phys.—JETP **34**, 610 (1971)].
[3] J. M. Kosterlitz and D. J. Thouless, J. Phys. C **6**, 1181 (1973).
[4] A. F. Hebard and A. T. Fiory, Physica **109&110B**, 1637 (1982).
[5] J. E. Mooij, in *Advances in Superconductivity,* Vol. 100 of *NATO Advanced Study Institute, Series B: Physics,* edited by B. Deaver and J. Ruvalds (Plenum, New York, 1983).
[6] J. E. Mooij, in *Percolation, Localization and Superconductivity,* Vol. 109 of NATO Advanced Study Institute, Series B: Physics, edited by A. M. Goldman and S. A. Wolf (Plenum, New York, 1984).
[7] K. Epstein, A. M. Goldman, and A. M. Kadin, Phys. Rev. **26**, 3950 (1982).
[8] A. T. Fiory and A. F. Hebard, Phys. Rev. Lett. **52**, 2057 (1984).
[9] A. Kapitulnik and G. Kotliar, Phys. Rev. Lett. **54**, 473 (1985); G. Kotliar and A. Kapitulnik, Phys. Rev. B **33**, 3146 (1986).
[10] L. N. Bulaevskii and M. V. Sadovskii, JETP Lett. **39**, 641 (1984).
[11] A. F. Hebard and M. A. Paalanen, Phys. Rev. Lett. **54**, 2155 (1985), and references therein.
[12] D. R. Nelson and J. M. Kosterlitz, Phys. Rev. Lett. **39**, 1201 (1977).
[13] A. T. Fiory, A. F. Hebard, and W. I. Glaberson, Phys. Rev. B **28**, 5075 (1983).
[14] A. F. Hebard, in *Inhomogeneous Superconductors (Berkeley Springs, WV, 1979),* Proceedings of the Conference on Inhomogeneous Superconductors, AIP Conf. Proc. No. 58, edited by D. U. Gubser, T. L. Francavilla, J. R. Leibowitz, and S. A. Wolf (AIP, New York, 1979), p. 129.
[15] E. Abrahams, P. W. Anderson, D. C. Liccardello, and J. V. Ramakrishnan, Phys. Rev. Lett. **42**, 673 (1979).
[16] Y. Imry and Z. Ovadyahu, J. Phys. C **15**, L327 (1982).
[17] A. F. Hebard and A. T. Fiory, Phys. Rev. Lett. **38**, 1131 (1987).
[18] B. G. Orr, H. M. Jaeger, A. M. Goldman, and C. G. Kuper, Phys. Rev. Lett. **56**, 378 (1986).