Nonuniversal critical exponents for the superconducting critical current in lead thin films

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We present experimental results on electrical measurements of evaporated thin lead films (inverse swiss cheese model) and ion-milled thin films (swiss cheese model) at liquid-helium temperatures near the superconductor-insulator percolation threshold. We provide evidence for the nonuniversality of the critical exponents for the critical current in two dimensions for the two systems studied, in good agreement with theoretical results.

Since superconductivity is a long-range order phenomenon, the inhomogeneous scales and disorder present in a sample, will directly determine its superconducting properties.[1] Depending on the relative size of the length scales of disorder and superconducting correlations, different phenomena such as percolation, charging effects, and localization will be of importance. The study of two-dimensional superconducting systems near the superconductor-insulator threshold in amorphous and granular materials is of considerable interest because of the richness of phenomena that can be observed.[2,3] The threshold of superconductivity in granular films (with a mesoscale disorder) can be understood on the basis of phase fluctuation effects and some aspects of the insulator-superconductor transition can be explained using percolation models.[2] Furthermore, the percolation critical exponents for the critical current in a superconducting-insulating composite have been predicted to be nonuniversal depending strongly on their microstructure.

In this work, we present experimental results for the critical exponents of the superconducting critical current in lead thin films. The films are evaporated as well as ion milled at 4 K near the insulator-superconductor percolation threshold. In this regime, the lead thin films form a two-dimensional granular system and we can make use of percolation theory. We find, in both systems (evaporated and ion-milled films), critical exponents which are larger than the exponent expected for a discrete two-dimensional lattice. These results are in agreement with the theoretical values and simulations made for the continuous percolation models of inverse swiss cheese and swiss cheese, given the microstructure and coherence length of our thin films. To our knowledge, this is the first experimental report of the universality of these exponents.

Figure 1 shows the experimental setup used in our experiments. Samples were grown at 4 K on sapphire substrates attached to the bottom of a cylindrical dewar filled with liquid helium, shielded in its lower part by a copper shield. The sample mount is surrounded and thermally shielded by another cylinder filled with liquid nitrogen and also protected by the shutter from the surroundings. The films were formed, within a high-vacuum chamber, by thermal evaporation of lead through a copper mask to form a pattern of 1×7 mm. For the evaporated films, the nominal thickness (between 20 and 100 Å) was measured by a calibrated quartz oscillator and controlled by means of a mechanical shutter. Temperature measurements were made by using a calibrated germanium resistor. An ion gun at the bottom of the chamber allowed us to ion mill the films (roughly 500 Å thick), previously deposited at liquid-nitrogen tempera-

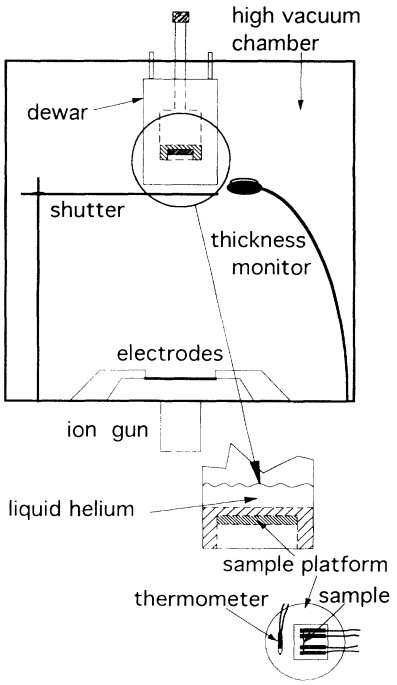

FIG. 1. Experimental setup.

ture to avoid problems of granularity. After this step, we remove the copper mask (to avoid redeposition during ion milling) and cool the film again at 4 K to erode it with an argon ion gun. In this case, if the erosion is constant, the thickness of sample may be assumed to be related to the ion-milling time as

$$d = d_0 - \alpha t \ , \tag{1}$$

where d_0 is the initial thickness, α is a positive constant and t is the ion-milling time.

The electrical characterization of the films was made by taking their voltage-current curves, using the four-contact technique, at various thicknesses and at constant temperature of about 4 K. From the current-voltage characteristic we can determine the critical current (defined experimentally as the current at which a voltage of 5 μV appears) and then, study how the critical current scales with thickness.

The voltage-current curves during thermal evaporation, were taken just when the sample started to conduct ($R \leq 20$ MΩ). During the deposition (when the shutter was opened), the temperature of the sample increased to a maximum value of 5 K. When the sample began to conduct (at a nominal thickness between 20 and 60 Å), I-V curves were taken at intervals of thickness between 3 and 5 Å and at 4 K. At the beginning of the evaporation, the voltage-current characteristic is strongly nonlinear and there is a threshold voltage at which conduction begins. In Fig. 2(a) we can see typical I-V characteristics at different thicknesses of the sample; we observe that the nonlinearity and threshold voltage decrease with thickness. This regime may be explained by tunneling of quasiparticles. Transmission electron microscopy analysis of evaporated films in this regime show (taking the most favorable case of small islands and separation) that there exist isolated superconducting islands of lead with an diameter of 200 Å separated 330 Å from center to center. In this case localization effects are negligible (the electrostatic energy between islands is lower than $k_B T$) and we can suppose that conduction is due to electron tunneling between isolated superconducting islands. We have that no current flows until the applied voltage reaches a value of nE_g/e, where n is the number of superconducting junctions assuming that they form a linear chain.[5] The energy associated with superconducting coupling between isolated grains of lead is $E_g = 2\Delta(T)$, which at 4 K is $E_g(4\text{ K}) = 1.97 \times 10^{-3}$ eV. From Fig. 2(a) we have a maximum threshold voltage of about 1 V (at 4 K), which implies a maximum value for $n = 508$ for a linear arrangement of superconducting junctions. This small number of uncoupled junctions can only be explained as arising from thermal fluctuations at 4 K ($k_B T = 3.62 \times 10^{-4}$ eV), which are comparable to the coupling energy of the rest of the junctions and which tend to reduce coupling.

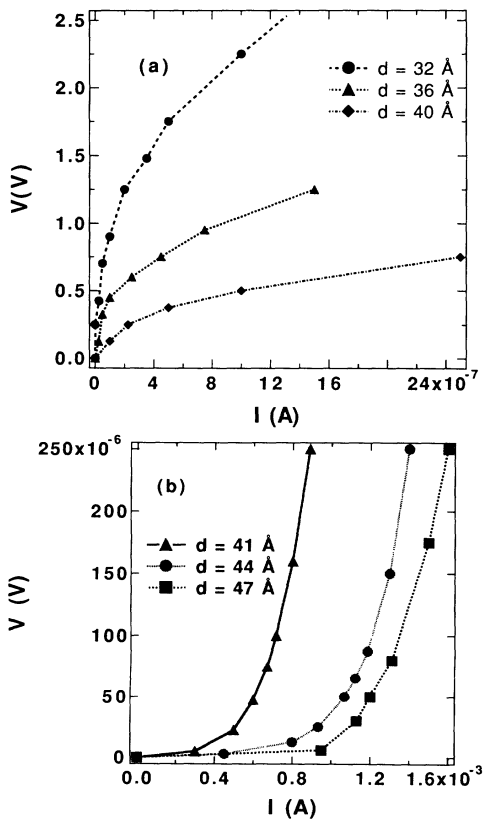

FIG. 2. I-V characteristics for evaporated films at 4 K: (a) for zero critical current, (b) for a measurable critical current.

During lead deposition, the curvature of the I-V curve changes from concave downward to concave upward (dV/dI increases with current) and a small critical current begins to appear in the I-V characteristics, which increases rapidly as more material is deposited. In Fig. 2(b) we present typical I-V characteristics in this regime. During the erosion process, the rate of erosion is kept at approximately 0.13 Å/sec. We let the ion beam erode the sample until the critical current decreases to less than 15 μA, and then we take I-V curves at different erosion times and at a constant temperature of 4 K. We determine the thickness from (1), assuming a constant erosion rate of the film. During ion milling, the sample temperature rises to 6 K. The I-V curves for different ion-milling times (thickness) have the same shape as that in Fig. 2(b) [for ion-milled films we were not able to reproduce the regime shown in Fig. 2(a)].

The critical currents measured from the I-V characteristic plotted as a function of thickness for a typical evaporated film are shown in Fig.3. We observe the same shape when we plot the critical current as a function of ion-milling time. At low thickness, the critical current varies strongly with thickness. In this regime we can assume that percolation theory is valid and that

$$I_c = (d - d_c)^{\gamma} \tag{2}$$

is applicable (I_c is the critical current, d is the film thickness, and d_c is the critical thickness at which a critical current first appears). From this, we can try to determine experimentally the exponent γ for both cases, if a value of d_c can be determined for evaporated and ion-milled thin films, and then attempt to understand the origin of the particular values of γ.

It is known that in continuous percolative systems the critical exponents can be nonuniversal and their values

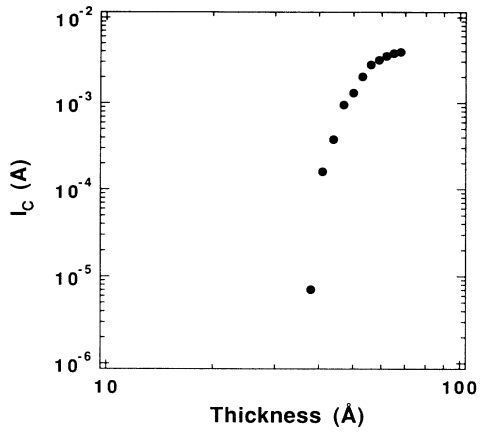

FIG. 3. Critical current at different thicknesses of the sample for an evaporated film [for the same sample as in 2(b)].

depend both on the dimension of the system and on its microstructure.[5] The transport properties in continuous disordered systems near the percolation threshold, cannot be treated, in general, with classic discrete percolative models, in which links randomly occupy a lattice with the same weight. Instead, one needs to use continuous percolative models in which there is a continuous distribution of weights for the occupied links.[4] In the generalization of these models to superconductor percolative systems we must consider two scales: the disorder scale of the system and the superconducting coherence length,[6] since the microstructure determines J_c as shown below.

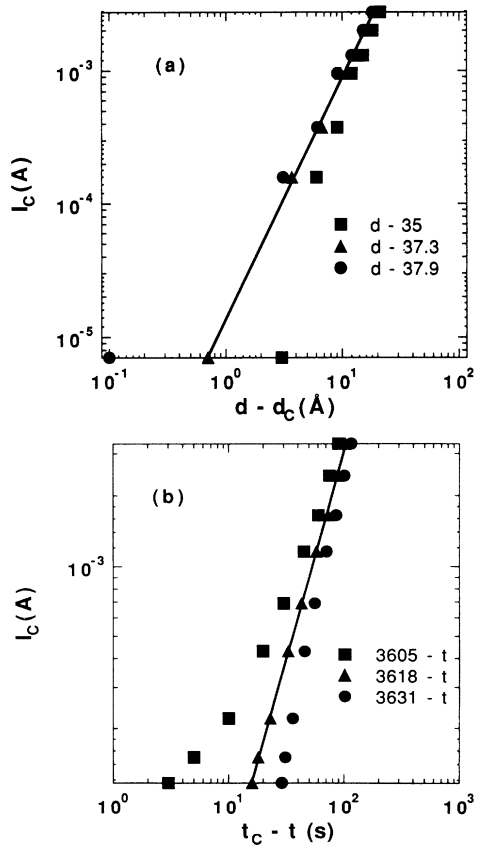

FIG. 4. (a) Critical current vs $(d-d_c)$ for evaporated films at 4 K; (b) critical current vs (t_c-t) for ion-milled films at 4 K.

TABLE I. Experimental results for the critical exponents.

	Evaporated films	Ion-milled films
v	1.88±0.09	1.84±0.05

The critical current density in a d-dimensional percolative superconductor system, has the form:[7]

$$J_c = I_c \xi_p^{1-d} , \tag{3}$$

where ξ_p is the percolation correlation length, which near the percolative threshold, varies like $(p-p_c)^{-\nu}$ (p is the fraction of superconductor present and p_c is the critical fraction). Using the nodes-links-blobs approximation, one can then assume near the percolation threshold, that the critical current of the sample I_c only depends on the critical current of the weakest bond $I_{\min}$, which scales like $(p-p_c)$, and $I_c \propto I_{\min}^m \propto (p-p_c)^m$, where m is an exponent that depends on the microstructure and characteristic scales of the system.[7]

The critical current density in a percolative two-dimensional ($d=2$) superconductor system is then

$$J_c \propto (p-p_c)^v , \quad \text{where } v = m+\nu . \tag{4}$$

From theory and in agreement with simulations[5] made assuming a distribution of critical currents, it is known that the values of v are those shown in Table II. The column corresponding to the Josephson case refers to the case in which the weakest bond behaves like a Josephson junction and the column named depairing corresponds to the case in which the weakest bond behaves like a microbridge with a critical current proportional to the width of the bond.

If we assume[8-10] that the superconducting fraction p is proportional to the nominal thickness d measured with the quartz crystal, then the dependence of the critical current with thickness in the percolation transition will be $I_c \propto (d-d_c)^v$ (for evaporated films), or $I_c \propto (t_c-t)^v$ for ion-milled films [in accordance with Eq. (1)]. Our data is then fitted to these forms adjusting d_c and t_c to obtain the best fit to a power law. In Fig. 4 we show two graphs: (a) the critical current I_c vs $(d-d_c)$ for evaporated films and (b) critical current I_c vs (t_c-t) for ion-milled films. The exponent v is determined experimentally with an average over three samples (for each case) and the slope is obtained from the two best fits varying d_c in steps of 0.1 Å (t_c in steps of 1 sec). The experimental value of v found in this measurements are shown in Table I. For evaporated films the exponent v determined is greater than both the exponent expected from a discrete two-

TABLE II. Simulated critical exponent for the discrete lattice, swiss cheese model, and inverse swiss cheese model.

Discrete lattice	Swiss cheese		Inverse swiss cheese	
	Josephson	Depairing	Josephson	Depairing
1.333	1.821	2.318	1.333	1.821

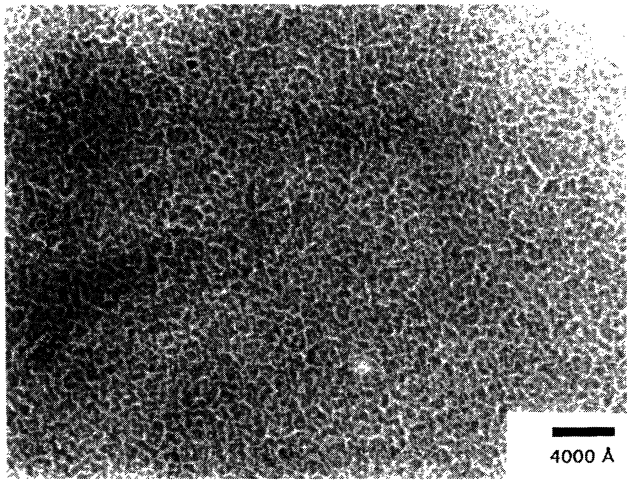

FIG. 5. Transmission electron micrography of an evaporated film, the dark zone corresponds to lead.

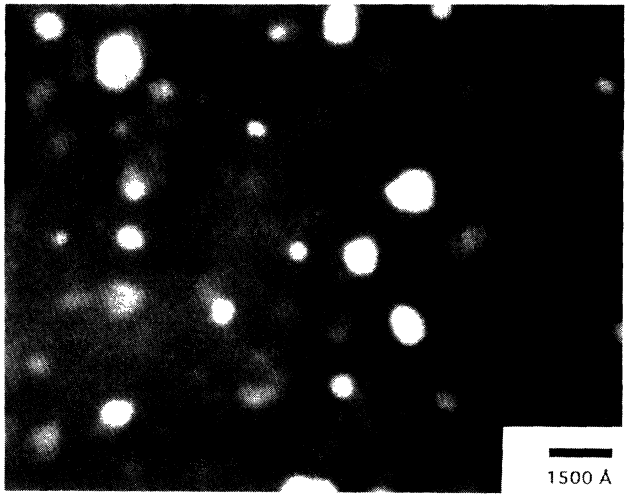

FIG. 6. Transmission electron micrography of an ion-milled film, the dark zone corresponds to lead.

dimensional lattice and the exponent measured in a similar system[3] (lead evaporated on a germanium substrate).

Comparing our results for evaporation with those expected from theory (see Table II), we see that our experiments will only be consistent with theory if the size of islands (using results of inverse swiss cheese model), is bigger than the coherence length of lead at 4 K. The temperature-dependent superconducting coherence length in the clean limit (from Gorkov theory) is $\xi(T)=0.74\xi_0(1-T/T_c)^{-1/2}$ (for lead $\xi_0=830$ Å), then we have that the coherence length of lead at 4 K is roughly 950 Å. From this, the exponent measured indicates that the typical size of the islands near the percolation threshold should be greater than 950 Å. This result has been verified by electron transmission microscopy of evaporated films at 4 K, near the superconductor-insulator percolation threshold; in Fig. 5 we show a TEM micrography of a 40-Å-thick evaporated lead film. Thus, the system is in the depairing regime where the smallest neck behaves like a one-dimensional wire.

The exponent obtained by ion milling suggests a size of holes (swiss cheese model) smaller than the coherence length, which is reasonable if we note that erosion in the film is due to collision of argon ions with the lead surface at low temperature. In Fig. 6 we show a TEM picture for one of these films (the thickness for this film is roughly 80 Å); a wide distribution of hole sites can be observed. While this is different from the evaporated films, we can note that the major part of holes is smaller than 950 Å, that is, smaller than the coherence length, and given that the smallest link would behave as a Josephson junction, the smallest hole should determine the critical current. Thus, both exponents measured are nonuniversal.

In conclusion, we present experimental results for the nonuniversality of the critical exponent for the critical current in a two-dimensional superconductor-insulator system for two cases: evaporated films (inverse swiss cheese model) and ion-milled films (swiss cheese model), and we find in both cases a critical exponent greater than the exponent for a discrete lattice. For evaporated films, the exponent found suggest an island size greater than the coherence length of lead at 4 K, in agreement with the image obtained by transmission electron microscopy. The critical exponent measured for ion-milled films corresponds to a typical size smaller than the superconducting length and the necks act like Josephson junctions. This is an experimental measurement of these critical current exponents for the two models commonly used in continuum percolation theory.

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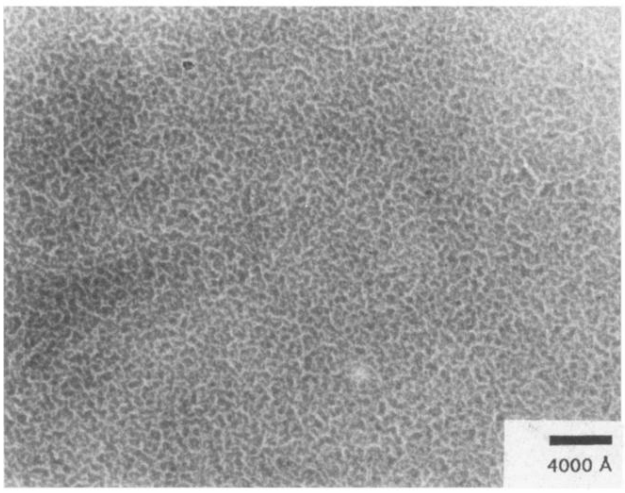

FIG. 5. Transmission electron micrography of an evaporated film, the dark zone corresponds to lead.

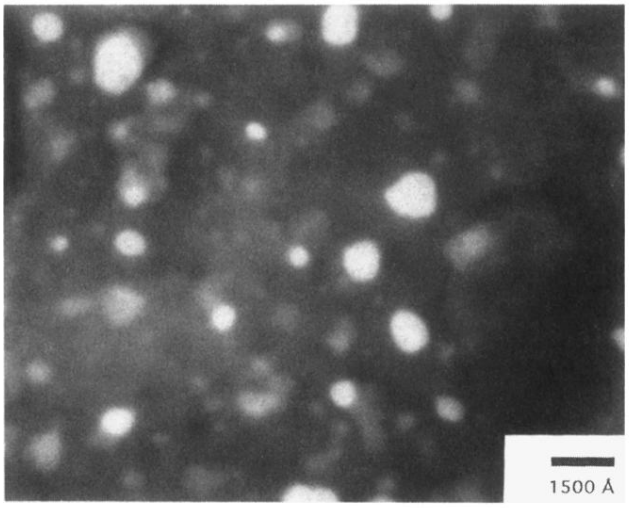

FIG. 6. Transmission electron micrography of an ion-milled film, the dark zone corresponds to lead.