SUPERCONDUCTOR–INSULATOR TRANSITIONS IN THE TWO-DIMENSIONAL LIMIT

Increasing the disorder of an ultrathin superconducting film may produce a quantum phase transition at zero temperature to an insulating state.

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The investigation of superconductivity in the presence of disorder began 60 years ago with the work of Alexander Shal'nikov at the Institute for Physical Problems in Moscow. The subject has played an ongoing role in condensed matter physics over the years. Interest has recently been heightened by the possibility that the disorder-driven or magnetic-field-driven quenching of superconductivity in systems at the limit of zero temperature and two dimensions might be quantum phase transitions.[1] That would link the physics of the superconductor–insulator transition in thin films to other systems believed to exhibit quantum phase transitions—for example, helium-4 in porous media, high temperature superconductors, Josephson-junction arrays, two-dimensional electron gases and various spin systems.

Disorder, which is relevant to superconductivity, can be morphological or chemical. Early in the game, Philip Anderson[2] showed that nonmagnetic impurities have no significant effect on the superconducting transition by pointing out that Cooper pairs are formed from time-reversed eigenstates, which have disorder included. Anderson's idea applies only to weakly disordered systems, with their extended electronic states.

One can, however, increase disorder to a level where electronic wavefunctions become localized. The investigation of superconductivity in that localized regime provides a unique opportunity for studying the competition between the attractive interaction responsible for superconducting pairing and the pair-breaking effects of localization and disorder-enhanced Coulomb repulsion. The latter is an important route to the localization of electronic wavefunctions.[3] Whereas superconductivity is a manifestation of long-range phase coherence between electron pair states, electronic localization involves a limitation of the spatial extent of the wavefunctions, which should preclude such pairing. One would therefore anticipate that superconductivity should disappear as disorder increases and states become localized. In 1985, Patrick Lee and Michael Ma at MIT presented a scenario for the persistence of superconductivity even when all states are localized.[4]

Of particular interest are very thin films, where both superconductivity and metallic behavior are marginal. In two dimensions, a superconducting transition is barely possible. It would be a topological Kosterlitz-Thouless-Berezinski (KTB) transition.[5] Weakly interacting electron systems in two dimensions are always localized, even for arbitrarily weak disorder.[3] (This view is changing; recent investigations of strongly interacting systems suggest that there can be a metal–insulator transition in two dimensions.[6])

The subject of the superconductor–insulator transition in two dimensions is an active area of experimental and theoretical study. There is no consensus as to the nature and applicability of the bosonic models of the quantum phase transition that have been proposed.[7] Indeed, the conventional picture of the suppression of superconductivity by disorder contains no quantum phase transition.

Disordered thin films

Thin films are good candidates for studying superconductor–insulator transitions, because they can be fabricated with disorder on different length scales. The superconducting state in metallic and alloy systems is usually forgiving of even substantial amounts of disorder. The modification of a film's superconducting properties caused by disorder depends on the strength of the disorder and its geometrical scale relative to intrinsic scales. The relevant intrinsic scales include the interatomic spacing, the inverse Fermi momentum (a few angstroms), the electronic mean free path, the London penetration depth, the Bardeen-Cooper-Schrieffer coherence length (10^4 Å) and the zero-temperature Ginzburg–Landau coherence length, which can be as short as 50 Å. This last is an approximate measure of the size of the normal core of a superconducting vortex.

The control and quantification of a film's disorder length scale represent a critical experimental problem. The disorder can be on any scale ranging from 2 to 500 Å. It is determined by processing. Metallic or metal-oxide films can be made inhomogeneous with a mesoscopic (about 100 Å) disorder length scale. With care, disorder can be created on atomic scales in sputtered films, or in films deposited on specially prepared substrates cooled to liquid helium temperatures. Such films can contain metallic clusters connected electrically by relatively narrow necks, or insulating junctions with tunneling through the substrate or through space.

In sputtered films that are nominally homogeneous on atomic scales the superconducting transition temperature is observed to be a decreasing function of the sheet resistance $R_\square$ of the film, measured in the normal state.

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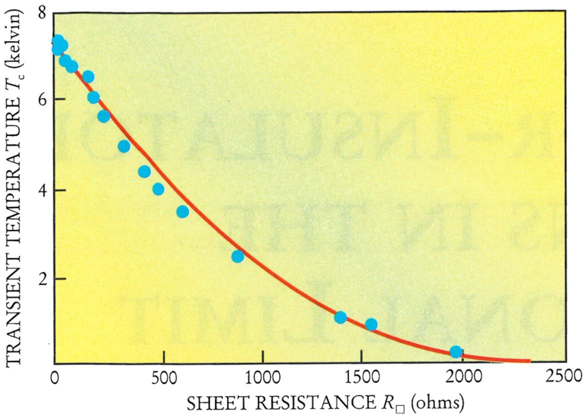

FIGURE 1. CRITICAL TEMPERATURE for the transition of a thin $Mo_{77}Ge_{23}$ film to superconductivity falls with increasing sheet resistance (decreasing thickness) in good agreement with models (red curve) based on weakening of Coulomb screening by disorder. (Adapted from ref. 8, Graybeal and Beasley.)

(Because the resistance of a film is proportional to its length and inversely proportional to its width, the resistance of a *square* film, denoted $R_\square$, is independent of the size of the square, but not of the film's thickness. It is used to characterize resistive behavior in two dimensions.) As we see in figure 1, the transition temperature falls with increasing $R_\square$ (or decreasing film thickness) in very much the way predicted by perturbative theoretical models based on the weakening of Coulomb screening with increasing disorder.[8] The response of such films to magnetic fields turns out to resemble a quantum critical point.

In films produced by evaporating metal onto cold substrates, repeated deposition of small amounts of metal followed by measurement reveals the evolution of superconductivity with increasing thickness. If such studies are carried out on substrates precoated with a thin layer of amorphous germanium or antimony, one seems to get disorder on atomic rather than mesoscopic scales. In figure 2 we see such films exhibiting a clear separation between superconducting and insulating behavior with increasing thickness, in the limit of zero temperature.[9] For bismuth, the resistance at the separation between insulators and superconductors is very close to the quantum resistance for electron pairs: $h/4e^2$ or 6450Ω.

It has been suggested that films formed this way are amorphous and free of clustering, because the underlayer promotes the wetting of the substrate by the film. Thus it permits the onset of conductance at thicknesses on the order of a monolayer.[10] There may, however, be mesoscale clusters. But the underlayer, though itself nonconducting, can provide a tunneling channel for the electrical coupling between clusters, thus giving rise to electrical connectivity at very early stages of surface coverage.

Films made by low-temperature evaporation without an underlayer of amorphous germanium or antimony are believed to be granular. They do possess mesoscale clusters, and they exhibit a separation between superconducting and insulating behavior in the limit of zero temperature. But their properties differ in certain details from the nominally homogeneous amorphous films described above. In particular, their superconducting transition temperatures are independent of thickness, and there is evidence that the mesoscale clusters are superconducting, with transition temperatures close to the bulk values. These systems may exhibit quantum critical behavior. So may Josephson junction arrays, which have similar properties.

High-temperature superconductors have been studied in a variety of configurations: Films have had their superconductivity weakened by disruptive ion bombardment or oxygen depletion, and they have been driven normal by applied magnetic fields. Single crystals of $YBa_2Cu_3O_{7-x}$ have been made to behave like two-dimensional systems by controlled oxygen depletion. High-temperature (oxide) superconductors are serving as models for the study of the superconductor–insulator transition. This modeling is, however, severely complicated by the contrast between the simple metallic systems and the structural, chemical and physical complexity of the oxides.

Aspects of superconductivity

To achieve an elementary understanding of the superconductor–insulator transition, it is useful to characterize the superconducting state by the complex order parameter

$$\Psi = \Psi_0 \, e^{i\phi} \ .$$

Ψ is, in effect, a macroscopic wavefunction for the superconducting electrons. Even though superconductors are complicated interacting many-body systems, many of their properties can be described by a very simple picture in which the aggregate collection of electrons behaves much like a single electron. Resistance vanishes when the order parameter is nonzero and there is long-range phase coherence—that is to say, when the phase ϕ becomes time-independent. In the presence of phase fluctuations, it is, in principle, possible to have finite resistance even with a nonzero order parameter.

Fluctuations in either the amplitude or phase of the order parameter are central to various models of the superconductor–insulator transition in disordered films. One can regard models in which electronic localization weakens superconductivity as primarily affecting the amplitude Ψ_0. By contrast, the "dirty boson" models discussed below are mostly concerned with phase fluctuations.

Fluctuations in the phase play a central role in the superconductivity of films not so thin or disordered that their electrons are strongly localized, but thinner than the London penetration depth, which is the length scale for the variation of currents and magnetic fields associated with Abrikosov vortices. The onset of superconducting behavior in this regime can be characterized as a topological (KTB) phase transition.[5] By contrast with three-dimensional superconductors, this transition is not accompanied by the onset of true long-range phase coherence.

Vortex–antivortex pairs, which are topological excitations produced in this regime by thermal excitation, become bound below a characteristic temperature and cease to contribute to the flux-flow resistance. Flux flow is a process by which resistance can develop in a superconductor. Under the action of an applied current, vortices generally move at right angles to the current, so that the induced emf results in a nonvanishing resistance. That is one way of producing a time-varying phase.

KTB transitions have been studied in many different types of film and junction array systems with sheet resistances on the order of a kΩ. One should see a KTB transition except when there are sample-dependent length scales due to granularity or defects that cut off the long-range vortex interaction. One also fails to see KTB transitions in films with sheet resistances close to $h/4e^2$, the quantum resistance for pairs. See, for example, the separatrix region in figure 2. In such cases, evidence of the KTB transition appears to be suppressed by localization effects and quantum fluctuations of the order parameter's phase.

The resemblance of what we see in figure 2 to renormalization flow diagrams in statistical mechanics has led to the suggestion of a superconductor–insulator transition at zero temperature very much like the dirty-boson quantum phase transition.[7] This approach assumes that the crossover from superconductor to insulator is a consequence of phase fluctuations. It attempts to go beyond perturbative treatments, which are unsatisfactory in the limit of strong disorder.

Quantum phase transitions

A quantum phase transition (QPT) is a transition at absolute zero brought about by changing a parameter in the Hamiltonian of a quantum system.[1] The quantum mechanical ground state changes when the critical point is crossed. By contrast with phase transitions at nonzero temperature, quantum effects are central to QPTs. In other phase transitions, the order parameter itself may be quantum mechanical, but classical thermal order-parameter fluctuations govern the behavior of the transition at the relevant long wavelengths. In superconductors, for example, Ψ is related to an underlying many-body electronic wavefunction, but fluctuations are described by a classical phenomenological Landau–Ginzburg free energy. The fluctuations are classical, because the thermal energy $k_B T$ is much greater than $\hbar\omega$ for all frequencies of interest. (k_B is the Boltzmann constant.) But in QPTs, where the temperature must be zero, the fluctuations themselves are quantum mechanical.

Near a quantum phase transition, ξ and ξ_τ, the spatial and temporal correlation lengths, are divergent. ξ_τ is associated with a vanishing energy scale. If we define a control parameter δ as the absolute value of the difference between the tuning parameter (for example, film thickness or magnetic field) and its critical value, then we can write

$$\xi \propto \delta^{-\nu} \quad \text{and} \quad \xi_\tau \propto \xi^z \ . \qquad (1)$$

That defines the correlation-length exponent ν and the dynamic critical exponent z. In the critical region close to the QPT, physical properties are homogeneous functions of the independent variables in the problem.[1]

A key feature of QPTs is the interplay of dynamics and thermodynamics. As a consequence of this interplay, a d-dimensional quantum system at finite temperature is described in the $T \to 0$ limit as a classical system of $d+1$ dimensions. (This is strictly true for $z = 1$; otherwise the dimensionality is $z + d$.) The finite extent of the system in the extra dimension is given, in units of time, by $\hbar / k_B T$; it becomes infinite only in the $T \to 0$ limit.

It is remarkable that important features of the quantum transition can be studied extensively in a classical context. One can treat the quantum mechanical problem

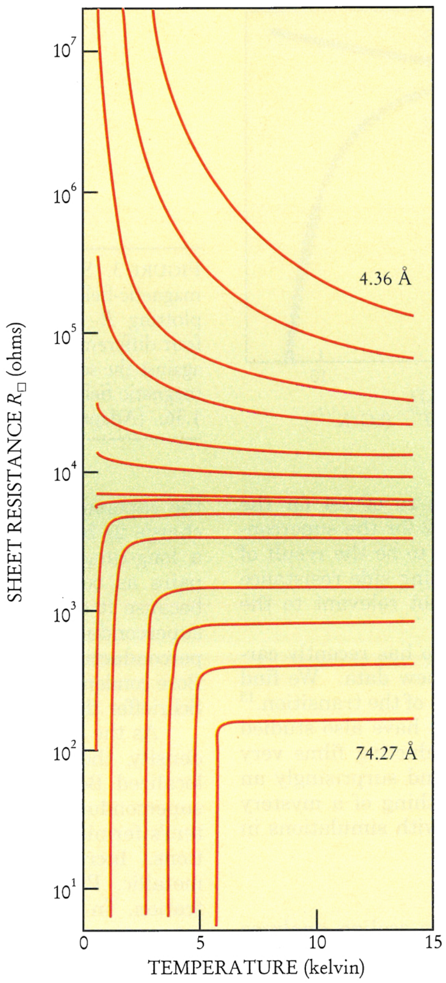

FIGURE 2. TEMPERATURE DEPENDENCE of sheet resistance for various thicknesses of bismuth film exhibits a striking separatrix near 6450 $\Omega = h/4e^2$. Film thicknesses range from 4.36 Å (top curve) to 74.27 Å (bottom). (Adapted from ref. 9, Liu *et al.*.)

computationally with simulations of the $d+1$ dimensional classical problem. But disorder can change the universality class of the equivalent classical problem. Furthermore, space and time do not, in general, enter the equivalent classical problem in the same way unless the dynamical exponent z is unity. And the value of z depends on the interactions in the system.

The effect of considering nonvanishing temperature in statistical mechanical analysis is to force the "temporal" dimension of the problem to be finite. One can then use a finite-size scaling model to analyze data at nonzero temperatures. The success of such analyses for superconductor–insulator transitions argues that they have quantum critical points. The change in behavior at nonzero temperature can be a phase transition or a crossover.

The scaling dependence of the sheet resistance on the temperature and the control parameter in two dimensions has the form:

$$R_\square = R_c f(\delta / T^{1/z\nu}) \ , \qquad (2)$$

where R_c is the critical resistance. The tuning parameter is most often the magnetic field or film thickness, but it could also be charge density, stress or disorder at fixed thickness.[11] Indeed, if the system is close enough to the transition, it probably doesn't matter what tuning parameter the experimenter uses.[7]

Although the data shown in figure 2 suggested that that the insulator–superconductor transition was a quantum phase transition, the first quantitative attempt at a finite-size scaling analysis was in the 1990 work of Arthur Hebard and Mikko Paalanen on the field-driven transition of amorphous composite In_2O_3 films.[11] Figure 3 shows the scaling behavior of equation 2 in more recent data taken by Ali Yazdani and Aharon Kapitulnik with amorphous MoGe films.[12] They find that the exponent product $z\nu = 1.36$.

One can determine the dynamical exponent z without a scaling analysis of the resistance: Hebard and Paalanen have argued[11] that the critical magnetic field B_c goes like $T_c^{2/z}$. Yazdani and Kapitulnik scaled the electric-field dependence of the dynamical resistance at fixed temperature and found the exponent product $\nu(z+1) = 2.73$. That yielded $z = 1.0 \pm 0.1$. That's what one expects in systems with long-range Coulomb interactions. For the magnetic-field-driven transition, both ν and z were found to agree with theoretical expectations.

Until recently it was less clear that there was a quantum critical point in the transitions one gets by varying the film thickness or disorder. The scaling of the

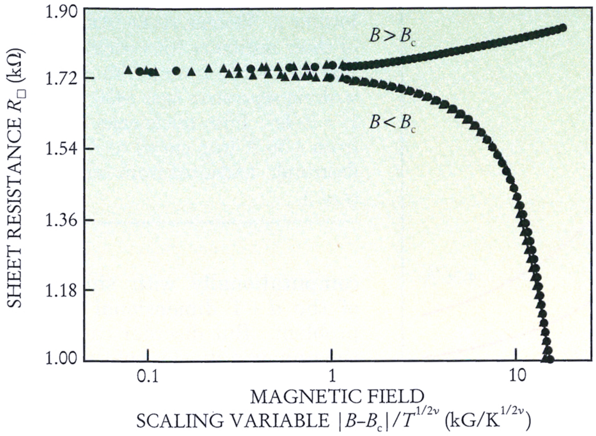

FIGURE 3. SCALING BEHAVIOR of the temperature and magnetic-field dependence of sheet resistance is exhibited by plotting $R_\square$ of a sputter-deposited MoGe film measured at four different temperatures (from 80 to 110 mK) together against the scaling variable $|B - B_c|/T^{1/z\nu}$. Here the critical magnetic field B_c = 4.19 kG and the exponent product $z\nu$ = 1.36. (Adapted from ref. 12.)

figure 2 data gave exponent products $z\nu$ of 2.4 for the insulating side of the transition and 1.2 for the superconducting side.[9] This mismatch appears to be the result of including in the analysis of the insulating side resistance data that involve a transport mode not relevant to the insulator–superconductor transition.

Our University of Minnesota group has recently carried out a more complete study with new data. We find an exponent product of 1.3 on both sides of the transition.[13] The results are shown in figure 4. We have also studied the field-driven transition of superconducting films very close to the critical resistance and found surprisingly an exponent product of 0.7. That's something of a mystery at this point, because it is consistent with simulations in which there is no disorder.

Dirty bosons

Despite some rough edges, the various scaling analyses appear to establish the existence of QPTs associated with both the disorder- and field-driven transitions. The issue of the applicability of the bosonic models proposed as a description of these transitions is not well established. Let us examine this issue.

The problem of Bose particles in a random medium, originally considered in the context of helium in porous media, has come to be called the dirty-boson picture. In the superconducting case, films are modeled by pointlike charge $2e$ bosons in a random potential, interacting with a long-range Coulomb force. The treatment of Cooper pairs as composite bosonic particles has been justified because it does correctly describe the critical behavior of superconductors. In three dimensions, models of the superconducting transition based on a finite-temperature Bose condensation and those based on the Bardeen–Cooper–Schrieffer theory belong to the same universality class.

As the boson density is increased through the critical density, there is a $T = 0$ transition from an insulating localized Bose-glass phase (localized Cooper pairs) to a superconducting phase. That picture ignores all the system's fermionic properties, such as single-particle excitations. Increasing the thickness of a film makes it more metallic. Presumably it also increases the carrier concentration. In the presence of an attractive electron–electron interaction, that results in an increased Cooper-pair (boson) density.

Applying a magnetic field adds vortices, which interact with a logarithmic potential and behave like quantum point particles. In the presence of disorder, these vortices are pinned and the usual vortex-lattice phase is replaced by a vortex glass. At temperatures above zero in a magnetic field, the resistance can be nonvanishing, because vortices can move either by thermal activation or quantum tunneling. But as the magnetic field is increased, the quantum gas of point vortices can, in principle, become a Bose condensate at some critical field. This condensation results in a Bose insulator with localized Cooper pairs.

There is a parallel between the disorder-controlled and field-controlled transitions. Adding charged bosons (Cooper pairs) by increasing film thickness results in a Cooper-pair condensate, and adding uncharged bosons (vortices) by applying a perpendicular magnetic field leads to a vortex condensate. Vortices and charges are related by a duality transformation. In two-dimensional super-

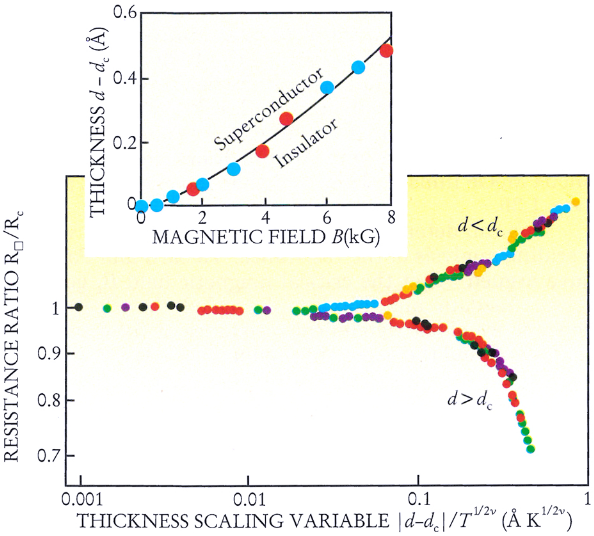

FIGURE 4. DEPENDENCE OF SHEET RESISTANCE (divided by its critical value) on film thickness d shows the expected scaling behavior when measurements in different magnetic fields (from 0 to 10 kG) are plotted together against the scaling variable $|d - d_c|/T^{1/z\nu}$. Here the exponent product $z\nu = 1.4 \pm 0.1$, and d_c is the critical thickness in the absence of a magnetic field. The inset is a phase diagram for the critical thickness as a function of magnetic field. The curve indicates the power law $d - d_c \propto B^{1.4}$. (Adapted from ref. 13.)

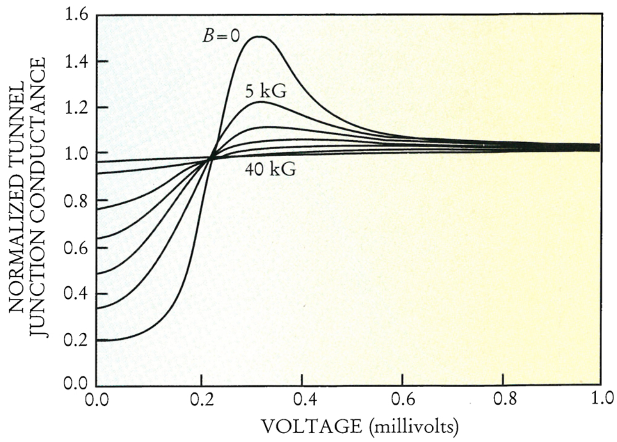

FIGURE 5. TUNNELING JUNCTION CONDUCTANCE of a PbBi/Ge film measured at T = 360 mK and various magnetic fields. (The temperature T_{c0} at which the resistance has dropped to half its normal-state value is 1.64 K.) The curves are for five different magnetic fields, ranging from 0 to 40 kG. No energy-gap feature is found for insulating films. (Adapted from ref. 15, Hsu, Chervenak and Valles.)

conductors this transformation interchanges particles and vortices and maps the insulating and superconducting phases onto each other.[14] We can view the insulator at zero temperature as a Bose condensate of vortices with the original Bose particles now localized. The dissipationless flow of vortices in the insulating phase is thus the dual of superconductivity. Just as moving charges induce current, moving vortices induce voltage.

An important additional prediction of the dirty boson theory is that, at the critical point (with both T *and* δ at zero), the conductivity is finite. This means there should be true metallic conduction. Furthermore, renormalization-group arguments lead to the conclusion that the sheet resistance at criticality is universal, its value depending upon the universality class of the transition, and not on microscopic details.

In the very special case in which the insulator and superconductor are self-dual, the resistance at criticality would be $h/4e^2$, the quantum resistance for pairs. That can be understood by a simple argument: When the system switches from superconductor to insulator, both charges and vortices move. A flow of Cooper pairs results in a current $I = 2e(dn/dt)_{cu}$. Vortices moving at right angles to the current produce a voltage $V = (h/2e)(dn/dt)_v$. When a superconductor is self-dual, vortices and charges behave identically and the two time derivatives are equal, so that the resistance $R = V/I$ becomes $h/4e^2$.

The experimental situation

An important feature of the dirty-boson model is the prediction that there is a universal limiting resistance at the critical point. Although some studies[9] find limiting resistances close to $h/4e^2$, different values of the critical resistance have, in fact, been reported for a number of thin-film systems. The spread of measured values may be extrinsic; there are morphological differences between films of different materials. In particular, films believed to be homogeneous may, in fact, be granular. The Josephson coupling between grains in such films would be determined by a critical value of the ratio of the electrostatic energy to the Josephson coupling energy, which would depend on geometry. Alternatively, material-specific features such as the strength of spin–orbit coupling might influence the universality class. Finally, the data may not really be from the critical regime of the $T = 0$ phase transition. The size of this regime is not known. It may require studies at lower temperatures, with values of the control parameter closer to criticality.

Tunneling studies of the density of states in ultrathin films also suggest that dirty-boson physics cannot be the whole story.[15] In particular, when superconductivity is tuned by varying the film thickness, the energy gap obtained by measuring the tunneling conductance is found to scale with the transition temperature. It disappears when the superconductivity vanishes. A similar study has been carried out on the magnetic-field driven transition, although in that case the density of states associated with the gap, rather than the gap itself, decreases with increasing field and vanishes in the insulating state. This is seen very clearly in figure 5.

The simplest interpretation of such studies is that there are serious amplitude fluctuations as well as phase fluctuations associated with the superconductor–insulator transition. One might also conclude from this interpretation that the vanishing of the energy gap implies the vanishing of the order parameter in the insulating state. That would call into question our earlier discussion of the superconductor–insulator transition. Such a conclusion should be treated with care. The vanishing of the gap aspect of tunneling could result from other effects, such as pair breaking by phase fluctuations. In any case, tunneling experiments raise serious questions as to the completeness of a phase-only picture of the transition.[16]

In the magnetic-field-driven transition, the values of the critical resistance cluster about the quantum resistance for pairs. But there too, the data indicate that the limiting resistance at the transition is not universal. Yazdani and Kapitulnik have suggested that this may be due to parallel electronic conduction channels.[12] It may also turn out that bosonic models incorporating local ohmic dissipation, in which the dynamical critical exponent is damping-dependent, will provide the clue to what is happening.[17]

The various bosonic models assume that Cooper pairs exist in the insulator. This certainly is the case in granular films and Josephson junction arrays. It might also be true for "uniform" films: Hall effect studies on indium oxide films suggest a crossover between two distinct insulating phases. When the longitudinal resistance R_{xx} and the transverse Hall resistance R_{xy} are measured on the same film,[11] we see in figure 6 that R_{xx} crosses over to insulating behavior at a field strength lower than that of a second crossover, found in the dependence of R_{xy} on B. At the higher-field feature there is also a drop in the resistance. It has been suggested that the first crossover indicates a transition between a superconductor and a Bose insulator—a state with nonzero pairing but infinite resistance at zero temperature—and the second feature is a crossover or transition to a Fermi electronic insulator without pairing.[11]

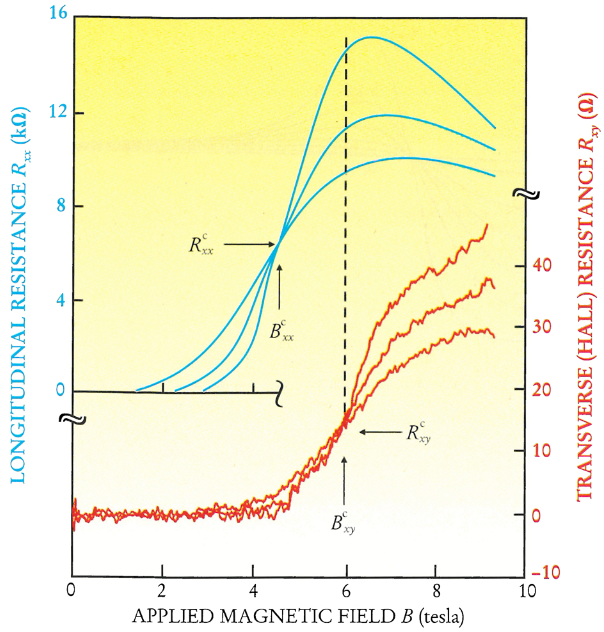

FIGURE 6. LONGITUDAL AND TRANSVERSE resistances of an amorphous In_2O_3 thin film superconductor, measured as a function of applied magnetic field, exhibit quite different critical fields B^c, as manifested by the crossover point for different isotherms from 40 to 200 mK. The right-hand axis refers to the transverse (Hall) resistance R_{xy} (red curves). The separation of the critical field values suggests an insulating phase of mobile bosonic vortices above the critical field for the field-driven superconductor–insulator transition. The high-field (fermionic) insulating phase appears to be less resistive than this bosonic phase. (Adapted from ref. 11, Paalanen, Hebard and Ruel.)

A second piece of supporting evidence for Cooper pairing in the insulator comes from the magnetoresistance of quench-deposited films. James Valles and Shih-ying Hsu have seen[15] a crossover from activated conduction (where conductance varies like $\exp(-\sqrt{T_0/T})$ to conduction that goes like log T. This crossover is accompanied by a change in the sign of the magnetoresistance.

Our group has observed these effects to be correlated with the development of anisotropy in the magnetoresistance of sets of films that ultimately become superconducting.[13] We found the difference between the measurements with the applied field perpendicular and parallel to the film plane—which should be a measure of the orbital magnetoresistance—to be linear in magnetic field. That linearity could be due to the flux flow of vortices *in the insulating state*. Theorist Efrat Shimshoni and coauthors have recently considered a system of quantum disordered Cooper pairs subject to a penetrating magnetic field, with one flux quantum per Cooper pair.[18] These objects might result in flux-flow resistance, which would exhibit linear magnetoresistance.

Challenges

The success of finite-size scaling analyses of the superconductor–insulator transitions as a function of film thickness or applied magnetic field provides strong evidence that $T = 0$ quantum phase transitions are occurring. On the other hand, the superconducting gap experiments and the lack of a universal limiting resistance at zero temperature raise serious questions as to whether the theoretical picture based on dirty-boson physics and phase fluctuations correctly describes the critical behavior of these two-dimensional systems. Certainly the correct theory should not ignore electronic degrees of freedom.

There are challenges to the experimentalist as well. We need lower temperatures, so we can be sure that the scaling analyses are really in the critical region. The nature of the insulating state must be investigated further. There are also concerns about the frequencies at which the measurements are made. To be confident of having reached the quantum limit,[1] one would like to achieve the condition $\hbar\omega \gg k_B T$.

There is also the issue of the chemical composition and structure of the various films used in these investigations. To assert that these films are homogeneous is, for quench-vaporated films, a minimalist assumption or, for sputtered films, an extrapolation from measurements on thicker films. It would be useful to obtain more structural information on films much thinner than 50 Å. It would also be important to investigate the behavior of films as the length scale for disorder changes from the microscopic to the mesoscopic scale.

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