Fizika Nizkikh Temperatur, 2000, v. 26, No 12, p. 1191–1196

Nature of critical current and coherent phenomena in granular MoN_x thin films

V. G. Prokhorov and G. G. Kaminsky

Institute for Metal Physics, 36 Vernadsky Blvd., 03680 Kiev, Ukraine
E-mail: pug@imp.kiev.ua

Y. P. Lee

Honyang University, Seoul, 133-791, Korea

I. I. Kravchenko

Department of Physics, University of Florida, Gainesville, FL 32611, USA

Received March 27, 2000, revised May 11, 2000

Investigations of the critical current versus temperature and applied magnetic field were carried out for granular MoN_x films. All sampels display a two-stage superconducting transition and can be treated as a percolating network of *SNS* contacts with a Josephson coupling between grains. The temperature behavior of the critical current for the films studied is the same as the $I_c(T)$ dependence for a *SNS* junction in the diffusive limit. The value of critical current in a magnetic field is governed by the pinning of Josephson vortices.

PACS: **74.50.+r**, 74.76.Db, **74.80.–g**

1. Introduction

The study of superconducting properties of nonuniform high-resistance films has attracted a great deal of attention recently due to the discovery of the superconductor–insulator transitional in two-dimensional (2*D*) systems [1–3]. However, another remarkable feature of these materials is connected with the possibility of forming a regular granular structure. The interplay between microstructural disorder and space modulation of the superconducting order parameter allows us to treat them as a network of Josephson junctions that are very similar in Bi- and Tl-based anisotropic high-T_c superconductors [4]. The granular nature of these objects clearly plays a major role in their superconductivity, and the understanding of the behavior of spatially nonuniform conventional superconductors is essential for establishing which properties of the cuprates might follow simply from their geometry and microstructural inhomogeneity. Even though the superconducting properties of the granular films have been studied for a long time, the physical nature of this effect continues to be a matter of controversy.

In this paper we propose a new method for the preparation of granular MoN_x films, which can be treated as a percolating network of the superconductor–normal metal–superconductor (*SNS*) contacts with a Josephson coupling between grains. The main peculiarity of the studied films is the occurrence of a two-stage transition from the normal to the long-range superconducting (coherent) state. It is shown that the observed linear dependence of the transition temperature to the phase-coherent state on the residual resistance can be explained in the framework of the proximity-effect model for the superconductor–normal metal (*SN*) system. The temperature behavior of the critical current I_c for the films studied is very similar to the $I_c(T)$ dependence obtained for a *SNS* junction in the diffusive limit. The microscopic superconducting parameters estimated from fitting the experimental $I_c(T)$ curves coincide with ones obtained by independent methods. In contrast with $I_c(T)$, the magnetic field dependence of the critical current cannot be described on the basis of the theoretical models developed for single or multiple *SNS* junctions. We suggest that the transition of the MoN_x granular films to the resistive state under the action of current in an applied magnetic field results from the start of the flux lines' motion rather than from the switching of the Josephson junctions. Therefore the value of the critical current is determined by the pinning force of the Josephson vortices owing to the magnetic interaction with the grains.

2. Experimental procedure

The MoN_x films were prepared on Al_2O_3 substrates by thermal evaporation in a vacuum chamber with a base pressure of $6.7 \cdot 10^{-4}$ Pa. A Mo strip (0.25×2×85 mm) was used for the evaporation. The deposition of the films was carried out with the following technological parameters. The current was about 50 A, the output power was ~ 50 W, the pressure of nitrogen was around 10^{-3} Pa, and the temperature of substrate during the deposition was ~ 350 K.

The deposited films have a thickness $d \simeq 200$ nm. An x-ray diffraction analysis showed that a hexagonal MoN phase [5] ($a = b \simeq (0.572 \pm 0.001)$ nm, $c \simeq (0.560 \pm 0.005)$ nm) is formed in about 90% of the film volume. On the other hand, the low value of the critical temperature T_c of 5.0–6.5 K shows that the prepared films have a non-stoichiometric concentration of nitrogen and their composition is represented as MoN_x . The average grain size estimated by high-resolution electron microscopy turned out to be $\langle D \rangle \approx 100$ nm. The geometric dimensions of the samples were set by a photolitographic method to have a width of 0.325 mm and a length between voltage contacts of 3.65 mm. The resistance was measured by the conventional dc four-probe techniques. A voltage drop of 1 μV/cm served as the criterion for choosing the critical current. The applied magnetic field was always maintained perpendicular to the film surface and to the direction of the transport current.

3. Results and discussion

Figure 1 shows the resistance curves of the superconducting transition for MoN_x films with different values of the residual resistivity ρ_0 measured at 10 K. It can be seen that all the films with $\rho_0 > 6.2$ μΩ·m have a two-stepped shape of $R(T)/R_0$, where R_0 is the resistance before the superconducting transition. Therefore, the transition of the films to the superconducting state is carried out in two stages. The first superconducting state occurs at a high temperature, T_{c0} , inside the grains, and a long-range phase-coherent superconducting state is formed at the lower temperature, T_c . Unfortunately, we have not observed any rela-

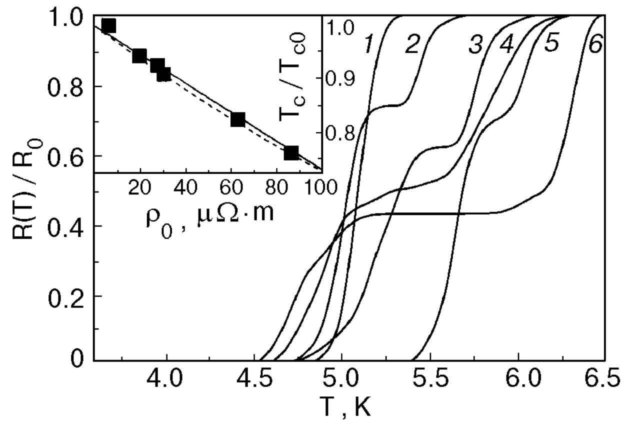

Fig. 1. Superconducting transition curves for MoN_x films with different values of the residual resistivity ρ_0 , μΩ·m: 6.2 (*1*); 30.2 (*2*); 19.8 (*3*); 62.4 (*4*); 27.5 (*5*); 86.1 (*6*). The inset shows the dependence of T_c/T_{c0} ratio on ρ_0 . Solid and dashed lines are theoretical curves obtained by using Eqs. (1) and (3), respectively.

tion between the value of the residual resistance (or sheet resistance, $R_\square = \rho_0/d$) and the temperature positions of the two superconducting transitions, which is very often found in disordered thin films [6,7]. However, the difference between the first and second resistance steps (T_{c0} and T_c) depends directly on the total resistivity of the film.

The disorder-induced depression of T_{c0} is in good qualitative agreement with the percolation description of a weakly coupled random 2D network of Josephson junctions. Using the theoretical results for the percolating network of Josephson-coupled grains, together with the temperature-dependent BCS energy gap, one can write [7]

$$\frac{T_c}{T_{c0}} = 1 - \frac{0.3e^2\rho_0}{\pi\hbar L} , \qquad (1)$$

where T_{c0} is the superconducting transition temperature of the grain and T_c is the depressed transition temperature of the grain boundary as mentioned above; L is the characteristic length of the microstructure morphology (proportional to the grain size), and $\pi\hbar/4e^2 = 6.45$ kΩ is the quantum of resistance. A better agreement between experimental and theoretical data is observed by fitting with $L \simeq 25$ nm, as is seen in the inset of Fig. 1. If it is assumed, according to Ref. 7, that $L \simeq 0.3\langle D \rangle$, where $\langle D \rangle$ is the average grain size in the film, $\langle D \rangle$ turns out to be about 83 nm, which is of the same order of magnitude as the data obtained by electron-microscopic analysis. The observed linear dependence of the ratio T_c/T_{c0} on the residual resistance suggests that our samples are very similar to a composite In–InO_x film with an island microstructure [7].

On the other hand, one can obtain a similar linear-like behavior of T_c/T_{c0} versus resistivity from Kresin's theoretical model [8] of the proximity effect for strongly coupled superconductors. Let us consider the film as a *SNS* proximity sandwich system, where S is the grains and N is the grain boundaries. If the thickness of the normal metal (the width of the grain boundaries) is smaller than the coherence length for the boundaries, the superconducting critical temperature for the whole *SNS* system can be described by [9]

$$T_c = T_{c0}(\pi T_{c0}/2\gamma u)^{\alpha}\,, \qquad (2)$$

where $\alpha = n_N\, L_N\, /n_S\, L_S$, n_N and n_S are the electron density of states of the normal and superconducting layers, and L_N and L_S are the layer thicknesses, respectively. For a granular film L_N corresponds to the width w of the grain boundary, as mentioned, and L_S to the average grain size $\langle D\rangle$. In case of a nearly ideal *SN* contact, $u \simeq \omega_c$, where ω_c is the average phonon frequency (equal to the Debye frequency in this model). $\gamma = 0.577$ is Euler's constant. Taking into account that the resistivity is inversely proportional to the electron density of states, $\rho = 1/(e\mathcal{D}n)$ where $\mathcal{D} = v_F^2\,\tau/3$ is the electron diffusion coefficient (v_F is the Fermi velocity and τ is the mean free time between collisions of electrons), Eq. (2) can be simplified to

$$\frac{T_c}{T_{c0}} \approx 1 + \frac{\rho_{0S}}{\rho_{0N}}\,\frac{w}{\langle D\rangle}\,\ln\left(\frac{2.72\;T_{c0}}{\omega_c}\right), \qquad (3)$$

where ρ_{0S} is the resistivity of the grains, and ρ_{0N} is that of the grain boundaries.

This equation is formally equivalent to Eq. (1), since the logarithmic term in Eq. (3) has the negative sign. The value of the electron diffusion coefficient is little different between grain and grain boundary a covalent compound of the type considered, and $\ln x \approx x - 1$ when $x << 1$. If we also assume that the minimum metallic conductivity of the grain boundary is realized as $\sigma_{\min} \simeq 0.03e^2/\hbar a$ according to Mott and Koveh [10], we can estimate the value of $\rho_{0N} = \sigma_{\min}^{-1} \simeq 156\ \mu\Omega\cdot$m, where a is the crystal lattice constant. With $\omega_c \approx 350$ K and $T_{c0} \simeq 5$ K for our films [11], the ratio of the grain boundary width to the average grain size, $w/\langle D\rangle$ is estimated to be about 0.1. Therefore, the average width of the grain boundaries turns out to be 10 nm in our case. This is an absolutely reasonable value for the films deposited at a low temperature. Consequently, the prepared MoN_x thin films can be treated as a percolating network of *SNS* contacts with a Josephson coupling between grains.

3.1. Temperature dependence of the critical current

Figure 2 shows the temperature dependence of the critical current for the MoN_x film with $\rho_0 = 30.2\ \mu\Omega\cdot$m at different values of applied magnetic field. The $I_c(T)$ behavior displays a fundamental deviation from the Ambegaokar–Baratoff (AB) expression developed for the case of the ideal superconductor–insulator–superconductor (*SIS*) Josephson junction, $I_c(T)R_N = \pi\Delta(T)/2 \times \times \tanh(\Delta(T)/2T)$ [12]. R_N is the normal resistance of the junction and $\Delta(T)$ is the superconducting energy gap. A better agreement between the experimental data and the AB model can be obtained by using a severely depressed value of the superconducting energy gap as a fitting parameter. The dashed line in the inset of Fig. 2 is plotted with a value of $\Delta(0) \simeq 1.76k_B\,T_c$, while the solid line requires $\Delta(0) \simeq 0.12k_B\,T_c$. It is worthy of note that the decrease in the energy gap of the Josephson junctions has already been observed experimentally.

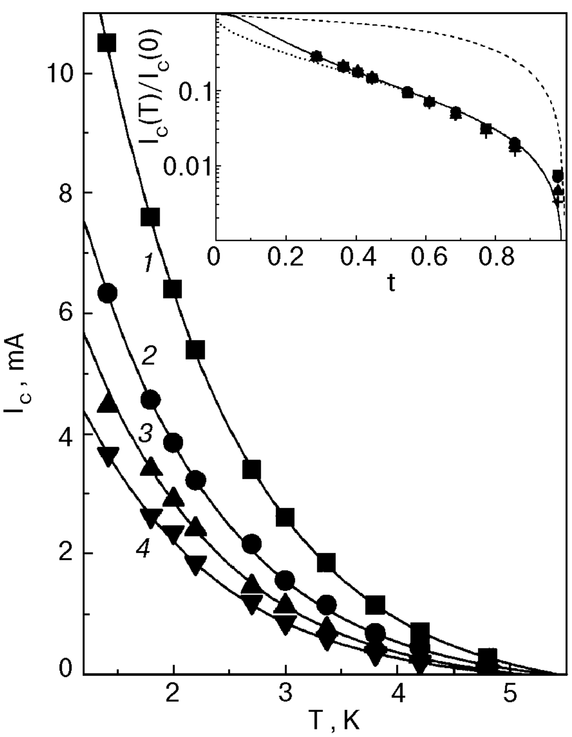

Fig. 2. Temperature dependence of the critical current for a MoN_x film with $\rho_0 = 30.2\ \mu\Omega\cdot$m at different values of applied magnetic field, T: 0 (*1*); 0.1 (*2*); 0.2 (*3*); 0.3 (*4*). The inset shows the logarithmic dependence of $I_c(T)/I_c(0)$ on the reduced temperature $t = T/T_c$. The dashed line corresponds to the Ambegaokar–Baratoff expression, the solid line is calculated for the model of a *SIS* junction with a suppressed superconducting energy gap, and the dotted line corresponds to the model of a *SNS* junction in the diffusive limit.

For example, the energy gap for the high-T_c layered superconductors obtained from measurements of the intrinsic Josephson effect [13] is about a factor of two smaller than that determined by the tunneling [14] or spectroscopy [15] measurements. However, even though such an energy gap supression is predicted in the theoretical works [16,17] this problem is far from being totally understood. On the other hand, the small value of the energy gap suggests that the transition temperature for the phase-coherent superconducting state (the second step on the resistance curves in Fig. 1) is lower than T_{c0}, an effect which has not been observed experimentally. We are proposing that the strong deviation of the experimental data from the AB expression in our case results from another physical reason.

Taking into account the good agreement between the experimental dependence of T_c/T_{c0} on ρ_0 and Kresin's model [9] for the *SN* proximity sandwich, we expect that the temperature behavior of the critical current of the films can be described in the framework of the approaches for *SNS*-type Josephson junctions [18–20]. The critical current is written as

$$I_c(T) = AI_c(0)\exp\left(-\frac{L_N}{\xi_N}\right), \quad (4)$$

where $I_c(0)$ is the critical current at zero temperature, and $A \approx 12/\pi$ for $T << T_c$ and $A \approx 1 - T/T_c$ at $T \to T_c$. For the normal layer in a *SNS* system, ξ_0 is replaced by the normal metal coherence length, $\xi_N = \hbar v_F/2\pi k_B T$ (in the ballistic limit $L_N << l$, where l is the impurity scattering length) or $\sqrt{\hbar\mathcal{D}/2\pi k_B T}$ (in the diffusive limit $L_N >> l$). Because the resistance of the grain boundary is larger than that of the grain itself, it is reasonable to assume the diffusive limit for our case. Using the well-known expression for the superconductor coherence length, $\xi(0) \approx (\xi_0 l)^{1/2}$, where $\xi_0 = 0.18\hbar v_F/k_B T_c$, one can write Eq. (4) in a form that is more convenient for fitting the experimental data:

$$\frac{I_c(T)}{I_c(0)} \simeq \left(1 - \frac{T}{T_c}\right)\exp\left(-\frac{0.57L_N}{\xi(0)}\left(\frac{T}{T_c}\right)^{1/2}\right). \quad (5)$$

It is shown in the inset of Fig. 2 that a better agreement with the experimental data is provided with $L_N/\xi(0) \simeq 2.5$. In our case $L_N = w$, the width of grain boundary. Therefore, the value of the coherence length estimated from the temperature dependence of the critical current is equal to $\xi(0) \simeq 4$ nm. This is in the same order of magnitude as the value calculated from the expression for upper critical magnetic field, $H_{c2}(0) = \phi_0/2\pi\xi^2(0) \simeq 0.69(dH_{c2}/dT|_{T_c})T_c$, where ϕ_0 is the magnetic flux quantum ($2.07\cdot10^{-15}$ T·m^2). The measured value of $dH_{c2}/dT|_{T_c}$ for our films was $\simeq 3$–3.5 T/K, leading to $\xi(0) \simeq 6$–7 nm.

3.2. Magnetic-field dependence of the critical current

It is well known that in a *SIS* system, the Josephson current as a function of magnetic field shows a Fraunhofer pattern [21]. However, in the case of a Josephson medium consisting of *SNS* junctions in which N is a $2D$ normal metal on the mesoscopic scale, the dependence of the Josephson current on magnetic field is expected to be not so simple. The Fraunhofer oscillations of the critical current have not been observed even in the stacked intrinsic Josephson junctions of $Bi_2Sr_2CaCu_2O_{8+\delta}$ layered high-T_c single crystals [4,22]. Figure 3 shows the $I_c(B)$ dependence for the MoN_x film measured at different temperatures, which does not display any manifestation of oscillatory behavior.

Two theoretical approaches have been used for the explanation of the magnetic field dependence of the critical current in spatially nonuniform superconductors included in a system of the Josephson junctions. The first of them is based on the solution

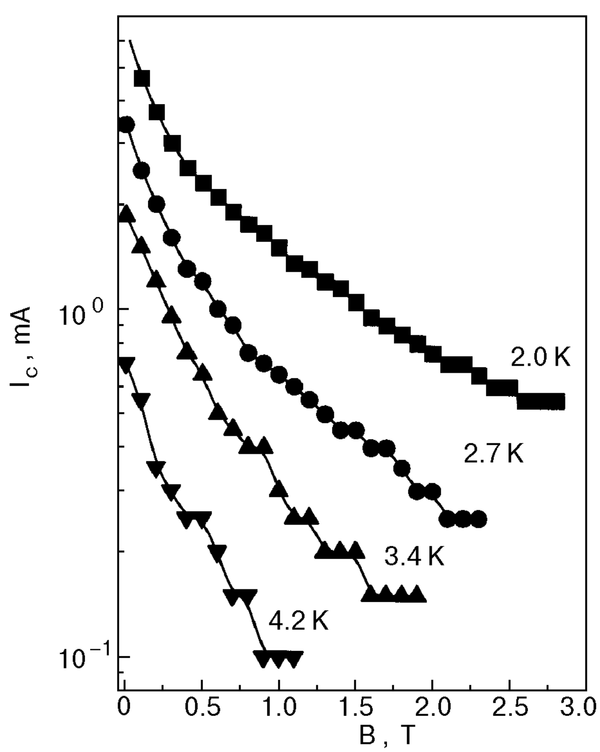

Fig. 3. Dependence of the critical current on applied magnetic field for a MoN_x film with $\rho_0 = 30.2$ μΩ·m at different temperatures.

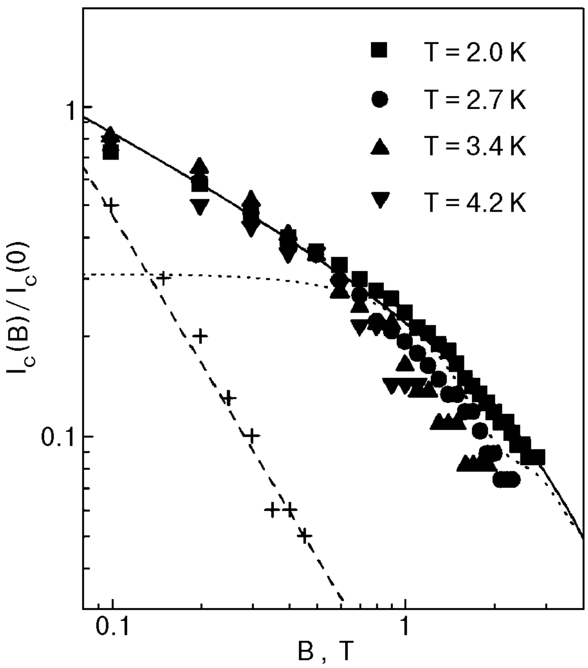

Fig. 4. Dependence of $I_c(B)/I_c(0)$ on applied magnetic field for a MoN_x film with $\rho_0 = 30.2$ μΩ·m. The solid line is the theoretical curve based on the model of pinning of the Josephson vortices, the dotted line corresponds to the *XY* model, and the dashed line indicates the dependence $I_c \sim 1/B^{1.5}$. The cross symbols represent the experimental data for the $Bi_2Sr_2CaCu_2O_{8-\delta}$ intrinsic Josephson junctions from Ref. 4.

of a Hamiltonian that is formally the same as for an *XY* disordered ferromagnet in 3*D* [23,24]. The second one suggests the pinning of the Josephson vortices by microstructural inhomogeneties, and the value of the critical current is determined by the driving force necessary for flux line motion [25,26].

For the system of a Josephson junction network, the magnetic field dependence of the critical current can be described by [23]

$$I_c(B)/I_c(0) \simeq 0.82(1 - \gamma_S^{-1} \sin \gamma_S)^{1/2} B_0/B \ , \quad (6)$$

$$\gamma_S = 2.7(\Lambda/\lambda_c)(B/B_0) \ ,$$

where B_0 is the first critical magnetic field, and Λ and λ_c can be treated as a correlation length for the phase-coupled junctions and a penetration depth, respectively. Unfortunately, Eq. (6) is not able to describe well the experimental $I_c(B)$, especially, in the low magnetic field range, as seen in Fig. 4. Moreover, the obtained fitting parameter $B_0 \simeq 0.25$ T is too large for our films.

Because of the growth mechanism and the tendency to form the so-called columnar texture of the grains, the grain boundaries of the prepared MoN_x films are arranged perpendicularly to the plane of substrate. Consequently, a 2*D* network of Josephson junctions (grain boundaries) with a 2*D* array of Josephson vortices can be formed. The rather small size of the grains ($\langle D \rangle << \lambda_J$), where $\lambda_J = (\phi_0/2\pi\mu_0 j_c\, t)^{1/2} \simeq 10$ μm in our case is the Josephson penetration depth, allows us to assume that the interaction between Josephson vortices leads to the formation of a flux line lattice. The dynamics of such a system can be described by the pinning theory. Since the Josephson vortices do not have normal cores, in contrast to the Abrikosov vortices, the mechanism of the flux line pinning is governed by the magnetic interaction, and the grains play the role of the pinning centers. In this case the critical current is expressed by $I_c(B) = 2cM\phi_0^{1/2}/\lambda B^{1/2}$, where the magnetization $M \simeq (H_{c2} - B)/[4\pi(2\kappa^2 - 1)]$; λ is the London penetration depth, and κ is the Ginzburg–Landau parameter [27]. Taking into account that the elementary pinning force has to decrease proportionally to the drop in the order parameter according to the rise of the field [27], the final expression for $I_c(B)$ is simplified to

$$\frac{I_c(B)}{I_c(0)} \simeq \frac{4\pi}{\kappa d} \left(\frac{\phi_0}{B}\right)^{1/2} \left(1 - \frac{B}{H_{c2}}\right)^2 . \quad (7)$$

Figure 4 shows that the theoretical curve is in good agreement with the experimental data practically in the whole range of magnetic fields. A fitting to the experiment allows us to estimate the values of the Ginzburg–Landau parameter, $\kappa \simeq 70$, and the upper critical field, $H_{c2} \simeq 10$ T, which coincide with those obtained from an analysis of the $(dH_{c2}/dT|_{T_c})$ data for these films.

For comparison, we also show in Fig. 4 the experimental results for the stacked intrinsic Josephson junctions of $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal [22]. It is seen that the magnetic field behavior of the critical current is described by a power law ($I_c \sim 1/B^{1.5}$), as well. However, the pinning mechanism has a more exotic nature and is connected with the collective pinning phenomenon [26].

Summary

We have developed a new method for the preparation of granular MoN_x films that can be treated as a 2*D* network of *SNS* Josephson junctions. It was shown that the temperature dependence of the critical current can be described in the framework of the *SNS* junction model in the diffusive limit. For an explanation of the superconducting transport properties in an applied magnetic field, a simple model for the magnetic pinning of the Josephson vortices is suggested, where the grains play the role of the pinning centers.

This work was supported by the Science and Technology Center in Ukraine through Project No. 1455, and by the Korea Research Foundation through the BSRI program (1998-015-D00087).

1. A. M. Goldman and N. Marković, *Phys. Today* **51**(11), 39 (1998).
2. A. F. Hebard and M. A. Paalanen, *Phys. Rev. Lett.* **65**, 927 (1990).
3. S. Tanda, S. Ohzeki, and T. Nakayama, *Phys. Rev. Lett.* **69**, 530 (1992).
4. A. Yurgens, D. Winkler, T. Claeson, G. Yang, I. F. G. Parker, and C. E. Gough, *Phys. Rev.* **B59**, 7196 (1999).
5. S. L. Roberson, D. Finello, and R. F. Davis, *Thin Solid Films* **324**, 30 (1998).
6. I. A. Parshin, I. L. Landau, and L. Rinderer, *Phys. Rev.* **B54**, 1308 (1996).
7. A. F. Hebard and A. T. Fiory, *Phys. Rev. Lett.* **58**, 1131 (1987).
8. V. Kresin, *Phys. Rev.* **B25**, 157 (1982).
9. V. Z. Kresin, A. Bill, S. A. Wolf, and Yu. N. Ovchinnikov, *Phys. Rev.* **B56**, 107 (1997).
10. N. F. Mott and M. Koveh, *Adv. Phys.* **34**, 329 (1985).
11. V. G. Prokhorov, *Fiz. Nizk. Temp.* **24**, 544 (1998) [*Low Temp. Phys.* **24**, 410 (1998)].
12. V. Ambegaokar and A. Baratoff, *Phys. Rev. Lett.* **10**, 486 (1963); *Phys. Rev. Lett.* **11**, 104 (1963).
13. A. Yurgens, D. Winkler, N. V. Zavaritsky, and T. Claeson, *Phys. Rev.* **B53**, R8887 (1996).
14. K. Kitazawa, *Science* **271**, 313 (1996).
15. A. G. Loeser, Z.-X. Shen, D. S. Dessau, D. S. Marshall, C. H. Park, P. Fournier, and A. Kapitulnik, *Science* **273**, 325 (1996).
16. X.-J. Fan, G. Li, and X.-G. Li, *Physica* **C311**, 93 (1999).
17. R. A. Klemm, C. T. Rieck, and K. Scharberg, *Phys. Rev.* **B58**, 1051 (1998).
18. K. K. Likharev, *Rev. Mod. Phys.* **51**, 101 (1979).
19. U. Ledermann, A. L. Fauchere, and G. Blatter, *Phys. Rev.* **B59**, R9027 (1999).
20. P. W. Brouwer and C. W. J. Beenakker, *Chaos, Solitons and Fractals* **8**, 1249 (1997).
21. M. Tinkham, *Introduction to Superconductivity*, Mc Graw-Hill, New York (1975).
22. V. M. Krasnov, N. Mros, A. Yurgens, and D. Winkler, *Physica* **C304**, 172 (1998).
23. C. Lebeau, A. Raboutou, P. Peyral, and J. Rosenblatt, *Physica* **B152**, 100 (1988).
24. J. Rosenblatt, A. Raboutou, P. Peyral, and C. Lebeau, *Rev. Phys. Appl.* **25**, 73 (1990).
25. J. R. Phillips, H. S. J. Van der Zant, J. White, and T. P. Orlando, *Phys. Rev.* **B47**, 5219 (1993).
26. L. N. Bulaevskii, D. Dommgues, M. P. Maley, A. R. Bishop, and B. I. Ivlev, *Phys. Rev.* **B53**, 14601 (1996).
27. A. Campbell and J. Evetts, *Critical Current in Superconductors*, Taylor and Francis, London (1972).