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Upper Critical Field of a Percolating Superconductor

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The upper critical field H_{c2} of the random percolating superconductor InGe has been measured as a function of the metal volume fraction x. Near the percolation threshold x_c, H_{c2} diverges with a critical exponent which is significantly smaller than that of the normal-state resistivity. An interpretation of this behavior is proposed in terms of the properties of the infinite cluster.

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It has long been recognized that the high upper critical field H_{c2} of dirty superconducting alloys is due to the existence of a short impurity-limited mean free path[1] rather than to macroscopic inhomogeneities.[2] Early work on solid-solution alloys[3] demonstrated that, in agreement with theoretical predictions, H_{c2} increases linearly with the normal-state resistivity ρ_n of the alloy (itself proportional to the impurity concentration). In this Letter, we wish to report on the critical behavior of a different class of superconducting alloys, consisting of unmiscible metal and insulator distributed at random.[4] In contrast with the case of solid solutions, we find that the inhomogeneity of these alloys has a determining influence on their upper critical field.

We find that near the percolation threshold[5] where ρ_n becomes much larger than the values typical of solid solutions (i.e., $\rho_n \gtrsim 50\ \mu\Omega$ cm), $H_{c2} \propto \rho_n{}^{\omega}$ with $\omega = 0.5 \pm 0.05$, in contrast with the behavior of solid solutions. We interpret this result as due to the diffusion process on a percolating network, for the case where the percolation correlation length ξ_p is of the order of or larger than the effective superconducting coherence length $\bar{\xi}_s$. We propose that the value of $\omega < 1$ results from the fact that in this limit the dead ends of the infinite cluster contribute very little to its superconducting properties. The experimental data suggest that the effective superconducting density of the infinite cluster is close to that of its backbone.

Samples were prepared by coevaporation of indium and germanium from two distinct electron beam guns, spaced apart, onto a room-temperature glass substrate. Nine samples, about 2000 Å in thickness, were simultaneously evaporated onto one glass substrate, their metal concentrations varying by about 2% between neighboring samples.[5] The pressure during the evaporation was held at approximately 1×10^{-6} Torr and never exceeded 5×10^{-6} Torr. In-Ge films prepared in this way have a random (rather than granular) structure[4] with both constituents being crystalline, in contrast with most cermet films.[6] Their percolation threshold is close to 15 vol.%, in agreement with theoretical predictions for percolation in a random continuous medium and the In and Ge clusters have the ramified structure characteristic of percolating systems.[4,5]

For samples with $\rho_n \lesssim 1000$ $\mu\Omega$ cm, the superconducting resistive transition in zero field $R(T)$ was fairly sharp, the 90% to 10% drop taking place typically in a few times 10^{-2} K (Fig. 1). For higher resistivities, a significant drop of T_c was observed and the transitions became very broad. For $\rho_n \lesssim 1000$ $\mu\Omega$ cm, the resistive transitions in an applied field $R(H)$ were typical of type-II high-κ thin films with a width of the order of 30%.[7]

The parallel ($H_\parallel$) and the perpendicular ($H_\perp$) critical fields were measured as functions of temperature, the critical field being defined as the field at which half of the normal-state resistance was restored. For $10 < \rho_n < 35$ $\mu\Omega$ cm the anisotropy ratio $H_\parallel/H_\perp$ was found to be somewhat higher[7] than the value $H_{c3}/H_{c2} = 1.69$ expected for thick $[d \gg \xi(T)]$ type-II films.[8] For $\rho_n \gtrsim 50$ $\mu\Omega$ cm the anisotropy was found to decrease and to reach a value of the order of 30% which we understand as resulting from the internal, randomly oriented, In/Ge interfaces as opposed to the external surfaces of the film which lead to the anisotropy ratio of 1.69.

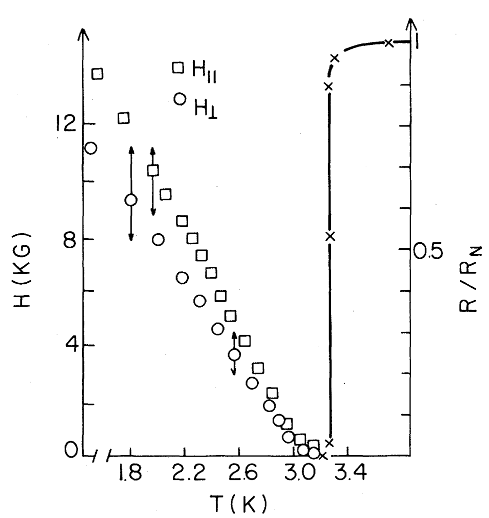

FIG. 1. Resistive transition and critical fields of a sample with $\rho_n = 300$ $\mu\Omega$ cm. The transition temperature is about 0.1 K lower than that of pure In; the lower end of the resistive transition presents a small tail; the temperature dependence of $H_\perp(T)$ presents an upward curvature very close to T_c. These features are more pronounced in samples with $\rho_n \gtrsim 1000$ $\mu\Omega$ cm. The arrows indicate the 10%–90% transition widths.

Figure 2 gives the variation of $(dH_\perp/dT)_{T=T_c}$ as a function of $\rho_n \lesssim 50$ $\mu\Omega$ cm. Again, there is clearly a change of behavior around $\rho_n \simeq 50$ $\mu\Omega$ cm. For $\rho_n \lesssim 50$ $\mu\Omega$ cm, the data points can be fitted to a straight line, with a slope [65 Oe/($\mu\Omega$ cm)] about 20% higher than would be expected for a type-II In based alloy.[3] For $\rho_n \gtrsim 50$ $\mu\Omega$ cm the data fit fairly well a $\rho_n^{1/2}$ variation. In this high-resistivity regime, the respective variations of $(dH_\perp/dT)_{T=T_c}$ and of ρ_n as functions of $x - x_c$ are displayed in Fig. 3. They follow distinct power laws, $(dH_\perp/dT)_{T=T_c} \propto (x-x)^{-k}$ with $k = 0.6 \pm 0.05$, and $\rho_n \propto (x-x)^{-t}$ with $t = 1.2 \pm 0.1$ ($x_c = 15\%$). The value of t is close to what is expected for a two-dimensional percolating system, in agreement with previous results on similar films.[9]

We interpret the change in behavior of the anisotropy ratio $H_\parallel/H_\perp$ and of the amplitude of $H_\perp$, which we observe around $\rho_n \simeq 50$ $\mu\Omega$ cm, as a crossover between two regimes: a quasihomogeneous type-II regime at low resistivities and a percolation-dominated regime at high resistivities near the percolation threshold. We propose that this crossover occurs when the effective superconducting coherence length of the medium $\bar{\xi}_s$ is of the order of the percolation correlation length ξ_p. When $\bar{\xi}_s \gg \xi_p$, the medium is effectively homogeneous on the scale of $\bar{\xi}_s$ and the structure of the infinite percolation cluster is unimportant for the superconducting properties. This is the regime where $H_\parallel/H_\perp \propto H_{c3}/H_{c2}$ and $(dH_\perp/dT)_{T=T_c} \propto \rho_n$. In the opposite limit, the

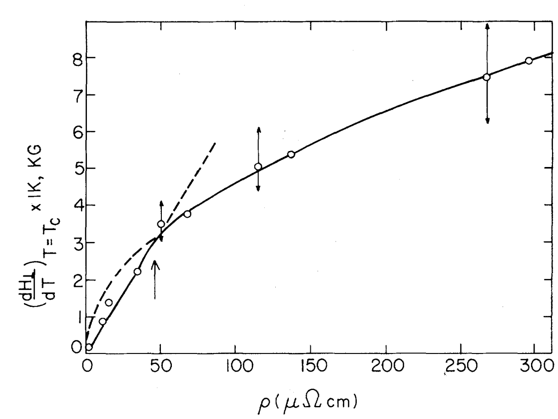

FIG. 2. Critical field as a function of the resistivity ρ_n. $(dH/dT)_{T=T_c}$ measures the slope of the linear variation of $H_\perp$ vs T near T_c (see Fig. 1). The arrows indicate the 10%–90% transition widths.

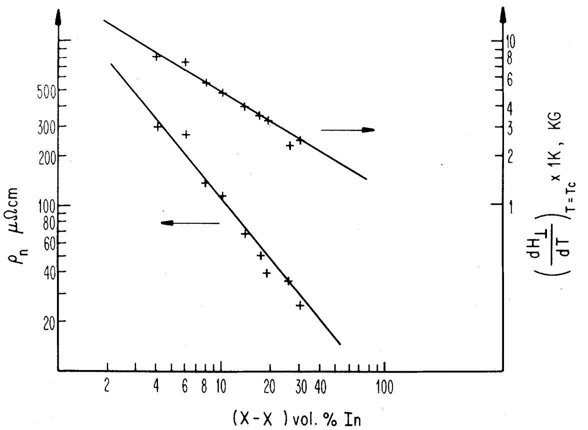

FIG. 3. Concentration dependence of the critical field and of the resistivity near the percolation threshold.

structure of the infinite cluster is important: In/Ge interfaces become effective internal boundaries, thus lowering the anisotropy ratio; the topology has a direct influence on the value of the upper critical field.

By using $\overline{\xi}_s^{\,2} = \varphi_0/2\, H_\perp$ and $\xi_p = d(x-x_c)^{-\nu}$ we can estimate the value of $x = x_{\rm cr}$ at the crossover. With $d \simeq 150$ Å, $\nu = \nu_3 = 0.85$, we obtain $x_{\rm cr} \simeq 0.30$ and $\rho_n(x = x_{\rm cr}) \simeq 25$ $\mu\Omega$ cm, in fair agreement with the experiments.[10]

The quasihomogeneous limit result $(dH_\perp/dT)_{T=T_c}$ can be formally obtained by noting that at the upper critical field H_{c2} the superconducting order parameter Δ is solution of a Landau-Ginsburg equation:

$$A(T-T_c)\Delta + D(i\,\nabla - 2e\hbar\vec{A}/c)^2\Delta = 0. \qquad (1)$$

In the limit $\overline{\xi}_s \gg \xi_p$ the energy density coefficient A must be proportional to the density of the entire infinite cluster, $A \propto P_\infty \propto (x-x_c)^B$ in the notation of Ref. 5. In the same limit, D measures the long-time diffusion on the infinite cluster, $D = (\langle r^2(\tau)\rangle/\tau)_{\tau\to\infty}$. This is given by $D = D_0(x-x_c)^{t+\beta}$,[11] where D_0 is the diffusion coefficient along an uninterrupted chain of In grains. The resulting coherence length is then given by

$$\overline{\xi}_s^{\,2} \propto (D/A) \propto (x-x_c)^t, \qquad (2)$$

and

$$H_{c2} = (\varphi_0/2\pi)\xi_s^{\,2} \propto (x-x_c)^{-t} \propto \rho_n, \qquad (3)$$

as expected.

The situation is more delicate when $\overline{\xi}_s$ approaches ξ_p. Two effects should then in principle be taken into account: (i) Long dead ends of the infinite cluster (along which $|\Delta|$ is not directly affected by the applied field because their diameter d is small compared to the London penetration depth) become effectively decoupled from the backbone, whole loops sustain all the Meissner currents. This reduces the value of the density coefficient A and hence the value of H_{c2}, compared to the quasihomogeneous-limit result. It is tempting to take $A \propto (x-x_c)\beta'$, with β' the density index for the backbone.[12] (ii) The value of D may be modified because, on the (reduced) scale of $\overline{\xi}_s$, it may depart from its $\tau\to\infty$ value.

The experiment suggests that, at least for not too high values of ρ_n, this second effect is not very important and $H_{c2} \propto (x-x_c)^{-t+\beta-\beta'}$: The experimentally measured value of $t = 1.2 + 0.1$ and the two-dimensional values $\beta = 0.14$, $\beta' = 0.55 \pm 0.05$ Ref. (12) give $k = 0.74 \pm 0.15$ in reasonable agreement with the measured value $k = 0.60 \pm 0.05$. We note, however, that the validity of this assumption is by no means obvious. It is consistent with the nodes-and-links model of the infinite cluster,[13] but not with the gasket model of Gefen *et al.*[14]

Finally, for alloy concentrations very close to x_c, we have $\xi_p \gg \overline{\xi}_s$ since $\xi_p \to \infty$. However, the meaning of $\overline{\xi}_s$ in this limit is not clear. In particular it becomes position dependent since on a scale much smaller than ξ_p the infinite cluster appears highly inhomogeneous. What probably happens in this regime is that superconductivity is quenched in a nonuniform fashion. The better connected parts of the cluster, with a fairly high local value of D, are quenched first. Long single strands, if they exist—and recent results[15] as well as the qualitative agreement of the critical-field data with the nodes-and-links model suggest that they do—remain superconducting up to higher fields. On these strands, one-dimensional fluctuation effects become important. In short, the alloy appears macroscopically inhomogeneous. This is probably the behavior observed in the higher-resistivity samples, $\rho_n \gtrsim 1000$ $\mu\Omega$ cm.

In conclusion, we have shown that the upper critical field of a superconducting alloy is sensitive to the topology of the infinite cluster. Critical-field measurements can be used to check specific models of the infinite cluster. The nodes-and-links model, which completely neglects the fact that at short times diffusion is faster than at long times, seems to be in better agreement with the experiment than the gasket model. The latter emphasizes these short-time effects and assumes that no long single strands exist near

x_c. The critical-field experiments indicate that this feature should not be neglected in any realistic picture of the infinite cluster.

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Additional Boundary Conditions: Critical Comparison between Theory and Experiment

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A linear-response theory leads to a generalized additional boundary condition (ABC) which includes as special cases all the well-known ABC's. Calculations for the reflectivity are compared with an experimental spectrum of CdS. Strong evidence is found in favor of the Pekar ABC and against other ABC's. Comparison with an attenuated total reflectivity spectrum of ZnO suggests that a more sophisticated surface response is necessary for semiconductors with a small exciton-free surface layer.

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Certain semiconductors exhibit anomalous optical properties associated with excitonic transitions. These properties are a manifestation of the translational motion of the exciton. They are described by nonlocal models: Namely the dielectric function ϵ depends on the wave vector $\vec{q}$, as well as on the frequency ω. As a consequence calculations of optical properties of these materials necessitate an additional boundary condition (ABC) which supplements those normally applied for electromagnetic fields at the interface between spatially nondispersive (local) media. The nature of this ABC has been under intense discussion for the past ten years.

The ABC's are usually stated in terms of constraints on the excitonic polarization $\vec{P}(z)$ and its normal derivative $\vec{P}'(z)$ at the crystalline surface or at the interface between the bulk and an exciton-free surface layer. Pekar[1] originally suggested $\vec{P} = 0$. Other frequently used ABC's are

