Upper Critical Field of Superconducting Pb Films above and below the Percolation Threshold

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The upper critical field of percolating lead films above and below the percolation threshold has been measured by monitoring the sample's electrical resistance as a function of an applied magnetic field. We find that it remains constant across the threshold, as predicted by theory based on the fractal nature of the infinite clusters and of large finite clusters.

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The upper critical field of percolating superconductors has been the subject of intensive interest during the last years. This research became even more important now because of its possible applications to ceramic high-T_c superconductors. The understanding of superconducting properties of random superconducting media is based on the distinction between the homogeneous and inhomogeneous limits.[1] The boundary between the two regimes is defined by the condition $\bar{\xi}_s = \xi_p$, where $\bar{\xi}_s$ (Refs. 2 and 3) is the effective superconducting coherence length of the medium, and ξ_p is the percolation correlation length. In the limit $\bar{\xi}_s > \xi_p$ the medium is effectively homogeneous on the scale of $\bar{\xi}_s$ and the structure of the infinite cluster is unimportant for the superconducting properties. The properties of the samples in this limit are predicted to be similar to those of dirty type-II superconductors: H_{c2} is proportional to the normal-state resistivity and increases linearly with T near T_c. In the opposite limit the topology of the sample has a direct influence on the upper critical field. A saturation of H_{c2} and an anomalous power-law temperature dependence near T_c are expected in this fractal regime $\bar{\xi}_s < \xi_p$, and have been observed experimentally in three-dimensional In-Ge samples.[4] No experimental data have yet been published on the superconducting properties of percolating superconductors below the percolation threshold. We want to report here for the first time results of measurements of the upper critical field of two-dimensional percolating films of lead both above and below the percolation threshold.

Samples were prepared by electron-beam deposition of lead on preevaporated 400-Å-thick layers of germanium at room temperature. Nine samples were simultaneously evaporated, their lead coverage varying slightly as a result of their different geometrical locations. Pressure during the evaporation was $(1-2)\times 10^{-6}$ Torr; the deposition rate was about 20 Å/sec. A typical film thickness when it becomes continuous under these deposition conditions is about 110 Å. Lead films prepared in this way have been shown[5] to have a two-dimensional percolative structure. In order to prevent the films from destruction by oxidation in atmosphere they were covered by an additional layer of 400 Å of germanium. (Results reported on samples uncovered by an additional Ge layer and measured immediately after their removal from the evaporation chamber are qualitatively similar.)

All samples were evaporated under similar conditions, their room-temperature resistivities varying from about 10 Ω/sq up to about 100 kΩ/sq, which corresponds to about 1 Ω/sq up to a few GΩ/sq slightly above the critical temperature, defined below as the normal-state resistivity.

When the metal coverage is above critical (about 50%), an infinite cluster develops across the sample. These samples have a metal-like resistivity temperature dependence $dR/dT > 0$ at temperatures above T_c and reach zero resistance below T_c. Samples very close to the percolation threshold have weak links in the infinite cluster. These samples also reach zero resistivity below T_c but their behavior above T_c is semiconductorlike $dR/dT < 0$. The maximum normal resistivity above T_c for which we have observed a zero resistivity in the superconducting state is 6.25 kΩ/sq. This value confirms again that predicted[6,7] and previously observed in quench-condensed tin[8,9] and other thin films[10-12] as the universal value of the critical resistance of a single Josephson junction R_Q, above which the junction remains resistive down to temperatures $T \ll T_c$.

Investigation of the samples below the percolation threshold in a transmission electron microscope confirms a fractal morphology with finite clusters only. The grain size is approximately 200 Å and clusters sizes range up to 1 μm slightly below the threshold. Intercluster distances can be as small as a few tens of angstroms. The conductivity in this case is dominated by quasiparticle tunneling between clusters.

At average thickness slightly below the "random" percolation threshold where no metallic infinite cluster is developed, the samples display the so-called reentrant behavior.[13-15] The resistance of these films increases with decreasing temperatures above T_c; there is a local resistance minima or "kink" below T_c and then a rapid increase of the resistance as the temperature is decreased further. At smaller thicknesses the resistance decreases

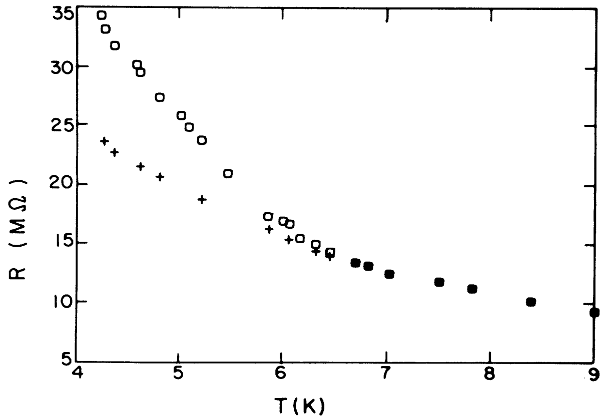

FIG. 1. Resistance of the sample below the percolation threshold as a function of temperature measured at 0- (squares) and 40-kG (crosses) magnetic field.

with temperature in the entire temperature range.

We present in Fig. 1 a typical dc resistance behavior of a Pb sample below the percolation threshold measured as a function of temperature respectively in 0- and 40-kG magnetic field (at which superconductivity is quenched). The two curves coincide at high temperature and deviate strongly below the superconducting transition temperature (essentially that of bulk lead). As shown Fig. 2, the superconducting transition of finite clusters in a discontinuous film can be monitored by the direct measurement of the sample's resistivity as a function of an applied magnetic field. Unlike the usual superconducting transition of a sample above the percolation threshold (dotted line in Fig. 2), the resistivity of a discontinuous film is maximum when the metal is in the superconducting state and gradually decreases down to a constant value as the clusters return to the normal state. While the return to the normal state of the infinite cluster above threshold is related to loss of coherence on the scale $\bar{\xi}_s$, we relate the opposite behavior of the magnetoresistance below the threshold to the progressive reduction of the order parameter in the finite clusters. Below threshold, the conductivity of the sample is dominated by intercluster quasiparticle tunneling. The conductivity in the normal state (*N*-*I*-*N* junction) is then larger than when the clusters are superconducting (*S*-*I*-*S* junctions), due to the opening of the superconducting energy gap.[16] Reduction of this gap by the applied magnetic field leads to the progressive reduction of the resistance to its normal-state value. This value is restored at a field H_{c2}^* when the order parameter in the cluster is reduced to zero.

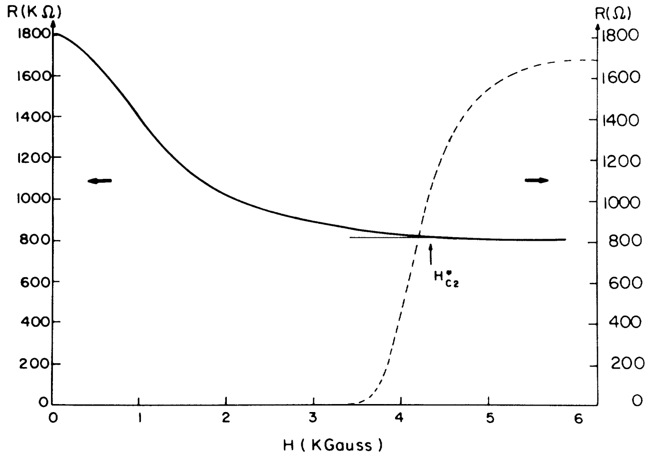

FIG. 2. Resistance as a function of applied magnetic field: solid line, sample below the percolation threshold; dotted line, sample above the percolation threshold. Upper critical field of the sample below the threshold H_{c2}^* is defined as a field at which $R(H)$ becomes saturated. Temperature 4.2 K.

We define H_{c2}^* as the field at which the sample's resistance deviates measurably from its field-independent

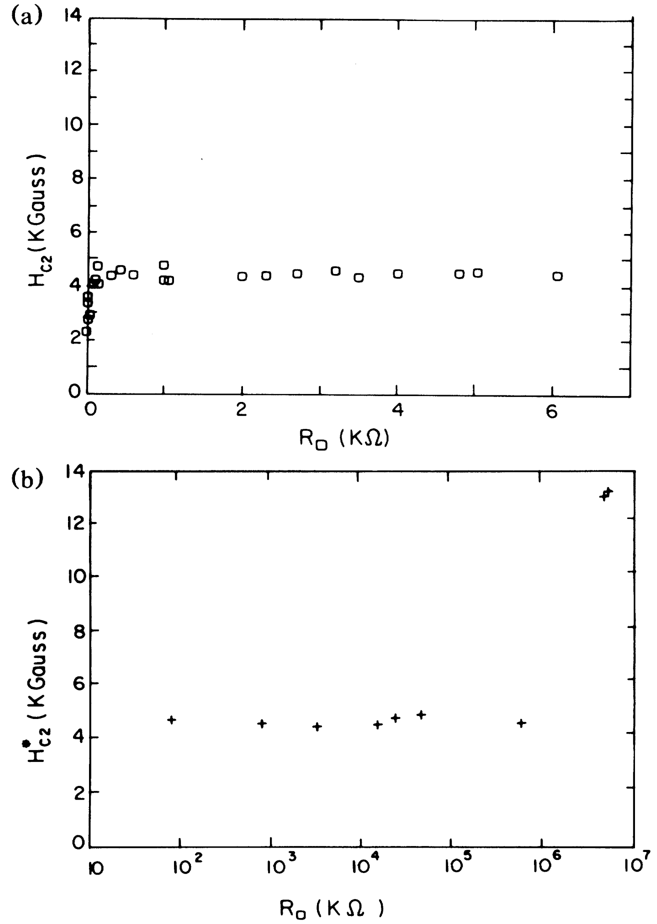

FIG. 3. (a) Upper critical field H_{c2} of lead samples above the percolation threshold as a function of normal resistivity per square at 4.2 K. (b) Upper critical field H_{c2}^* of lead samples below the percolation threshold as a function of normal resistivity per square at 4.2 K. Resistivity scale is logarithmic.

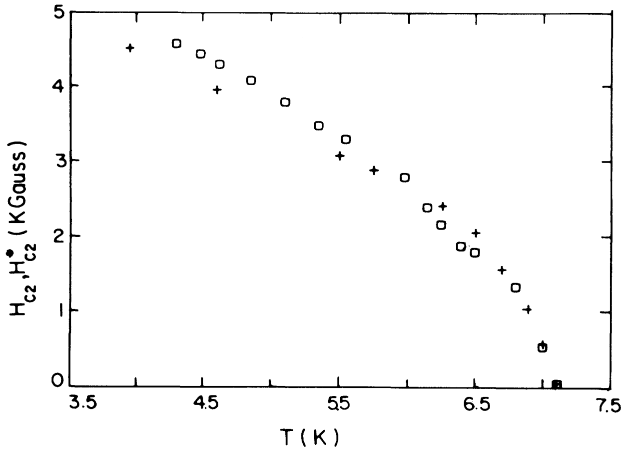

FIG. 4. Upper critical field H_{c2} of the sample closely above the percolation threshold [$R_\square = 2.1$ kΩ/□ (squares)], and H^*_{c2} of the sample closely below the threshold [$R_\square = 800$ kΩ/□ (crosses)], as a function of temperature.

normal-state value. We estimate that it can be determined to better than ±5%. The striking feature of Fig. 2 is that H^*_{c2} falls within the width of the resistive transition of a sample slightly above the percolation threshold.

Values of the perpendicular critical field as a function of normal-state resistivity are shown in Figs. 3(a) and 3(b). The behavior of H_{c2} and H^*_{c2} is in accordance with theoretical predictions concerning the critical field of percolating superconductors.[2,3] For $R \leq 100$ Ω/□, $\bar{\xi}_s > \xi_p$ and the films behave as homogeneous superconductors, H_{c2} being a linear function of R. For 1 kΩ/□ $< R_\square <$ 6 kΩ/□, $\bar{\xi}_s < \xi_p$ and H_{c2} remains constant. Below threshold, and for $R_\square < 1 \times 10^6$ kΩ/□, H^*_{c2} is also constant and equal to the value of H_{c2} above threshold. We interpret this behavior as characteristic of large clusters of size $\xi_p > \bar{\xi}_s$. We argue that tunneling between large clusters dominates the conductivity of the sample, because of their small electrostatic charging energy compared to that of the small clusters, and the smaller number of junctions that must be tunneled through across the sample. Hence, it is the critical field of the largest clusters, of size ξ_p, that is measured in these experiments. As we move towards higher $R_\square$, away from the threshold, ξ_p comes down and eventually the condition $\xi_p > \bar{\xi}_s$ is not met. The critical field then goes up as the cluster's size goes down ($R_\square > 1 \times 10^6$ kΩ/□).

The temperature dependence of the critical field of samples close to (above and below) the percolation threshold is shown in Fig. 4. It is clearly nonlinear near T_c, and if fitted by a power law $H_{c2}, H^*_{c2} \propto (T_c - T)^u$, the data provide a value of $u = 0.66 \pm 0.06$. The temperature dependence of the upper critical field, in the percolation-dominated regime $\xi_p > \bar{\xi}_s$, is predicted to be

$$H_{c2}(T) \propto \bar{\xi}_s^{-2} \propto (\Delta T)^{1/(1+\theta/2)},$$

where $\theta = (\mu - \beta)/\nu \simeq 0.9$ (2D). Our results are in excellent agreement with this theoretically predicted value, $u = 0.69$.

It is important to emphasize that the superconducting transitions reported here are those of large finite fractals and not of small clusters or single grains from which the clusters are built. The value of the single-grain critical field can be estimated to be at least that of the samples far below the percolation threshold, which is of the order of 12 kG at 4.2 K. Also, the anisotropy ratio, i.e., the ratio of parallel to perpendicular critical field of the films, is above 2.5 and not of the order of 1 as expected for a single grain.

The results presented here complete the earlier reported data for superconductors above the percolation threshold.[4] They give the full picture both above and below the percolation threshold. In the inhomogeneous limit the upper critical field remains constant across the threshold. Its temperature dependence near T_c is described by a power law with $u = 0.66 \pm 0.06$. These results fully confirm theoretical predictions for the upper critical field of large fractals.

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