Observation of the insulator-superconductor transition on solid inert gas and other substrates

2001 J. Phys.: Condens. Matter 13 889

(http://iopscience.iop.org/0953-8984/13/5/309)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **13** (2001) 889–899 www.iop.org/Journals/cm PII: S0953-8984(01)16390-3

Observation of the insulator–superconductor transition on solid inert gas and other substrates

K Das Gupta, G Sambandamurthy, Swati S Soman and N Chandrasekhar

Department of Physics, Indian Institute of Science, Bangalore 560 012, India

E-mail: samband@physics.iisc.ernet.in

Received 15 August 2000, in final form 25 September 2000

Abstract

Observations of the insulator–superconductor transition in amorphous ultrathin films of Bi deposited on solid xenon are presented. The resistance separatrix is found to be close to $h/4e^2$ and the crossover thickness close to 25 Å for all substrates. I–V studies and Aslamazov–Larkin analyses indicate that the superconductivity is inhomogeneous. Screening effects are observed, with the transition temperature increasing as the relative permeability of the substrate is increased, when studying films on quartz and Ge. The resistance separatrix that defines the transition remains unaffected. These results may be qualitatively understood in terms of a percolation-type model.

The insulator–superconductor (I–S) transition has been extensively investigated over the last decade, in a variety of systems such as thin films [1,2] single Josephson junctions [3], arrays [4], and one-dimensional wires [5]. Values of limiting resistance close to the quantum resistance for pairs in one-dimensional wires [5], and two-dimensional, nominally homogeneous ultrathin films [1] are reported. Differing values of the limiting resistance at the transition have been observed [2] in different systems, and this is attributed to the structure, i.e. homogeneous and granular films are expected to behave differently.

A phase-only picture, first proposed by Ramakrishnan [6], and further elaborated by Fisher [7], has been considered appropriate for such systems. A scaling theory of the I–S transition has been developed [8]. This theory predicts that the critical resistance will be universal, i.e. independent of all microscopic details, if the system is invariant under the interchange of the roles of the charge and flux. Thus the I–S transition may be self-dual [9]. The precise value of the critical resistance appears to depend on the nature of the interaction between the charges, being equal to $h/4e^2$ only when the interaction is logarithmic in the separation, one of the conditions which must be obeyed for self-duality. The interaction between charges can be logarithmic in their separation if the 2D films have a sufficiently high dielectric constant [10]. This condition may be met for semiconductor and semimetal films.

This has to be reconciled with experimental observations, which show that superconductivity in ultrathin films can be enhanced by the proximity of a metal or dielectric [11,12]. This is attributed to partial screening of the Coulomb interaction between conduction electrons

0953-8984/01/050889+11$30.00

in the films. Screening also affects the properties of insulating films, by changing the localization length [13, 14]. Glover studied the effect of the substrate dielectric constant on the transition temperature of thin films, over thirty years ago, inconclusively. Ge and Sb have been extensively used as underlayers in ultrathin-film studies [1, 15, 16]. A natural question arises at this point about the effect of the substrate/underlayer on the I–S transition.

For a film on an underlayer, the general expression for the interaction potential between charges in the film is

$$U(r) = \frac{e^2}{4\pi\epsilon_0\epsilon_r}\left(\frac{1}{r} - \frac{1}{\sqrt{r^2+4d^2}}\right) \tag{1}$$

where d is the screening distance (the distance between the mid-plane of the film and the mid-plane of the underlayer) and r the separation between the charges in the film. If the separation of two charges is small compared with the distance between a charge and its image ($2d$), the underlayer makes little difference, and the interaction remains monopolar. At large distances, however, the charge and its image behave as a dipole and the interaction falls off more rapidly with distance. For the films studied here, $2d$ would be close to the film thickness. Although screening by charge carriers in the film is considered in the usual BCS treatment, the relative permeability for a highly disordered film is unknown [17]. It is expected to lie between the metallic and insulating limits of $\epsilon_r \simeq \infty$ and $\epsilon_r \simeq 10$. This issue becomes important in the I–S transition region. $U(q)$, a term in the BCS interaction Hamiltonian, becomes

$$U(q) = \frac{4\pi e^2}{q^2 + k_s^2} \tag{2}$$

where k_s is the inverse screening length. In principle, a film with a transition temperature of 0 K defines R_c. As we show, screening influences the transition temperature of superconducting films, and the normal-state resistance of all films. Therefore it should affect the I–S transition.

It has also been proposed that the I–S transition in the limit $T = 0$ is a combined effect of pairing and localization in 2D systems [18]. Experiments have refuted this conjecture [19]. How disorder, cluster size, and the Coulomb interaction influence the behaviour of Cooper pairs is still not very clear. It has been conjectured that the Coulomb interaction may not influence the critical normal-state sheet resistance [18]. However, this is refuted by observations of the M–I transition in silicon MOSFETS and other systems in zero magnetic field [20]. It has been shown that interactions may increase or decrease the conductance of a disordered 2D electron system—weak interactions increase the dc conductance in the localized regime while they decrease the conductance in the diffusive regime [21]. Strong interactions were always found to decrease the conductance. These considerations motivated our experiments.

In superconductors, Coulomb interactions suppress the fluctuations in the number of electrons, and increase the fluctuations of the phase of the superconducting order parameter. This affects the Josephson coupling energy E_J in inhomogeneous systems, where the competition between the charging energy E_C and E_J drives the transition [19]. Typical systems exhibiting such behaviour are granular films and junction arrays. E_J may be estimated using the Ambegaokar–Baratoff equation, $E_J = \pi\hbar\Delta/4e^2R_N$, where Δ is the BCS gap for Bi, R_N is the normal-state resistance of the film. E_C can be estimated as $E_C = q^2/4\pi\epsilon_0\kappa d$ where $\kappa = \epsilon_r(d/2s + 1)$, where ϵ_r is the relative permeability of the substrate, d is the size of the grains/clusters, and s is the spacing. Since R_N and κ depend on the substrate/underlayer, one expects the I–S transition to occur at different R_N on different substrates. Since the effects of disorder, cluster size, and interactions on Cooper pairs are unclear, explicit predictions of R_N for the I–S transition on various substrates cannot be made.

In this work, we report studies of the I–S transition in quench-condensed bismuth films, as a function of disorder (or film thickness), on a variety of substrates—amorphous quartz, quartz

coated with Ge, and solid xenon condensed on quartz. The relative permeability ϵ_r ranges from 1.5 for Xe to 15 for Ge. Despite screening effects, the I–S transition is robust. Studies for other substrates such as single-crystal sapphire and sapphire coated with Ge, Xe yield similar results. The experiments were done in a custom-built UHV cryostat equipped with reflection electron diffraction (RHEED), capable of producing a hydrocarbon-free vacuum of $\sim 5 \times 10^{-10}$ Torr, described elsewhere [22]. The Ge underlayers were deposited on the substrates in a separate UHV system at room temperature. The Xe underlayers were grown *in situ*. Our experimental set-up resembles that of references [1, 2], although we cannot attain such low temperatures. We quench condense our ultrathin films in the temperature range 1.8 K to 15 K. The substrate temperature influences the disorder in the film, and thereby its properties. Here we report results on films that were quench condensed at 15 K, which facilitates comparison with published results [1].

Since the information obtained is in reciprocal space, it is difficult to comment on the real-space surface morphology. Our RHEED observations are consistent with previous results [23]. Superconductivity in granular systems of rhombohedral Bi clusters has also been reported [24]. Figure 1 shows oscillations in the intensity of the specular RHEED spot, as a function of time, during the deposition of Xe on sapphire. This indicates that Xe may be condensing in the layer-by-layer mode on sapphire. These oscillations enable us to deposit a controlled thickness of Xe on the substrate. Typical thicknesses used were 20 to 25 layers, worked out from the corresponding number of oscillations. Similar oscillations, albeit of smaller amplitude, were seen when the Xe was deposited on quartz.

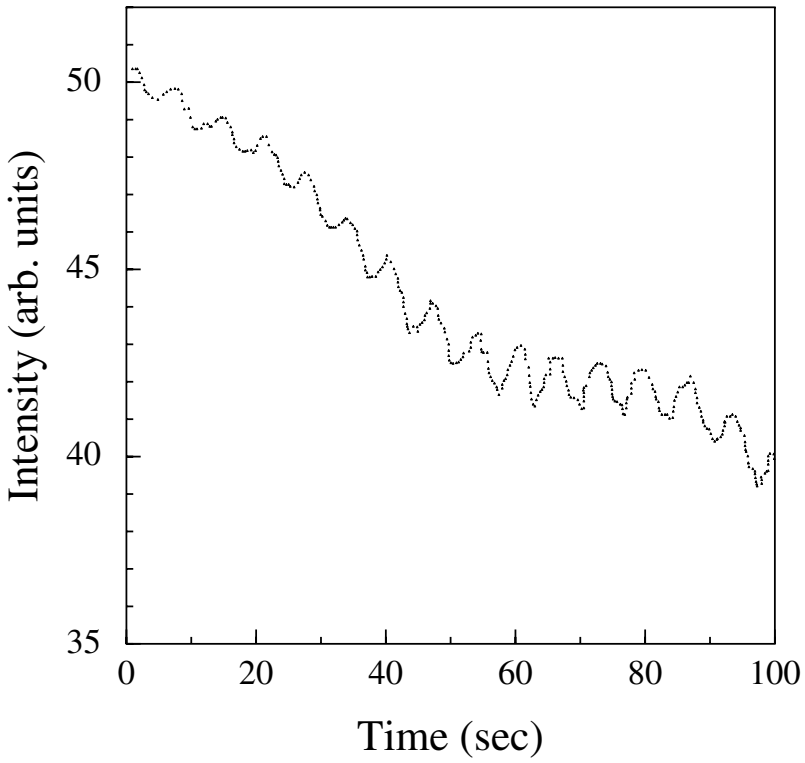


Figure 1. Oscillations in the intensity of the specular RHEED spot, as a function of time, during the deposition of Xe on sapphire.

RHEED studies on Bi films, grown on various substrates and underlayers, show that the Bi is almost amorphous. A RHEED picture is shown in figure 2. It is difficult to unambiguously distinguish between amorphous and nanocrystalline structure at such low temperatures, for poorly conducting films, due to charging effects. On the basis of the Scherrer formula for the peak broadening, we estimate that films thicker than 10 Å are composed of clusters that vary in size from 25 Å to 100 Å [22]. Since the information obtained is in reciprocal space, it is difficult to comment on the real-space surface morphology. Our RHEED observations are consistent with previous results [23]. Superconductivity in granular systems of rhombohedral Bi clusters has also been reported [24].

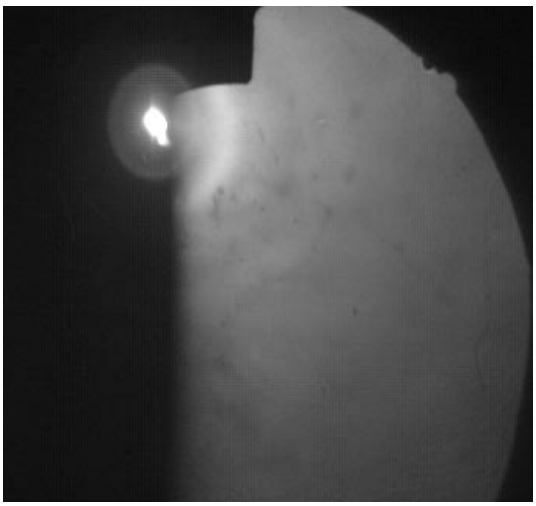


Figure 2. The RHEED diffraction picture from an 82 Å thick Bi film on HOPG. At least one diffuse ring is clearly visible.

Figure 3 shows the evolution of the temperature dependence of the sheet resistance $R(T)$ with thickness for (a) Bi films on Ge (10 Å thick), which has been deposited on amorphous quartz, (b) Bi films on solid xenon condensed on amorphous quartz. A transition from insulating-type behaviour to superconducting behaviour as the thickness of the films is increased is clear. This type of zero-field transition is considered a zero-temperature quantum phase transition, controlled either by disorder, carrier concentration, or thickness. The normal-state resistance at an arbitrarily high temperature R_N has traditionally been used to parametrize the transition, although it may be weakly temperature dependent above the superconducting transition temperature, and becomes ill defined as the I–S transition is approached. The value of the normal-state resistance of a film on the boundary between superconducting and insulating behaviour has been referred to as the resistance separatrix, and has been denoted by R_0 [1,2,5]. We obtain R_0 as an algebraic average of the sheet resistances of the last insulating and the first superconducting films, measured at a relatively high temperature (10 K). R_0 is close to $h/4e^2$ for all three sets of data. This observation indicates that the value of R_0 is substrate independent, and possibly experiment independent.

The transition temperature for a film of a given thickness is higher when the film is on substrates of higher relative permeability, and the normal-state resistance is also higher. A 65 Å film on Xe, quartz, and Ge has $T_c = 3.8$, 4.2, and 4.42 K respectively. These results are consistent with published data [11, 12]. This shows that the conductance of a disordered film depends on the interplay between interaction and disorder [21]. We caution that since

our experiments are limited to 1.8 K, we cannot rule out the possibility that a film which appears to be the last film on the insulating side may, at lower temperature, turn out to be superconducting [15]. A plot of conductance versus thickness at different temperatures gives the crossover thickness, which is shown in figure 4 for films on Xe. We find the crossover thickness to be between 25 to 28 Å for all substrates.

The insulating films follow a behaviour that is consistent with the results reported [25]. We find the conductivities of all of our insulating Bi films, on all substrates, to be given by

$$\sigma(T) = \sigma_0 \exp[-(A/T)^x] \tag{3}$$

where x changes with film thickness. For the thinnest films, x is close to 0.75, due to collective hopping as discussed by Markovic *et al* [25]. This is reasonable, since the experimental apparatus and physical vapour deposition technique used in our work resemble those of reference [25] most closely. As the thickness is further increased, x reduces to 0.5, the value from the Efros–Shklovskii form [26], which describes hopping modified by Coulomb interactions, and finally for the thickest insulating films x is close to 0.33, the Mott value [27].

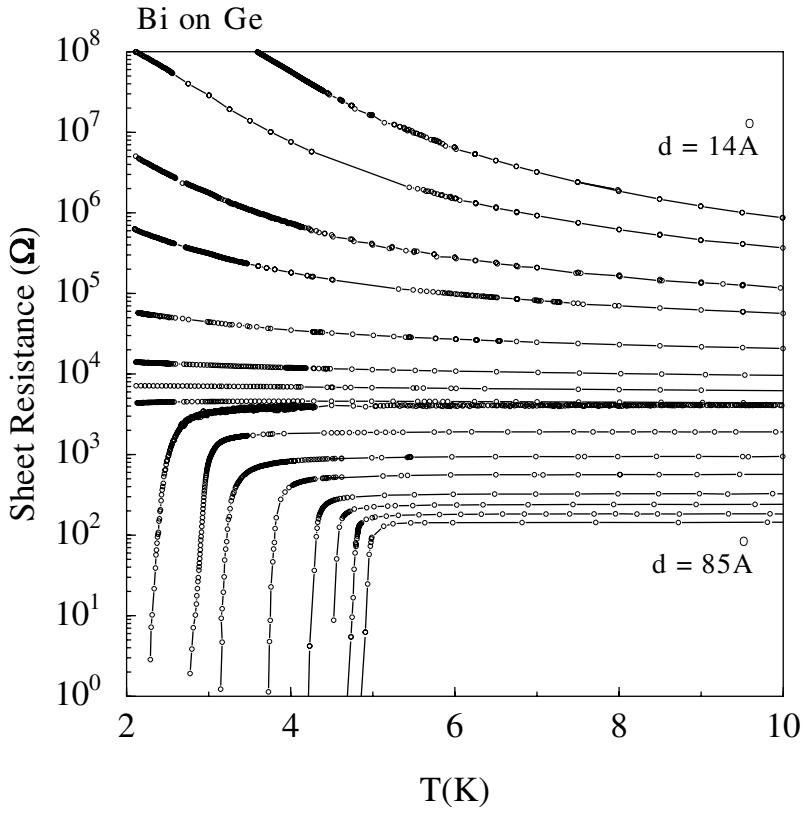
The form of $R(T)$ for these films may lead us to the conclusion that these films are nominally homogeneous. Such a conclusion is incorrect as we show below. Although STM studies of surface morphology [23] report that even at 75% coverage the films are not conducting, and hence show no evidence of percolative behaviour, we find that superconductivity in our films is indeed percolative in nature [30]. Aslamazov and Larkin [28] considered the possibility of fluctuations causing superconductivity. The total conductivity is given by $\sigma = \sigma_N + \sigma'_{2D}$, where σ_N is the normal-state dc conductivity and σ' the paraconductivity. Its temperature dependence is similar to that of the magnetic susceptibility at $T > T_c$. They derived the result

$$\frac{\sigma'_{2D}}{\sigma_n} = \frac{e^2}{16\hbar}\frac{R^N_\square}{\tau} = \frac{\tau_0}{\tau} \tag{4}$$

where $R^N_\square$ is the normal-state sheet resistance, $\tau_0 = 1.52 \times 10^{-5}\ R^N_\square$, and $\tau = (T - T_c)/T_c$ is the width factor. T_c is the mean-field transition temperature. $\tau/R^N_\square$ is a constant ($g_{AL} = e^2/16\hbar$) for all materials. We have evaluated $\tau/R^N_\square$ (g_{exp}) for various films. A systematic dependence of g_{exp} on the thickness d is shown in figure 5. This parameter deviates from g_{AL} for thinner films. It approaches the AL value (g_{AL}) as the thickness is increased. It is assumed that theory predicts $g_{ex} = g_{AL}$ for all films, independently of the microstructure. Both the normal-state conductance and the paraconductance depend on the sample shape. Felsch and Glover [29] have shown that as the microstructure deviates from a 'uniform rectangular slab', g_{exp} exceeds the AL value. The thinner the film, the higher the disorder, the larger the deviations from a slab geometry, and the larger the deviation of g_{exp} from the AL value. Hence, films close to the transition are inhomogeneous.

Further evidence is in the form of hysteretic I–V curves, shown in figure 6. These curves can be understood in terms of a resistively and capacitively shunted random Josephson-junction-array model. These I–V curves have been discussed in detail in a separate publication [22]. The physical picture is of superconducting islands connected by thin regions of normal metal, which act as the weak links between the islands, thus forming a random JJ array. The distribution of grains/clusters, each with a finite number of electrons, requires a finite-N BCS treatment. This results in a spread in T_c [31], so at some temperature all regions of the film are not superconducting. Normal regions exist, and act as weak links. Areal maps of quasiparticle conductance or superconducting order parameter amplitude can resolve such regions.

Feigel'man and Larkin [32] and Spivak *et al* [33] have discussed the quantum superconductor–metal transition in a 2D proximity-coupled array. Thickness larger than the



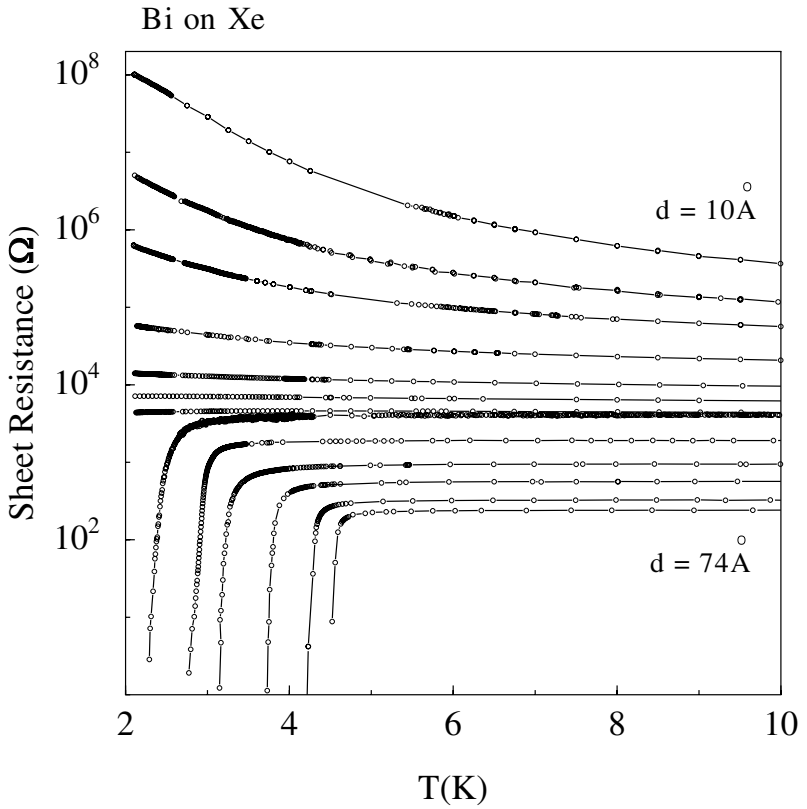
(a)

Figure 3. Evolution of $R_\square$ versus T for Bi on (a) 10 Å Ge on amorphous quartz and (b) solid Xe condensed on amorphous quartz. The substrate temperature during deposition was in all cases maintained at 15 K.

coherence length was considered by the former, with the opposite extreme analysed by the latter. They predict intervention of a normal phase. Limitations on the accessible temperatures prevent our study from confirming this. From the $H_{c2}(T)$ curve, we have determined the coherence lengths (ξ_c) to be close to the film thickness. This regime merits further theoretical study. We have evaluated the localization lengths (ξ_l), and we find that at the I–S transition, $\xi_l \gg \xi_c$, consistent with the results of reference [17].

Earlier theoretical models [7, 34] have shown that the separatrix is unaffected by the dielectric function. One important feature of the earlier papers is the assumption of a constant Josephson coupling energy. This assumption is valid only if the grains or clusters that make up the disordered films, such as those investigated in this work, are (i) on a lattice; and (ii) of uniform size. These parameters are quite difficult to control experimentally unless arrays with predetermined values of these parameters are fabricated. It is certainly implausible that E_J is single valued in our quench-condensed films. De Palo *et al* also consider a phase-only picture [35] similar to that of Fisher, and Emery and Kivelson [7,34]. Therefore, although these models predict the I–S transition to be unaffected by the dielectric constant, the assumptions made therein may not be realized in this and several other experiments.

An I–S transition in fabricated two-dimensional Josephson-junction arrays, which is influenced by the screening provided by a two-dimensional electron gas, has been reported by



(b)

Figure 3. (Continued)

Rimberg *et al* [36]. As the carrier concentration in the 2D electron gas was tuned by changing the voltage on a gate, the I–V characteristic changed continuously between superconductorlike and insulatorlike, demonstrating an influence of screening. The nature of the coupling between the array and the 2D electron gas, and the effect of the array dimensionality on the transition remain open questions. The work reported here differs from those discussed above in two respects: (i) we certainly do not have a single-valued E_J in our films; and (ii) the array is random, with a distribution of E_Js and E_Cs, and is more two dimensional than three dimensional, although we cannot prove this experimentally. The grains or clusters are certainly not on a lattice. We give below a brief description of what we believe is an appropriate description of the transition, drawing an analogy from the recent work on the M–I transition in 2D electron gases, where percolation turns out to be important.

Our results suggest that a percolation-type model, proposed by Meir [37] to explain the M–I transition in 2D systems, is the most appropriate and merits investigation. Meir suggested the following physical picture for the M–I transition in 2D: potential fluctuations due to disorder define density puddles of size L_ϕ or larger, within which the electron wave function totally dephases. Locally, between these puddles, transport is via quantum tunnelling. Support for this idea comes from fits of the conductance at a high temperature (10 K) for the insulating films and the normal-state conductance (at 10 K) for the superconducting films, which follow the same power law with an exponent of 1.33 characteristic of 2D percolation, but with different

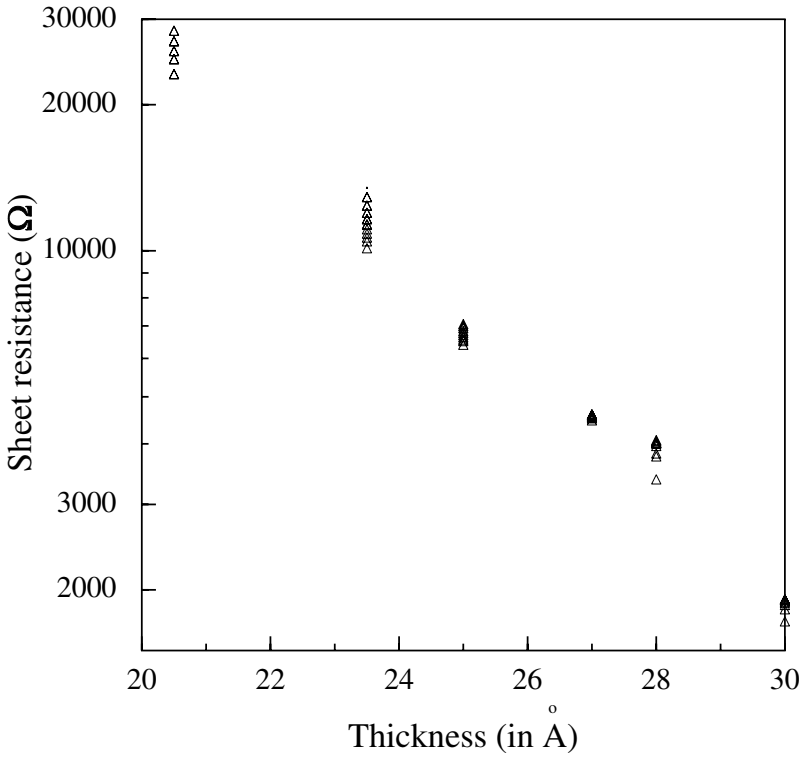


Figure 4. Plots of resistance versus thickness at different temperatures showing the crossover thickness of about 26 Å for Bi films on Xe. The crossover occurs within ±3 Å of this value for all substrates.

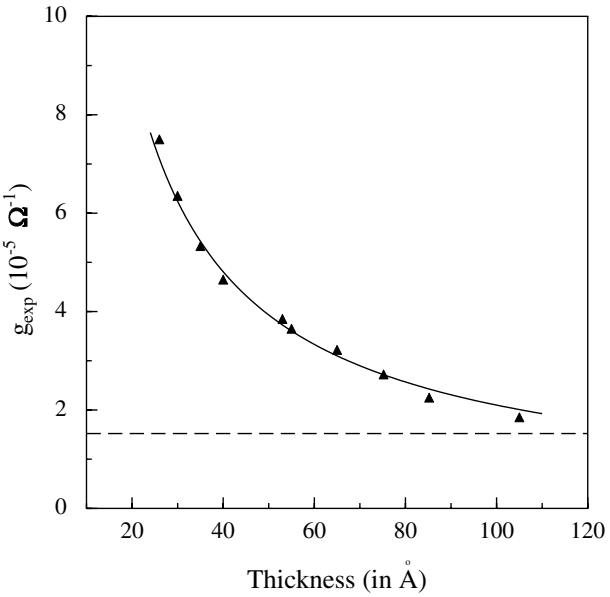


Figure 5. The parameter g_{ex} which characterizes the amplitude of the fluctuations as a function of film thickness. It asymptotically approaches the Aslamazov–Larkin value.

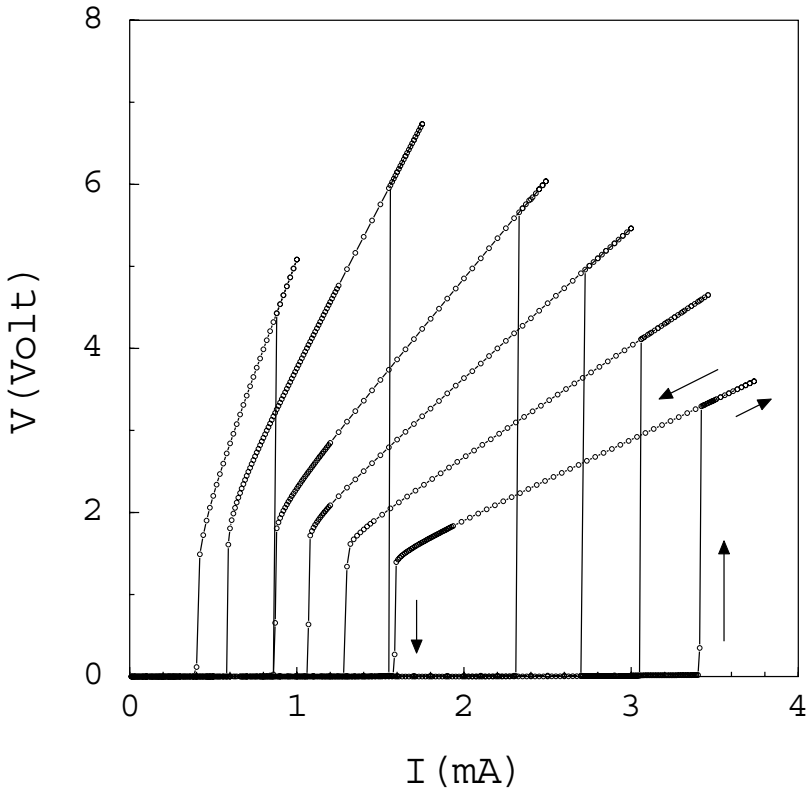


Figure 6. I–V for the superconducting films on quartz. The hysteresis loops show the dissipation due to resistances shunting the junctions of the random Josephson-junction array. The first film is 40 Å thick, and the thickness increment is 10 Å.

critical thicknesses as shown in figure 7. The critical thickness for the insulating films would be the thickness for the onset of electrical conduction. We have no explanation as to why the critical thickness for normal-state conduction depends on whether or not the film superconducts at lower temperature, but present this as an interesting observation. It is logical to associate the puddles with superconducting regions and the quantum point contacts with the normal regions. The competition between the E_C and E_J probably drives the transition. However, it is intriguing that the transition is robust at $h/4e^2$ on all substrates, despite E_C and E_J being dependent on ϵ_r. Percolation issues were studied earlier [39–41].

The conditions for 2D Coulomb interactions may have been met in this and other experiments [1]. Keldysh [10] has shown that this requires the dielectric constant of the film to be much greater than that of the substrate. In this study, substrate dielectric constants range from 1.5 for solid Xe to 15 for Ge underlayers. Hall measurements were done for the first superconducting film near the transition, and this yielded a high carrier concentration of the order of 10^{23} cm^{-3}, which is consistent with that reported in the literature [38], suggesting a high dielectric constant for the films. We have not made Hall measurements for films on the insulating side, since interpretation of data in the hopping regime is complicated. Vortices exist in these films, since they are random Josephson-junction arrays [42], due to the discrete nature of the array, which requires that the current take convoluted paths, consistent with a transition governed by percolation. Duality in JJ arrays has been observed before [4].

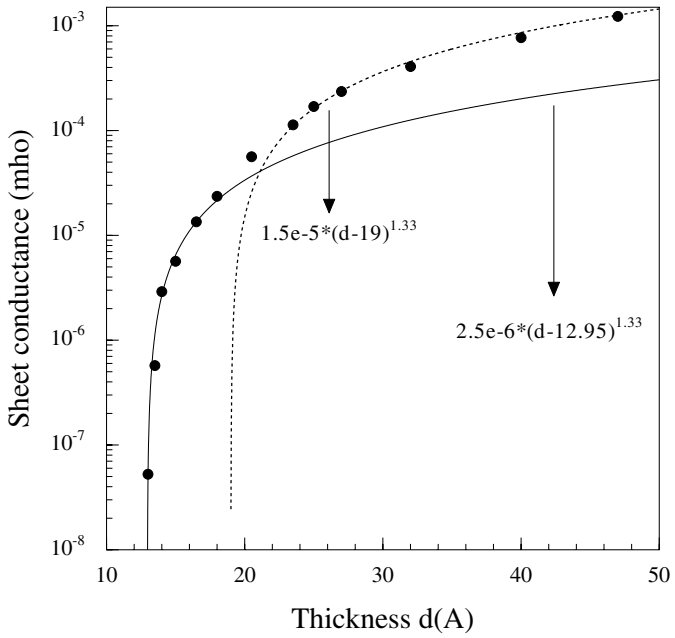


Figure 7. Fits of the conductance of the insulating films (taken at 10 K) and the normal-state conductance of the superconducting films (also taken at 10 K) to the 2D percolation function for Bi on Ge. Only the critical thickness is different in the two regimes, as discussed in the text.

In conclusion, we have observed robust I–S transition in ultrathin Bi on several substrates. We have presented, for the first time, the I–S transition on solid inert gas underlayers. Although the substrate influences T_c as well as R_N for the films, the transition itself is independent of the interaction potential between charges. An AL analysis indicates inhomogeneous films, contrary to $R(T)$. I–V curves indicate a percolation-type transition. A model similar to that proposed by Meir for the M–I transition in 2D merits further investigation. Further studies at lower temperatures are needed.

Acknowledgments

This work was supported by DST, Government of India. We thank Drs Allen Goldman, C J Adkins and Nandini Trivedi for discussions. We thank one of the referees for drawing our attention to reference [34]. KDG thanks CSIR, New Delhi, for financial support through a Junior Research Fellowship.

References

[1] Haviland D B *et al* 1989 *Phys. Rev. Lett.* **62** 2180
Markovic N *et al* 1998 *Phys. Rev. Lett.* **81** 701
[2] Graybeal J M and Beasley M R 1984 *Phys. Rev.* B **29** 4167
[3] Penttila J S *et al* 1999 *Phys. Rev. Lett.* **82** 1004
[4] van der Zant H S J *et al* 1992 *Phys. Rev. Lett.* **69** 2971
[5] Bezraydan A *et al* 2000 *Nature* **404** 3352
[6] Ramakrishnan T V 1989 *Phys. Scr.* T **27** 24
[7] Fisher M P A 1990 *Phys. Rev. Lett.* **65** 923

Fisher M P A 1987 *Phys. Rev.* B **38** 1917
[8] Cha M-C *et al* 1991 *Phys. Rev.* B **44** 6883
[9] Wen X G and Zee A 1990 *Int. J. Mod. Phys.* B **4** 437
[10] Keldysh L V 1979 *JETP Lett.* **29** 658
[11] Astrakharchik E G and Adkins C J 1998 *J. Phys.: Condens. Matter* **10** 4509
[12] Landau I L *et al* 1991 *JETP Lett.* **53** 263
[13] Adkins C J and Astrakharchik E G 1998 *J. Phys.: Condens. Matter* **10** 6651
[14] Entin-Wohlman O and Ovadyahu Z 1986 *Phys. Rev. Lett.* **56** 643
[15] Valles J M Jr *et al* 1992 *Phys. Rev. Lett.* **69** 3567
[16] Strongin M *et al* 1970 *Phys. Rev.* B **1** 1078
[17] Butko V Yu *et al* 2000 *Phys. Rev. Lett.* **84** 1543
[18] Pang T 1989 *Phys. Rev. Lett.* **62** 2176
[19] Kagawa K *et al* 1996 *Phys. Rev.* B **53** R2979
[20] Abrahams E *et al* 2001 *Rev. Mod. Phys.* at press
(Abrahams E *et al* 2000 *Preprint* cond-mat/0006055)
[21] Vojta T *et al* 1998 *Phys. Rev. Lett.* **81** 4212
[22] Sambandamurthy G *et al* 2000 *Solid State Commun.* **115** 427
[23] Ekinci K L and Valles J M Jr 1999 *Phys. Rev. Lett.* **82** 1518
[24] Weitzel B and Micklitz H 1991 *Phys. Rev. Lett.* **66** 385
[25] Markovic N *et al* 2000 *Phys. Rev.* B **62** 2195
[26] Efros A L and Shklovskii B I 1975 *J. Phys. C: Solid State Phys.* **8** L49
[27] Mott N F and Davis E A 1979 *Electronic Processes in Non-Crystalline Materials* (New York: Oxford University Press)
[28] Aslamazov L G and Larkin A I 1968 *Phys. Lett.* A **26** 579
[29] Felsch W and Glover R E 1971 *J. Vac. Sci. Technol.* **9** 337
[30] Henning P F *et al* 1999 *Phys. Rev. Lett.* **83** 4880
[31] Tsai A P *et al* 1999 *Appl. Phys. Lett.* **75** 1527
[32] Feigel'man M V and Larkin A I 1998 *Chem. Phys.* **235** 107
[33] Spivak B *et al* 2000 *Preprint* cond-mat/0004058
[34] Emery V J and Kivelson S A 1995 *Phys. Rev. Lett.* **74** 3253
[35] De Palo S *et al* 1999 *Phys. Rev.* B **60** 564
[36] Rimberg A J *et al* 1997 *Phys. Rev. Lett.* **78** 2632
[37] Meir Y 1999 *Phys. Rev. Lett.* **83** 3506
[38] Buckel V and Hilsch R 1952 *Z. Phys.* **132** 420
Buckel V and Hilsch R 1954 *Z. Phys.* **138** 109
[39] Kapitulnik A *et al* 1998 *Phys. Rev. Lett.* **80** 3352
[40] Seshadri H *et al* 1995 *Phys. Rev. Lett.* **75** 4075
[41] Yazdani A *et al* 1995 *Phys. Rev. Lett.* **74** 3037
[42] Xia W and Leath P L 1989 *Phys. Rev. Lett.* **63** 1428
Leath P L and Xia W 1991 *Phys. Rev.* B **44** 9619