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Autor(en): **Hebard, A.F. / Palaanen. M.A.**

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Quenching of Superconductivity in Disordered Thin Films by Phase Fluctuations

A. F. Hebard and M. A. Palaanen
A T & T Bell Laboratories
Murray Hill, New Jersey 07974

Abstract. The amplitude Ψ_0 and phase ϕ of the superconducting order parameter in thin-film systems are affected differently by disorder and dimensionality. With increasing disorder superconducting long range order is quenched in sufficiently thin films by physical processes driven by phase fluctuations. This occurs at both the zero-field vortex-antivortex unbinding transition and at the zero-temperature magnetic-field-tuned superconducting-insulating transition. At both of these transitions Ψ_0 is finite and constant, vanishing only when temperature, disorder, and/or magnetic field are increased further. Experimental results on amorphous-composite InO_x films are presented to illustrate these points and appropriate comparisons are made to other experimental systems.

Introduction and thesis statement

There have been numerous studies[1] devoted to the competition between disorder and superconductivity. Disorder alone can induce localized eigenstates (the Anderson transition[2]) and coulomb interactions alone can give rise to localized electrons (the Mott transition[3]). It is the interplay of these localization and interaction effects which have a deleterious effect on superconductivity, sharply reducing the transition temperature and ultimately quenching all remnants of superconductivity. Particularly relevant is the dimensionality. Thus, for example, the inherent two-dimensionality of sufficiently thin films sets the stage for a delicate and marginally attractive pairing interaction from electronic states which are localized for arbitrarily weak disorder.

Experimentally, studies of superconductivity in disordered thin films are concerned with either granular[4] or homogeneously disordered[5-9] materials. For granular materials an energy gap or local order parameter develops near the bulk T_{c0} on each grain. The superconducting transition temperature T_c where long range order (LRO) appears is determined by the normal-state sheet resistance or, equivalently, the Josephson coupling between grains and can therefore be appreciably smaller than T_{c0}, The resistive transition of granular films usually takes place in two stages: the first comprising a sharp drop in resistance near T_{c0} where the individual grains become superconducting, and the second comprising a more gradual decrease towards the zero-resistance superconducting transition at T_c. Phase (ϕ) fluctuations, driven by thermally-suppressed intergranular Josephson currents, dominate in this latter regime[10]. On the other hand, for homogeneously disordered materials there is no obvious feature in the resistance transition which marks T_{c0}. The transitions are smooth and continuous, with a transition width which increases in proportion to the amount of disorder. Disorder is usually measured by the film resistivity for three-dimensional (3D) processes and the sheet resistance for two-dimensional (2D) processes. Thus, in contrast to granular films where Ψ_0 remains constant on each grain, as disorder increases both T_{c0} and Ψ_0 are continuously reduced to zero and it is Ψ_0-fluctuations which seem to be more important. This point of view is qualitatively supported by tunneling measurements which show a reduction in the density of states at the fermi energy occurring in proportion to the reduction in T_{c0}[11]. It has not been shown, however, that long range order (LRO) is destroyed by these same Ψ_0-fluctuations.

The central thesis of this work is that for sufficiently thin homogeneously disordered films the true superconducting phase transition (i.e., where LRO is established and the dc resistance is zero) is dominated by ϕ-fluctuations. This statement is certainly true for granular films where Ψ_0 remains relatively unchanged on each grain as disorder (intergranular coupling) is increased (decreased). For homogeneously disordered films this thesis presumes that Ψ_0, although strongly reduced from its clean-limit value, is non-zero at the superconducting transition[12], only to vanish when disorder, temperature, and/or magnetic field are further increased.

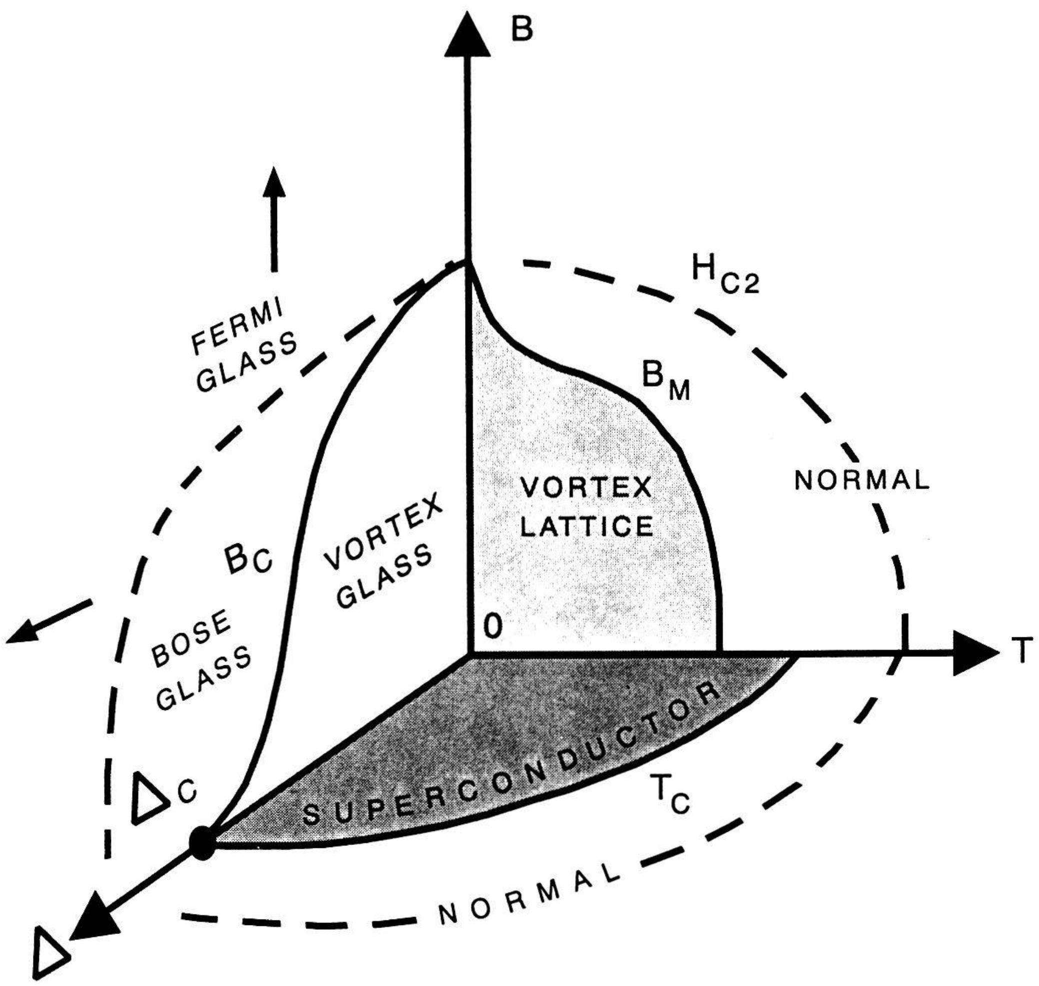

Fig. 1. Phase diagram for a homogeneously disordered 2D superconductor. The solid lines represent phase boundaries where long range order is quenched and the dashed lines delineate crossover regions.

Superconducting phase diagram in two dimensions

To illustrate and support these statements, we present in the following a discussion of experimental results on the 2D superconducting transitions of homogeneously disordered amorphous composite InO_x thin films. The point of reference for a theoretical interpretation of these results is embodied in the phase diagram[13] shown in Fig. 1 for a 2D superconductor. The amount of disorder Δ in a film is measured along an axis mutually perpendicular to the temperature, T, and magnetic field, B, axes. The magnetic field is applied perpendicular to the film surface. Superconducting phases with LRO, delineated in the figure by the solid lines, only occur on the BT, $T\Delta$, and $B\Delta$-planes. Thus, on the $T\Delta$-plane ($B = 0$) we have the vortex-antivortex Kosterlitz-Thouless transition[14] at T_c. At this transition, Ψ_0 is finite and LRO is destroyed by the phase fluctuations arising from the thermally induced unbinding of vortex pairs. At higher temperatures and/or greater disorder Ψ_0 is gradually suppressed to zero. This crossover (dashed line) to the fully normal state occurs near the mean-field transition temperature T_{c0} which in 2D is not precisely defined.

In like manner, on the $B\Delta$-plane ($T = 0$) there is a vortex-glass to bose-glass superconducting-insulating (S-I) transition at a critical field B_c[13]. At this transition Ψ_0 is again non-zero and LRO is destroyed by ϕ-fluctuations driven by the bose condensation of localized vortices into a single coherent quantum state with high resistance. Thus, for a given disorder Δ, an increasing magnetic field B induces a phase transition from a superconducting state of localized vortices and condensed pairs to an insulating state of localized pairs and condensed vortices. This duality at the transition involving two types of bosons (magnetic vortices and electron pairs) is a consequence of the boson Hamiltonian used to describe the dynamics of bosons in a random potential. There is also a crossover region on the $B\Delta$ plane (dashed line) where Ψ_0 is finally quenched to zero and the bose glass insulator comprising localized paired electrons transforms to a fermi-glass insulator comprising localized single electrons.

Characteristic length scales

Previously reported experimental results on InO_x films confirm the appropriateness of the phase diagram of Fig. 1 for both the $B = 0$ vortex-antivortex unbinding transition[15] and the $T = 0$ magnetic-field tuned superconductor-insulator transition[16]. Confirmation of the predicted behavior at each of these transitions requires that the longest length scales set by the experimentally available temperatures, currents, frequencies, and/or magnetic fields not be limited by sample nonuniformity. Thus the microscopic disorder must be uniform out to macroscopic length scales. For 100 Å-thick InO_x films these lengths have been found to be of the order of 10 μm for the vortex-antivortex unbinding transition[15] and 0.3 μm for the superconductor-insulator transition[16]. Agreement with theory out to these lengths implies that sample-related nonuniformities are not yet operative.

The characteristic length used in the scaling theory of the S-I transition is the superconducting coherence length ξ which diverges as $|\Delta_c - \Delta|^{-\nu}$ when disorder approaches criticality, $\Delta \to \Delta_c$. The variable Δ is assumed to have continuous behavior through the transition and the exponent ν is predicted to have a lower bound of unity. On the insulating side of the transition, ξ is a measure of the size of the superconducting regions over which the electron pairs are correlated whereas on the superconducting side of the transition, ξ sets the scale for fluctuations about a finite pair density. Interestingly, both T_c and B_c have straightforward functional dependences on ξ (i.e., $T_c \propto \xi^{-z}$ and $B_c \propto \xi^{-2}$) which lead to the relation,

$$B_c = A_1 T_c^{2/z} \quad , \qquad (1)$$

where A_1 is a constant and z is a dynamical exponent predicted to have a value of unity. Shown in Fig. 2 (solid line) is the experimental verification of this relationship for five 100 Å thick InO_x films prepared at different stages of disorder and for which T_c, B_c and T_{c0} have been independently determined[16]. The slope $2/z = 2.04 \pm 0.09$ is in excellent agreement with theoretical expectations.

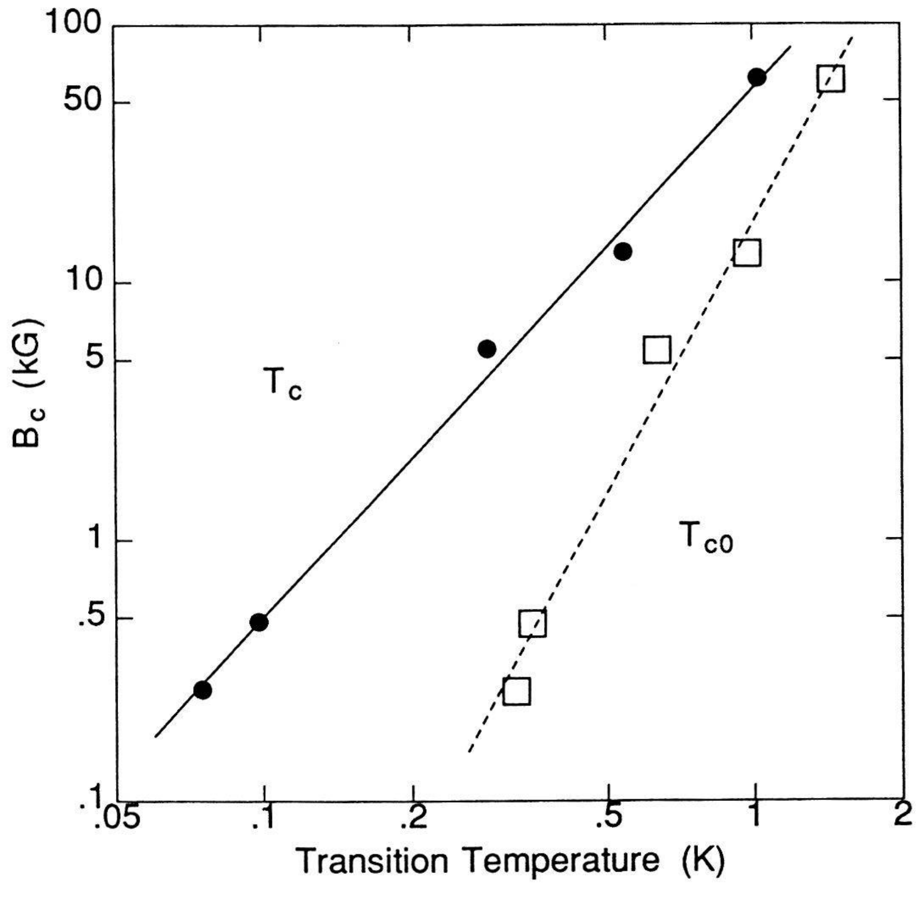

Fig. 2 Logarithmic plot of the critical field B_c versus transition temperature T_c (solid circles) and T_{c0} (open squares) for five 100-Å thick InO_x films. The regression fit solid line has a slope of 2.03 and the dashed line a slope of 3.49.

A quadratic relationship between B_c and T_{c0} rather than between B_c and T_c can be inferred from the expression, $\xi_o \propto v_F/T_{c0}$, for the Pippard length ξ_o. If we assume that the Fermi velocity v_F is relatively constant for different amounts of disorder and that the critical field scales as $B_c \propto \xi_o^{-2}$, then $B_c \propto T_{c0}^2$. Experimentally this dependence does not check as shown by the dashed-line fit to the data (□'s) in Fig. 2 which gives the more pronounced dependence $B_c \propto T_{c0}^{3.5}$.

Dependence of T_c and T_{c0} on normal-state properties

Since T_{c0} for films of different thickness depends on resistivity rather than sheet resistance[16-17] we conclude that the disorder-induced suppression of T_{c0} is 3D in character. This can be made more explicit by recalling the 3D scaling form for the temperature-dependent conductance $\sigma(T)$ which describes ($T > T_{c0}$) InO_x films, specifically,

$$\sigma(T) = (e^2/\hbar)[\xi_L^{-1} + A\ell_i^{-1}(T)] \qquad (2)$$

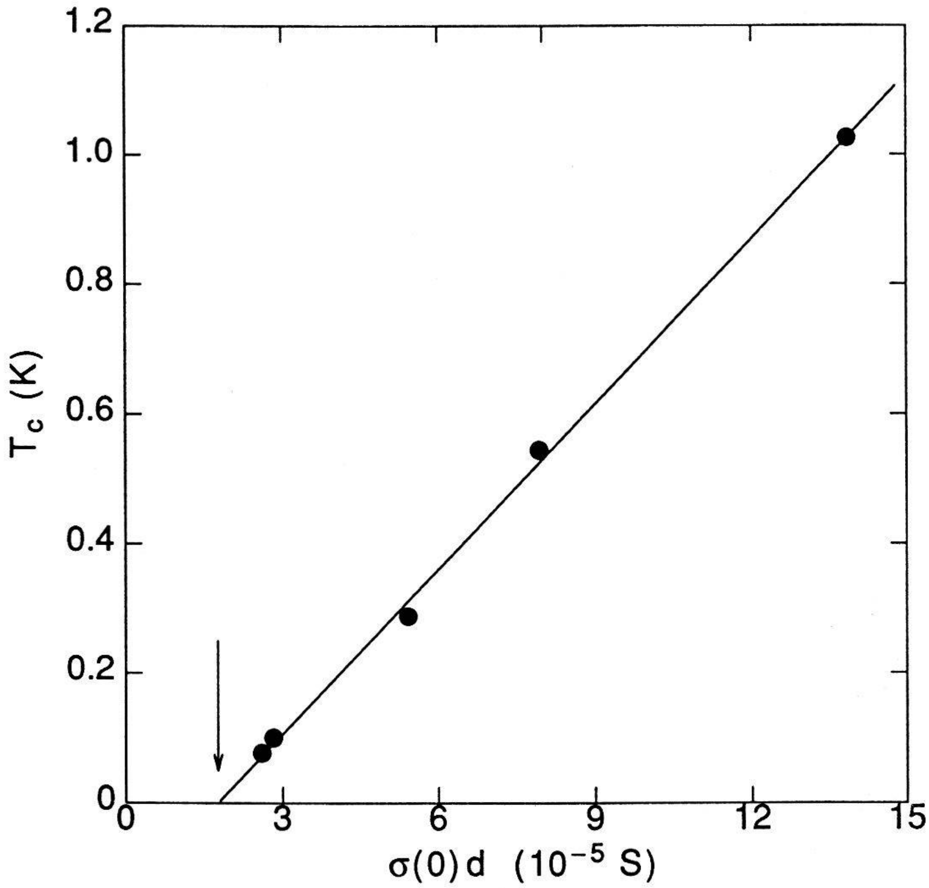

Fig. 3. Plot of T_c versus $\sigma(0)\,d$ for the five 100 Å-thick films shown in Fig. 2. The vertical arrow denotes critical disorder where $T_c = 0$.

where ξ_L is a localization length, A a constant and $\ell_i(T)$ the inelastic electron scattering length. As $\ell_i(T)$ has been experimentally shown to have a $T^{-1/4}$ temperature dependence[17-18], linear plots of $\sigma(T)$ vs $T^{1/4}$ allow estimates of $\sigma(0)$ for any given film. The extrapolated values of $\sigma(0)$ together with the measured values of T_{c0} reveal a linear dependence of T_{c0} on the ratio $\sigma(0)/\sigma(300K)$ which was found to extrapolate to the origin where T_{c0} and $\sigma(0)$ are simultaneously zero. Thus T_{c0} of InO_x films seems to vanish when disorder in 3D is sufficient to assure the 3D metal-insulating transition where $\sigma(0)$ is zero[17].

A revealing experimental dependence of T_c on the zero-temperature sheet conductance $G(0) = \sigma(0)d$ is shown in Fig. 3. for the same five 100 Å-thick films of Fig. 2. The solid line regression fit to the data can be parameterized by the equation

$$T_c = A_2 d\left[\sigma(0) - \sigma_c(0)\right] \quad , \tag{3}$$

where A_2 is a constant. As the thicknesses of the studied films were the same, we have explicitly retained d in this expression to emphasize the dependence of T_c on 2D (sheet conductance) rather than 3D (conductivity) quantities. Figure 3 and Eq. 2 thus imply that there is a critical disorder Δ_c, measured by the zero-temperature conductance $\sigma(0)d = 1.8\times10^{-5}$ S, at which T_c vanishes. Thus T_c is suppressed to zero at a critical disorder corresponding to, $\sigma(0)=\sigma_c(0)$, which is less than the critical disorder, $\sigma(0)=0$, where T_{c0} in previous work was found to be suppressed to zero. These observations are consistent with the statement that there is a finite gap or order parameter Ψ_0 at critical disorder Δ_c where $T_c = 0$.

A self consistency check

Equation 1 for $z = 1$ and Eq. 3 represent two separate and independent measurements which we now show are consistent with each other. We do this by writing each of these equations in the form $T_c \propto \xi^{-1}$ and comparing the numerical prefactors. For Eq. 1 this is easily done by making the substitution, $B_c = \xi^{-2}\Phi_0$, (Φ_0 is the flux quantum) to obtain,

$$T_c = \left[\Phi_0/A_1\right]^{1/2}\xi^{-1} = 1.93\times10^{-6}\xi^{-1} \quad , \tag{4}$$

where the experimentally determined value $A_1 = 5.55\times10^4\,\mathrm{G/K^2}$ was used. For Eq. 3 we make the substitutions $\sigma(0) = (e^2/\hbar)\xi_L^{-1}$ and $\sigma_c(0) = (e^2/\hbar)\xi_{Lc}^{-1}$ (cf. Eq. 2) to obtain $T_c = A_2 d(e^2/\hbar)[\xi_L^{-1} - \xi_{Lc}^{-1}]$ where ξ_{Lc} is the localization length corresponding to the critical zero-temperature conductivity where $T_c = 0$. With the plausible identification, $\xi^{-1} = \xi_L^{-1} - \xi_{Lc}^{-1}$, Eq. 3 becomes,

$$T_c = A_2 d(e^2/\hbar)\xi^{-1} = 2.07\times10^{-6}\xi^{-1} \quad , \tag{5}$$

where $A_2 d = 8.5 \times 10^{-3}$K cm/S has been determined experimentally from the slope of the solid-line regression fit to the data in Fig. 3. The coefficients of Eqs. 4 and 5 are fortuitously close to each other given the approximations involved. The agreement, however, is a good qualitative consistency check on the independent experimental measurements shown in Figs. 2 and 3 and described respectively by Eqs. 1 and 3.

Conclusions

There are cogent theoretical arguments that Ψ_0 is non zero when ϕ-fluctuations quench LRO in both granular and homogeneously disordered systems[1,10,12-14]. Experimental measurements on amorphous-composite InO_x films of both the vortex-antivortex and superconducting-insulating transitions show good agreement with the theories which make this assumption. The correlation of T_c and T_{c0} with $\sigma(0)$ which shows that T_{c0} is finite when T_c is suppressed to zero at Δ_c provides additional evidence for the correctness of this viewpoint. These results on InO_x are by no means observed in all homogeneously disordered superconductors. Part of the reason may be that there are other phase slip processes[19] which have heretofore not been taken into account and which might dominate in films with different microstructure, carrier density, or interfacial energies. Such films may even be in different universality classes. Also there are multitudinous manifestations of microscopic disorder which are not amenable to detailed experimental characterization. Any given microstructure which appears to be homogeneous might well have underlying nonuniformities with associated length scales which impose unrecognized bounds on the finite lengths set by external experimental probes.

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