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Percolation-localization crossover and depression of the superconducting transition temperature in films of metal-insulator mixtures

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Recent experiments on the conductivity and the onset of superconductivity in thin metal-insulator-mixture films are interpreted in terms of an ansatz which provides a criterion for the crossover between percolation and localization. In agreement with experiment, this ansatz predicts that percolation dominates in large-grain structures and localization in amorphous ones, providing a unified view of the apparently disparate results on different types of metal-insulator mixtures.

A puzzling aspect of our research on metal-insulator mixture films has been the apparent diversity of the mechanisms needed to explain the metal-insulator (M-I) transition. While percolation appears to be the driving mechanism for the transition in In-Ge, Pb-Ge, and Al-Ge,[1,2] localization can be identified as responsible for the M-I transition in quenched-condensed Bi-Kr (Ref. 3) and both mechanisms evidently play a role in quench-condensed Hg-Xe.[4] In this Rapid Communication we describe a means of reconciling these apparently diverse results in the context of a model which considers the crossover between percolation and localization. The latter is an outstanding issue in the description of the M-I transition. With the use of the model, the differences between the data of our different groups can be understood by considering both the behavior of the electrical resistivity and the superconducting transition temperature.

We assume that all of the above systems are disordered and random, albeit on different length scales. This claim is supported by direct electron microscopy observation for In-Ge, Pb-Ge, and Al-Ge, which have grain sizes the order of 200 Å.[1,2] Quenched-condensed Bi films are known to be amorphous.[5] Hence, it is reasonable to assume that Bi-Kr mixtures are amorphous as well, since it is well known that impurities tend to stabilize amorphous phases. The characteristic length scale for disorder in that case is the interatomic distance. Quench-condensed Hg films have been observed to be polycrystalline, with small crystallites.[6] Although there is no direct evidence as to the structure of Hg-Xe mixture films, as discussed below, their resistivities and the critical concentration for the M-I transition are consistent with a structure consisting of a random network of random crystallites. In that case the length scale for disorder would be intermediate between that of the large-grained and amorphous systems.

We observe that the threshold concentration on the critical concentration for the M-I transition is close to that predicted for percolation in a random continuum in three dimensions (3D)—about 15 vol.%—for In-Ge, Pb-Ge, and Hg-Xe films, but is much higher for Bi-Kr films (43 vol.%). Related differences occur in the superconducting properties. While the superconducting transition temperature T_c (determined as the 50% point of the resistive transition) remains almost constant down to the threshold concentration for the large-grained systems, there is a more progressive drop in Hg-Xe films commencing at a higher concentration, and in Bi-Kr films the drop in T_c is already present at very small Kr concentrations. Percolation, being a purely classical phenomenon, does not by itself affect T_c. On the other hand, electronic localization is known to weaken superconducting properties.[7] Hence, we can already state qualitatively that localization effects marked by an increase in the critical volume fraction and a reduction of T_c appear progressively as the elementary scale for disorder becomes small.

It is useful to present the data from these diverse systems in a unified way. In Fig. 1 we plot T_c/T_{c0} as a function of $\log_{10}(\rho/\rho_0)$, where ρ_0 is the resistivity of the undoped metal. For the case of Hg-Xe a very large resistivity increase is needed before T_c drops. On the other hand, for Bi-Kr the ratio T_c/T_{c0} falls rapidly with increasing ρ. For Pb-Ge, T_c/T_{c0} hardly changes over the entire range of resistivities over which the samples are conducting. Superconductivity disappears abruptly at the percolation concentration. The decrease of T_c as has been mentioned can be associated with localization-like behavior. Consequently Bi-Kr is always in the localized regime, Hg-Xe would appear to exhibit a crossover from the percolation to the localization regime with increasing Xe doping, and Pb-Ge is the percolation regime except negligibly close to the percolation threshold.

We can now attempt to "calculate" the T_c/T_{c0} vs $\log_{10}\rho$ curves of Fig. 1, using the idea that there are different characteristic minimum length scales for each material system over which a resistivity ρ_0 can be defined. The approach is similar to that of Khmelnitskii[8] in describing localization in percolative structures. The central idea of the proposed picture is that as the M-I transition is approached, as a function of increasing dilution, percolation crosses over to localization. In the model this will occur when the percolation correlation length at the localization transition is smaller than the localization correlation length. Thus localization sets in before the classical percolation threshold is reached. The importance of localization effects in a percolative structure can be seen by considering the normalized resistance R_{ξ_p} of a cube of size ξ_p measured in units of $\hbar/e^2$. Here ξ_p is the percolation correlation length given by $\xi_p = L(x - x_{cp})^{-\nu_p}$, where x is the volume fraction of metal, x_{cp} is the value of x at the classical percolation threshold, and L is the minimum length scale over which a resistivity ρ_0 may be defined. The latter is a characteristic of the particular structure. The Anderson transition is then assumed

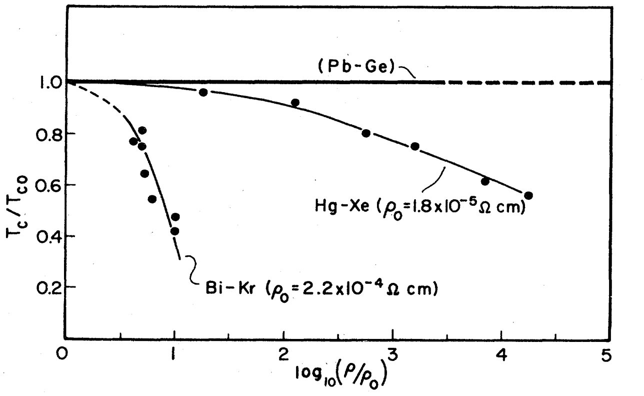

FIG. 1. Plot of the reduction of the superconducting transition temperature T_c/T_{c0} (resulting from doping) as a function of $\log_{10}(\rho/\rho_0)$. The data for Pb-Ge, Bi-Kr, and Hg-Xe are from Refs. 1, 3, and 4, respectively.

to take place when

$$R_{\xi_p} = \left[\frac{e^2}{\hbar}\rho_0(x-x_{\mathrm{cp}})^{-\mu}\right]\Big/\xi_p \tag{1}$$

is the order of unity. This follows directly from the scaling arguments presented by Abrahams, Anderson, Licciardello, and Ramakrishnan.[9] Taking into account that on the percolation infinite cluster the resistivity is known to be a decreasing function of the length scale up to ξ_p. Hence, if the 3D electronic wave functions are localized on a scale smaller than or equal to ξ_p, then they must necessarily be localized on any larger macroscopic length scale. On the other hand, if they are not localized on the scale ξ_p they will not be localized on any larger length scale. In three dimensions $\mu=2$ and $\nu_p=0.88$.[10] The value of x at the Anderson transition x_c is then given by solving Eq. (1) with $R_{\xi_p}=1$.

$$x_c - x_{\mathrm{cp}} = y^{1/(\mu-\nu_p)} \quad . \tag{2}$$

Here we have defined $y=\rho_0 e^2/L\hbar$. The magnitude of this parameter we will show determines the location of the crossover between percolation and localization.

We now implement the central idea by making the ansatz that the relevant consideration for the description of the percolation-localization crossover is the magnitude of the parameter is $z=\xi_l/\xi_p(x=x_c)$ where ξ_l is the localization correlation length.[11] Also in the limit where the effective superconducting coherence length $\xi_s>\xi_l$, ξ_p, the magnitude of z determines whether there is a weakening of superconductivity. With $\xi_l=L(x-x_c)^{-\nu_l}$ we obtain

$$z = y^{\nu_p/(\mu-\nu_p)}(x-x_c)^{-\nu_L} \quad . \tag{3}$$

These considerations are summarized in the schematic representation of the phase diagram for the M-I transition which is given in Fig. 2. The solid line corresponds to Eq. (2) and the slanted dashed line shows the condition $z \sim 1$. We can now use scaling to eliminate concentration in favor of resistivity in the condition for the crossover between percolation and localization. There are two limits of interest.

(i) $z \ll 1$. The resistance of the sample in the metallic phase is dominated by percolation where $\rho=\rho_0(x-x_c)^{-\mu}$.

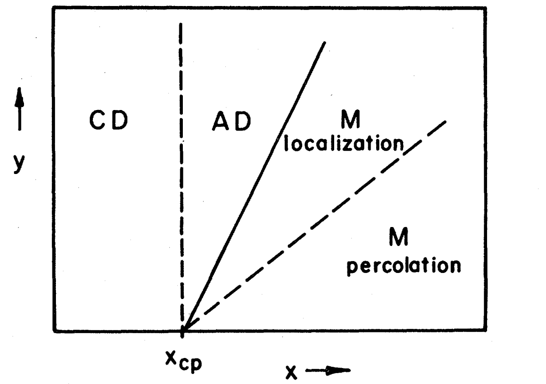

FIG. 2. Schematic diagram of the M-I transition in a system in which there is a percolation localization crossover. The M-I phase boundary is the solid line given by Eq. (2) and the slanted dashed line, which is a measure of the percolation-localization crossover in the metallic phase, roughly corresponds to $z \sim 1$. In this figure, following Ref. 7, AD, CD, and M refer to the Anderson dielectric, classical dielectric, and metallic phases, respectively.

Hence,

$$z = y^{\nu_p/(\mu-\nu_p)}(\rho/\rho_0)^{\nu_l/\mu} \quad . \tag{4}$$

(ii) $z \gg 1$. Localization effects dominate, and with $\rho=\rho_0(x-x_c)^{-\nu_l}$

$$z = y^{\nu\rho_p/(\mu-\nu_p)}(\rho/\rho_0) \quad . \tag{5}$$

Is should be noted that for 2D percolating systems (films with thicknesses smaller than ξ_p) these results are replaced by

$$z = y^{\nu_p/\mu}(\rho/\rho_0)^{\nu_l/\mu} \quad z \ll 1 \quad , \tag{4$'$}$$

$$z = y^{\nu_p/\mu}(\rho/\rho_0) \quad z \gg 1 \quad . \tag{5$'$}$$

If $\nu_l=1$, and since $\mu=1.3$ in 2D, the difference in behavior between the two limits is much less pronounced than in the 3D case.

We can now discuss the behavior of the conductivity and the transition temperature depressions observed in the various measurements in light of the above scaling results, bearing in mind that ν_l should be in the range $0.5<\nu_l<1$.[11] For the large grained structures such as Pb-Ge our estimates for L and ρ_0 yield for y the value 1×10^{-4}. According to Eq. (4) a value of ρ the order of 1 Ω cm or greater would then be required for crossover to the localization regime. As observed experimentally, percolation dominates in practice over the entire range of concentrations. The facts that the M-I transition occurs at x_{cp} and the conductivity exponent has the percolation value ~ 2, together with the drop in T_c, signify that the onset of the localization occurs abruptly, *negligibly* close to the percolation threshold.

In the original analysis of the concentration dependence of the resistivity of quenched-condensed Hg-Xe films there were difficulties in obtaining a single conductivity exponent. Over a wide range of concentrations above x_c the data could be fit with an exponent of 1.6, a value close to, but somewhat smaller than the percolation value. On the other hand, a fit carried out very close to x_c gave an exponent somewhat smaller than unity. These results imply a crossover from one value of the exponent to another as a function of increasing Xe doping. The absence of any discontinuous change in the resistivity as a function of Xe doping suggests that the change in the behavior is not associated with a crystallographic change in the sample such as from a random-crystalline to an amorphous structure. For the He-Xe system we estimate $y=2\times10^{-2}$, taking $\rho_0=1.8\times10^{-5}$ Ω cm and $L=2.0\times10^{-7}$ cm. The former is the resistivity of a pure Hg film. The latter has been taken to be the initial thickness at which films of various concentrations become continuous. It is a reasonable estimate of the length scale. Once again $z \ll 1$ in the slightly doped limit. Consequently using Eq. (4) the M-I transition is expected to occur very near the percolation concentration x_{cp}. However, in contrast with the case of the large-grained structures, the model predicts a percolation-localization crossover before x_{cp} and a depression of T_c associated with the onset of the localization regime over an accessible range of concentrations. The condition $z \sim 1$ is met for $\rho/\rho_0 \sim 460$ or $\log_{10}(\rho/\rho_0) \sim 2.7$ assuming $\nu_L=1$, $\mu=2$, and $\nu_p=0.88$. This value of the logarithm is in rough agreement with the value of Fig. 1 at which T_c begins to fall. If ν_L were close to 0.5 the falloff would occur at $\log_{10}(\rho/\rho_0)=5.4$ which is beyond the range of the data for Hg-Xe. These results suggest that ν_L is closer to 1 than to 0.5.

Finally, for Bi-Kr we obtain $y \simeq 1$, as L is the order of atomic dimensions and ρ_0 is two orders of magnitude smaller than the corresponding result for Pb-Ge. In this case, localization dominates at all concentrations. Indeed, x_c is now much larger than x_{cp}, consistent with the considerations in Fig. 2. The condition $z \approx 1$ is met for $\rho \approx \rho_0$, i.e., T_c drops immediately starting from the slightly doped value. The resistivity data can be fit with an exponent ν_L close to unity which is very different from the percolation exponent. This value of ν_L is also consistent with the qualitative value inferred from the analysis of the percolation-localization crossover in the Hg-Xe system.

In conclusion, the proposed scaling form for the crossover allows a comprehensive qualitative description of all of our data on 3D random-mixture films. It is possible to understand the differences between the various systems as to the composition dependence of their superconducting transition temperatures in terms of this scaling model of the percolation-localization crossover. For very small values of the parameter y all of the localization, and associated reduction of T_c, occurs negligibly close to the percolation threshold, at very high sample resistivity. As the parameter y increases the crossover occurs further from the percolation threshold, at decreasing values of the resistivity. For large values of y, such as those found in amorphous systems, the effects of localization may be evident even in the undiluted material. We have excluded from this analysis granular films for which x_{cp} is shifted from the value for percolation in a random continuum because of the lack of topological symmetry between the metal and the insulator, and for which charging effects may be important. The direct fit in the case of Bi-Kr, and the comparison in the case of Hg-Xe between the condition for the crossover between percolation and localization in the scaling approach and the data imply that ν_L is closer to 1 than to 0.5. The analysis suggests that it may be fruitful to undertake more detailed studies of mixture systems, attempting to measure the crossover critical exponents in the context of a more elaborate scaling theory. It would also be useful to study the dependence of the resistivity at the percolation-localization crossover on ρ_0 and L, the latter being determined by the metallic cluster size which may be determined spectroscopically.[12]

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