Tunneling percolation model for granular metal films

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The evolution of conductivity in granular metal films, as a function of thickness and temperature, has been modeled by percolation within the framework of the theory of stochastic transport in disordered systems. The model was analytically worked out by means of the effective-medium approximation. The analytical expression obtained spans the entire range of resistance, from insulating to globally superconducting behavior, with increasing thickness. Quasireentrant or local superconductivity was found in our results as a consequence of tunneling junctions allowed in the model. Experimental data on tin films are considered. [S0163-1829(97)05905-5]

I. INTRODUCTION

The conditions for the onset of bulk solid behavior and the development of associated phenomena, such us superconductivity in ultrathin films, are important problems in condensed matter physics. Recently fine experiments[1] have been realized in ultrathin metal films that present granular structure. The general behavior of sheet resistance $R(T)$ with thickness resulting in these experiments was surprisingly reproducible for a variety of soft metals such as Sn,[2,3] Ga,[4,5] Pb,[5] Al, and In. The thinnest films exhibit strong insulating behavior and activated conductivity with no trace of superconductivity. Manifestations of local superconductivity first become noticeable for thicker films as a ''kink'' and before a local minimum in $R(T)$. The latter behavior has been called ''quasireentrant'' superconductivity because the resistance never actually drops to zero. Subsequent addition of material then leads to films that present global superconductivity. Quasireentrant superconductivity has also been observed in three-dimensional films of granular aluminum.[6] However, this particular behavior has not been found in homogeneous films. Granular films result when glass or ceramic substrates are used. Homogeneous films are formed on amorphous Ge substrates. The resistance of homogeneous films is always a monotonic function of the temperature T (see Ref. 7 for a review).

A remarkable feature in thin films is the fact that global superconductivity is found when the normal-state sheet resistance falls below a value close to $h/4e^2$. This is a universal observation uncorrelated with either structural or material parameters such as thickness or transition temperature.[1] Very much attention has been addressed to the role of Josephson junctions in the interpretation of the experimental data of thin granular films. A random array of Josephson-coupled grains was considered in order to explain the origin of the resistance minimum. This was theoretically treated by a bond percolation model[8] and also experimentally studied.[9] The resistive transition was also treated by renormalization-group analysis of the effects of zero-point fluctuations on the vortex-unbinding scenario.[10]

Our aim in this paper is somewhat different. We will present a tunneling percolation model for describing the film's electrical behavior from quasireentrant (local) to global superconductivity in the context of the stochastic transport theory.[11] The stochastic transport theory maps the conduction process in disordered media on a random walk on lattices with random transition rates. The randomness in our model is a crucial ingredient that mixes two different phenomena: the insulator-conductor transition at the metallic grain concentration $\overline{p}$ and the metallic connectedness percolation one at the concentration p_c. Analytical results are done within the framework of the effective-medium approximation (EMA). The EMA is a powerful approach to study analytically diffusion problems and stochastic transport processes in disordered media. The EMA was developed independently by several authors.[12,13] Whether the approximation gives exact results was analyzed recently.[14]

The random walk approach has been used to study the random two-component mixture of good and poor conductors. The limit case in which the poor-conductor species has infinite resistance is known as a random resistor network (pure ''ant'' limit), and the limit case in which the good-conductor has zero resistance is known as a random superconducting network (pure ''termite'' limit). The ''termite'' behaves like an ''ant'' on the normal conductor, where it performs a random walk in steps of one lattice spacing. However, the clock is held still while it walks on the superconductor. The scaling properties of random resistor and superconducting networks have been extensively studied by Hong *et al.*[15] who developed a scaling theory for the general two-component alloy.

The EMA has also been extended to study the first-passage time problem in random media.[16] The first-exit time analysis is a useful tool for describing the problem of ''termites'' diffusion.[17] Recently, the role of percolation in diffusion on square and simple cubic lattices with a random distribution of energy barriers has been investigated by

means of Monte Carlo simulation and the EMA.[18] The analytical predictions of the EMA in this model are in good agreement with numerical results both in the asymptotic long-time limit and in the whole transient regime. Thus, the reliability of the EMA has been well established in the framework of percolation processes. However, the universal behavior at the percolation threshold cannot be described correctly by an approach like the EMA.[19]

The starting point for the stochastic transport theory, within the linear response approximation, is the generalized Einstein equation for the conductivity:[11]

$$\sigma = \frac{nq^2}{k_B T} D, \tag{1.1}$$

where k_B is Boltzmann's constant, n is the number density of *effective carriers*, q is the charge of the carriers, and D denotes the frequency-dependent diffusion constant of the disordered medium. To evaluate D in a disordered medium we must know all the transition rates for the carrier between any pair of lattice sites. In the EMA, the problem of a particle's diffusion in a disordered lattice is replaced by a diffusion problem on an ordered lattice with an effective transition rate ω_{eff} that is frequency dependent:

$$D = \frac{1}{2d} Z a^2 \omega_{\text{eff}}, \tag{1.2}$$

where d is the dimensionality of the system, Z is the coordination number of the lattice, and a is the lattice constant. dc hopping conductivity is calculated in the zero-frequency limit and corresponds to the long-time limit of the diffusion process. For the resistance in a disordered media we have then

$$R = C \frac{T}{\omega_{\text{eff}}}, \tag{1.3}$$

where C is a constant that depends on the carrier properties n and q, on the lattice geometrical constants Z and a, and on the geometrical form factors of the sample. For growing films the geometrical form factors are given by the effective conducting cluster, and thus depend on the growth parameter (thickness). Moreover, on large length scales, only the carriers on the effective conducting cluster contribute to the conductivity. Their density n is therefore also proportional to the thickness.

This paper presents a tunneling percolation model for metallic granular superconductors. In Sec. II the transport process of the model is mapped on a random walk in a lattice and the basic quantities are constructed. The EMA is performed in Sec. III and the percolation thresholds $\overline{p}$ and p_c are calculated. Section IV provides the temperature behavior of hopping rates and the analytical expressions for the film resistance dependence in temperature and concentration of metallic grains are given. Section V contains the fits to the experimental data of tin films reported in Ref. 2. The fits with our approach suggest the relevance of the third dimension in the conduction process of the film. Section VI provides a final discussion and some concluding remarks. The results endorse an explication of quasireentrant superconductivity based on the geometrical interplay between tunneling and metallic junctions in the film.

II. TUNNELING PERCOLATION MODEL FOR GRANULAR SUPERCONDUCTORS

The present model is based on the nature of the considered films.[1,2] The samples were grown by precisely controlled exposures of a cold substrate to the metal vapor flux. Depositions of increments of average film thickness of less than 0.02 Å were reported as possible, and typically increments between 0.05 and 0.2 Å were used in the evaporations. The vapor flux density was estimated to be uniform to better than 1 part in 10^4 over the area of the sample. All films were grown on silica-glazed alumina substrates held between 15 and 18 K during metal deposition. The sample temperature remained below the highest temperature during deposition and annealing effects were avoided. The glaze provided a very smooth surface on atomic length scales with variations occurring only at long wavelengths. In consequence, it is reasonable to assume *equivalent uncorrelated metallic grains* on the film, with a narrow grain-size distribution across the sample in the successive depositions of metal.

We simplify the growth process by considering the insulating substrate as a hypercubic regular lattice in two (square) or three (simple cubic) dimensions. Each site of the lattice can be occupied by a metallic grain with probability p or can be unoccupied (vacancy) with probability $1-p$. We are not considering correlations between any pairs of sites. We assume the probability p to be proportional to the nominal thickness. For *true* two-dimensional films, p is the areal coverage and can be measured from digitized micrographs.[20] The lattice constant a is selected equal to the mean diameter of grains. In this manner, two nearest-neighbor grains are forming the smallest metallic cluster. This random manner of filling the space induces topological disorder on the site scale, but also represents a situation of structural disorder on the cluster scale.

We assume that the electronic states are *localized in the metallic sites* and that the effective electronic transport process takes place by hopping among neighbor grains. The transport process is mapped in the conventional way on a random walk in the lattice,[11,13] and all the randomness of the system is contained in the spatial distribution of hopping rates between sites. We assume hopping bonds between nearest-neighbor sites only.

There are three types of bonds between two nearest neighbor sites in the lattice. The bonds between two vacancies are insulators (with hopping rates $\omega_0 = 0$), the bonds between two grains are metallic junctions (with hopping rates ω_s), and the bonds between a grain and a vacancy are regarded tunneling junctions (with hopping rates ω_t). For the two-dimensional case, a given realization of the lattice is depicted in Fig. 1. Two metallic clusters will be joined into only one cluster if they have at least a pair of nearest neighbor grains, and will be connected by at least one tunneling path if each cluster has a grain nearest neighbor to a same vacancy in the lattice. There are several possible configurations of parallel tunneling paths between two grains according to the environment of them, which increase with the dimensionality of the lattice. Thus, the model takes into ac-

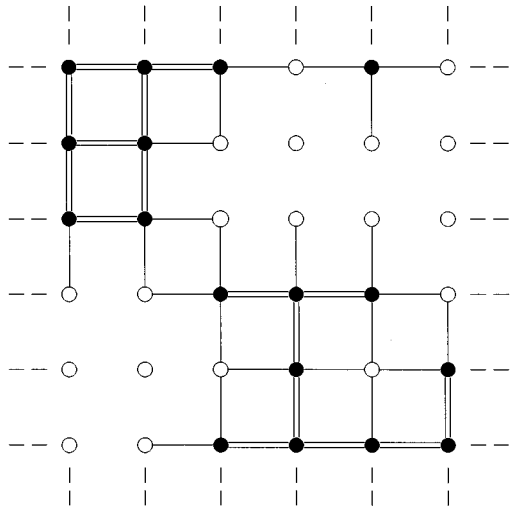

FIG. 1. A realization of the system into the square lattice. The sites are occupied by metallic grains with probability p or are vacant with probability $1-p$. References: (●) metallic grain, (○) vacancy, (=) metallic bond, and (−) tunneling bond.

count in an elementary way the variations in the bond strength due to randomness in the tunneling distance between metallic grains, which is present in the experimental samples.

The fractions P_0, P_s, and P_t of insulating, metallic conducting, and tunneling bonds depend on the concentration p in the following way:

$$P_0(p)=(1-p)^2,$$

$$P_s(p)=p^2, \tag{2.1}$$

$$P_t(p)=2p(1-p).$$

$P_0(p)+P_s(p)+P_t(p)=1$. These relations are completely general and hold in two and three dimensions, included for nonhypercubic lattices. Therefore, we are dealing with a three-component system where the lattice bonds have hopping rates ω randomly distributed with the probability density[21]

$$\Pi(\omega)=P_0(p)\,\delta(\omega)+P_s(p)\,\delta(\omega-\omega_s)+P_t(p)\,\delta(\omega-\omega_t). \tag{2.2}$$

From the different types of bonds we will obtain in Sec. III two critical concentrations $\overline{p}$ and p_c. $\overline{p}$ will be related to the conducting (tunneling and metallic) bonds and defines the percolation threshold where a insulator-conductor transition occurs. p_c will be related to the metallic bonds only and defines the lowest concentration where an infinite cluster of metallic bonds develops (connectedness percolation). For $p>p_c$ the model will show metallic conductor-superconductor behavior.

In the past another related model for tunneling percolation was proposed by Gefen *et al.*[22] and recently reanalyzed in Ref. 23. That model was found to be compatible with the experiments on a discontinuous thin gold film near the percolation metal-insulator transition. Recently, tunneling across dangling bonds or between metal grains in random two-components mixtures has been considered in the dc and ac electrical properties of insulator-conductor composites near the percolation threshold. Evidence for a new scaling law has been reported.[24]

A three-component model was developed for the conductivity of dispersed ionic conductors,[25] but that case corresponds to interfacial conductivity instead of conduction between domains as in our case. This and our model have similar expressions for the bond fractions in the two-dimensional case [Eq. (2.1)], but they are intrinsically different in the three-dimensional case.

III. EFFECTIVE-MEDIUM APPROXIMATION

The EMA self-consistent equation for the coherent transition rate $\omega_{\rm eff}$ is[13]

$$\left\langle \frac{\omega_{\rm eff}-\omega}{1+2\,\Theta(\omega_{\rm eff}-\omega)} \right\rangle_{\Pi(\omega)}=0, \tag{3.1}$$

where the angular brackets indicate the average over the distribution $\Pi(\omega)$ given in Eq. (2.2). The EMA for stochastic transport is outlined in full detail in Ref. 13. The reader is referred to this excellent reference for further details. In the zero-frequency limit and for hypercubic lattices we get $\Theta^{-1}=-2d\omega_{\rm eff}$. In the long-time or zero-frequency limit the hopping problem becomes equivalent to the random-resistor system and the effective-medium theory for that model was already given by Kirkpatrick[26] in classical bond and site percolation problems.

The self-consistent condition, Eq. (3.1), reduces to a quadratic equation for $\omega_{\rm eff}$:

$$[(d-1)\omega_{\rm eff}]^2+B(d-1)\omega_{\rm eff}-C=0, \tag{3.2}$$

where

$$B\equiv\omega_t\{[1-dP_t(p)]+[1-dP_s(p)]\tau\}, \tag{3.3a}$$

$$C\equiv\omega_t^2[d-1-dP_0(p)]\tau, \tag{3.3b}$$

and $\tau\equiv\omega_s/\omega_t$. The relevant root of Eq. (3.2),

$$\omega_{\rm eff}=\frac{1}{2\,(d-1)}(-B+\sqrt{B^2+4\,C}), \tag{3.4}$$

is positive for $C>0$. Thus, the threshold $\overline{p}$, i.e., the lowest concentration of metallic grain where conduction becomes possible, is a solution of $d-1-dP_0(\overline{p})=0$ ($B>0$ for p in a neighborhood of $\overline{p}$). This situation corresponds to percolation of a conducting path that is made of metallic and tunneling bonds in series. The strong insulating behavior of the film for $p\gtrsim\overline{p}$ will be given by the thermally activated conductivity, characteristic of the tunneling junctions. We obtain $\overline{p}=1-\sqrt{1-1/d}$, and thus $\overline{p}\approx0.293$ for $d=2$ and $\overline{p}\approx0.184$ for $d=3$.

For $C\gtrsim0$ we get $\omega_{\rm eff}\approx(d-1)^{-1}C/B$, and then if the

percolation threshold is approached from above ($p \gtrsim \overline{p}$), ω_{eff} approaches zero with a critical exponent μ,

$$\omega_{\text{eff}} \propto (p-\overline{p})^{\mu}. \tag{3.5}$$

In our model, we find $\mu=1$ independently of d.[27] The EMA values for the critical exponents in the scaling laws are only rough estimations.[19]

In the limit $\tau \gg 1$ (highly conducting metallic bonds), $B \approx \omega_t[1-dP_s(p)]\tau$ and

$$\omega_{\text{eff}} \approx \frac{B}{2(d-1)}\left[-1+\text{sgn}(B)\left(1+\frac{2C}{B^2}\right)\right] \tag{3.6}$$

if $B \neq 0$, but $\omega_{\text{eff}} \approx \sqrt{C}/(d-1)$ if $B=0$. Therefore, the condition $B=0$ separates two different conducting regimes: $\omega_{\text{eff}} \propto \omega_t$ for $B>0$ and $\omega_{\text{eff}} \propto \omega_s$ for $B<0$. Hence, the threshold p_c, i.e., the lowest concentration p where an infinite metallic cluster develops, is a solution of $1-dP_s(p)=0$. The superconducting behavior of the film for $p>p_c$ results from a divergence in the hopping transition ω_s. We obtain $p_c=\sqrt{1/d}$, and thus $p_c \approx 0.707$ for $d=2$ and $p_c \approx 0.577$ for $d=3$. The difference between both percolation thresholds is almost independent of d: $p_c-\overline{p} \approx 0.4$. In our model, the metallic fraction of bonds resulting for $p=p_c$ is given by $P_s(p_c)=1/d$.[19] This value can be compared with the critical fractions for uncorrelated bond percolation:[28] $P=1/2$ in $d=2$ (exact result) and $P=0.2488$ in $d=3$ (series expansion result). Thus, in the limit $\tau \gg 1$ we can write

$$\omega_{\text{eff}} \approx \begin{cases} \dfrac{d-1-dP_0(p)}{(d-1)[1-dP_s(p)]}\,\omega_t & (\overline{p} \leqslant p < p_c), \\ \dfrac{\sqrt{d-1-dP_0(p)}}{(d-1)}\sqrt{\omega_t \omega_s} & (p=p_c), \\ \dfrac{|1-dP_s(p)|}{(d-1)}\,\omega_s & (p_c < p \leqslant 1). \end{cases} \tag{3.7}$$

This expression will allow us obtain the general behavior of $R(T)$ with thickness. The particular behavior of ω_{eff} at p_c is due to the fact that the transport process is in the tenuous percolating backbone, while for $p \neq p_c$ transport is in a homogeneous disordered metallic support.

IV. TEMPERATURE-DEPENDENT RESISTANCE

To compare our tunneling percolation model with experimental results of granular superconductors we have to introduce a temperature dependence in the hopping rates ω_t and ω_s. We assume the simplest hypothesis, taking

$$\omega_t(T) = K_t \exp\left(-\frac{U}{T}\right), \tag{4.1a}$$

$$\omega_s(T) = \begin{cases} \infty & (T<T_c), \\ K_s\{\tanh[\alpha(T-T_c)]\}^{-1} & (T>T_c). \end{cases} \tag{4.1b}$$

The dimensional constants K_t and K_s give the strength of the bonds. In this manner, the tunneling junctions are thermally activated with activation temperature U (see Ref. 29) and the metallic bonds become superconductors for $T \leqslant T_c$. The temperature T_c is the critical threshold of the conductor-superconductor transition and will be the local transition temperature for p below the percolation threshold p_c. This simple model enables us to consider only granular films with constant local transition temperature, like Sn films.[1] The parameter α controls the temperature saturation range. Within this range the bulk metallic conduction reaches the high-temperature behavior. From Eqs. (3.7) and (4.1), the function $\omega_{\text{eff}}(T)$ results in being continuous in T_c for $p<p_c$ and diverges for $T \leqslant T_c$ if $p \geqslant p_c$.

Equations (1.3), (3.3), (3.4), and (4.1) give the analytical expression for the temperature dependence of the film resistance $R(T)$. In Eq. (3.4) the hopping rate ω_t can be factored out; therefore, ω_{eff} turns out to be a function of T through τ. Introducing the notation $\mathbf{K}=K_s/K_t$ and $R_0=C/K_t$, we obtain from Eqs. (1.3) and (3.7) the following behavior for $T<T_c$,

$$\frac{R(T)}{R_0} = \begin{cases} \dfrac{(d-1)[1-dP_s(p)]}{d-1-dP_0(p)}\,T\exp\left(\dfrac{U}{T}\right) & (p<p_c), \\ 0 & (p \geqslant p_c), \end{cases} \tag{4.2}$$

and for $T \gtrsim T_c$ (i.e., in the limit $\tau \gg 1$),

$$\frac{R(T)}{R_0} \approx \begin{cases} \dfrac{(d-1)[1-dP_s(p)]}{d-1-dP_0(p)}\,T\exp\left(\dfrac{U}{T}\right) & (p<p_c), \\ \dfrac{d-1}{\sqrt{d-1-dP_0(p)}}\,T\left[\exp\left(\dfrac{U}{T}\right)\mathbf{K}^{-1}\tanh[\alpha(T-T_c)]\right]^{1/2} & (p=p_c), \\ \dfrac{d-1}{|1-dP_s(p)|}\,T\,\mathbf{K}^{-1}\tanh[\alpha(T-T_c)] & (p>p_c). \end{cases} \tag{4.3}$$

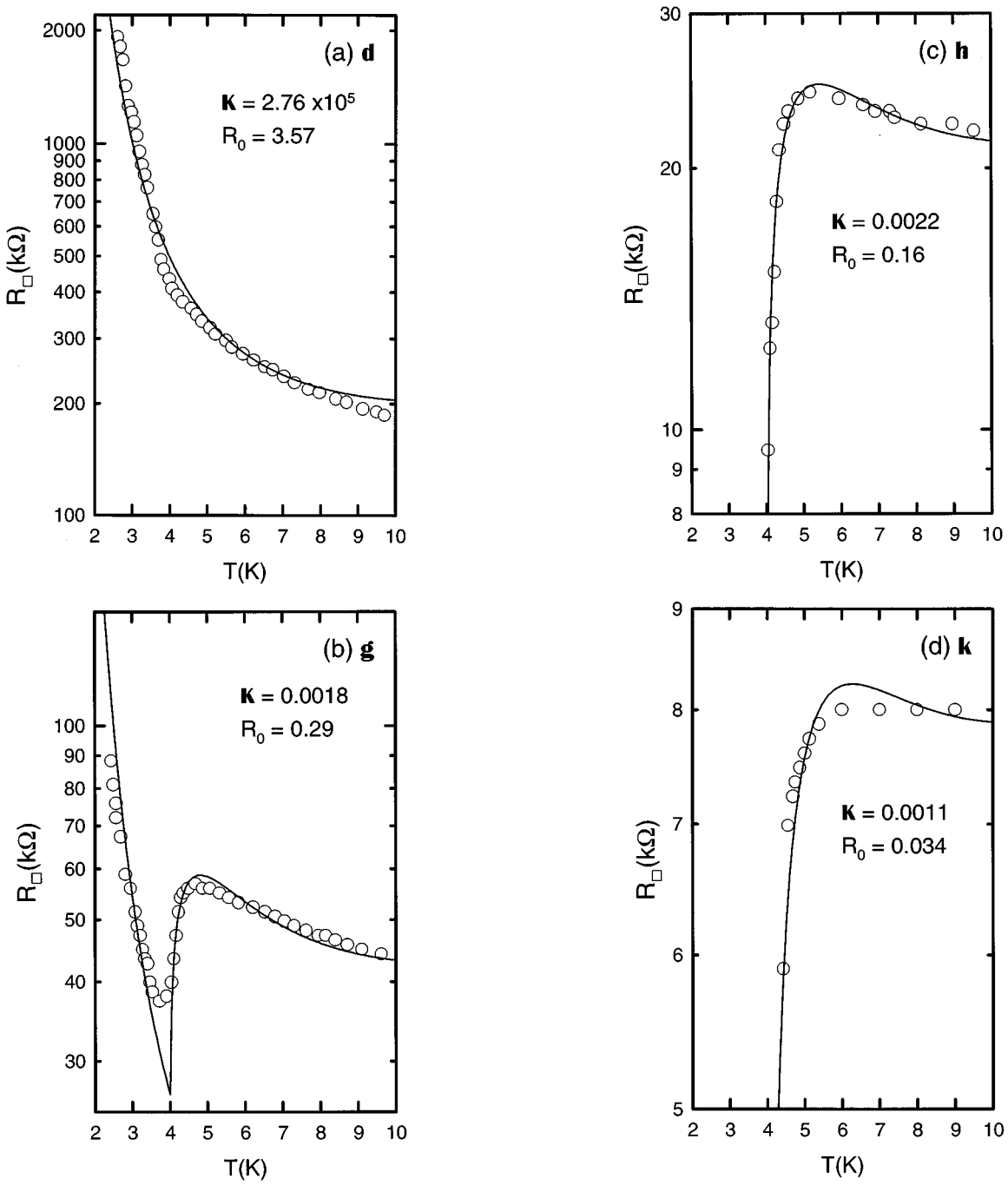

FIG. 2. The evolution of $R(T)$ curves for a tin film. Solid lines are fits of Eqs. (1.3), (3.4), (4.1), and (4.2) to the experimental data (open circles) with common fitting parameters $d=3$, $T_c=4$ K, $U=12$ K, and $\alpha=0.2$ K^{-1}. The small bold letters correspond to set B of Fig. 1 in Ref. 2. **K** has no dimensions and the units of R_0 are $k\Omega/\square$K. The values of p used are (a) 0.35, (b) 0.4, (c) p_c, and (d) 0.7.

For $p \lesssim p_c$, under the condition $B^2 \approx 4C$ we will find a ''quasireentrant'' superconductivity behavior in $R(T)$, where the resistant shows a nonzero minimum at T_c.

For $p \gtrsim p_c$, as the critical temperature is approached from above ($T \gtrsim T_c$), the resistance approaches zero with a critical exponent s,

$$R(T) \propto (T - T_c)^s. \tag{4.4}$$

We find $s=1/2$ if $p=p_c$ and $s=1$ if $p>p_c$, both independently of d. Thus, we find a different scaling behavior in the conductor-superconductor transition at the critical concentration p_c than for $p>p_c$.

V. DISCUSSION OF EXPERIMENTAL RESULTS

In this section we accomplish the fit to experimental data of resistance versus temperature corresponding to a Sn film set reported by Orr, Jaeger, and Goldman in part B of Fig. 1 in Ref. 2. The experimental data document the systematic evolution of 11 stages, from insulating to metallic behavior, as the sheet resistance of the film is decreased by increasing

its thickness from 19.4 Å to 21.2 Å. Typical thickness increments were 0.1 Å. Our choice of tin experimental data is based on the small dependence of the local transition temperature T_c as a function of the normal-state resistance $[R(14.0\,\mathrm{K})]$. However, some of the thicker Sn films also exhibit transition-temperature oscillations with thickness.[30] Moreover, on high-resistance Sn films, $R(T)$ increases sharply with decreasing temperature apparently without limit, whereas on Ga (Ref. 4) and Pb (Ref. 5) films $R(T)$ appears to flatten out at the lowest temperature even when there is a minimum. This fact suggests that the picture of tunneling junctions thermally activated must be modified in some manner for these films.

Four film growth stages are shown in Fig. 2. The small bold letters correspond to set B of Ref. 2. Digitalized experimental data are given as open circles and the solid lines represent fits to the experimental data using our approach from Eqs. (1.3), (3.4), (4.1), and (4.2). We have use the fixed values $T_c = 4$ K, $U = 12$ K, and $\alpha = 0.2\mathrm{K}^{-1}$. The critical temperature T_c is the average local transition temperature of the films. In our model we simplify the physical situation by considering the parameter U constant as a function of T. We evaluated U from the superconducting regime as $U \approx 2\Delta/k_B$, where Δ is the energy gap in the Sn superconductor. For the estimate α we have taken a saturation interval from T_c in the globally superconducting stages equal to 5 K. The parameters p, **K**, and R_0 are the remaining fitting constants. From our percolation model we associate the first experimental curve that presents globally superconductivity with a film at the critical threshold p_c [see Fig. 2(c)]. As one can see from Fig. 2, we find good agreement between experimental data and the calculated curves in all stages of film growth despite our oversimplified assumptions given in Eqs. (4.1). The agreement is excellent in the quasireentrant superconducting stage and near p_c. The fitted value of **K** in Fig. 2(a) is rather artificial because for $p \ll p_c$ the film resistance is driven by the tunneling junction and the only relevant strength constant is K_t, as we can see from Eqs. (4.2) and (4.3). From the simple assumptions given by Eqs. (4.1) **K** must be constant. We can assume the approximated value $\mathbf{K} = 0.002$.[31] The parameter R_0 is a function of p because of the thickness dependence of the geometrical form factors and the carrier density n.

The most striking feature of these fits to the experimental data is that all these were done with $d = 3$. For $d = 2$ our model looks like for the $d = 3$ case (qualitatively), but we can neither fit well the local minimum in quasireentrant superconductivity [see Fig. 2(b)] nor fit all the film set with the same parameters U and α. It is in apparent contradiction with the fact that the whole electrical evolution for all the films, experimentally studied, takes place over nominal thickness intervals corresponding *to less than one atomic layer*.[1] However, it is important to keep in mind that the total average nominal thickness at which this evolution takes place is about 20 Å or 30 Å, *near to four atomic layers*. For typical metals in a free-electron approximation, bulk superconductivity is expected in isolated grains with characteristic size greater than 30 Å. The stated thicknesses of films are *nominal* in that they are determined from summing the mass increments of material deposited on the thickness monitor during each deposition and using the standard bulk density.

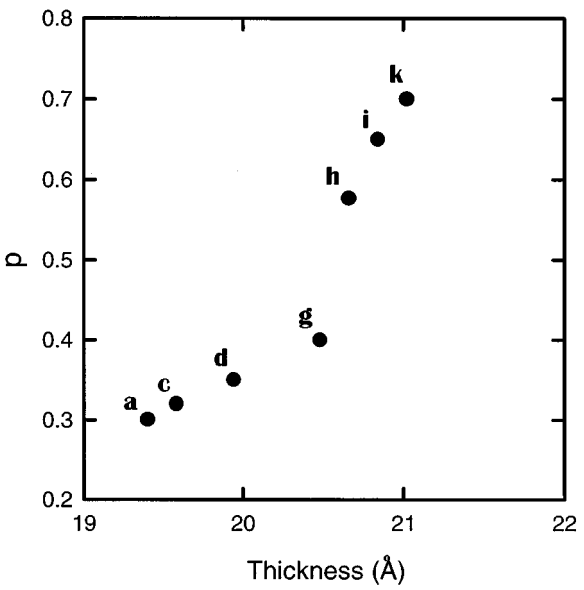

FIG. 3. The metallic grain fraction p as a function of the film thickness. The small bold letters correspond to set B of Fig. 1 in Ref. 2. We have assumed constant thickness increments of 0.18 Å between consecutive films in the cited set B.

The latter may be greater than the real film density; then the real thickness may be greater than the nominal thickness. Thus, the real film may present a disorder structure that is not confined to the film plane. We are dealing with a (2+1)-dimensional system. The extra dimension reduces both percolation thresholds $\overline{p}$ and p_c but preserving the difference $p_c - \overline{p} \approx 0.4$; then the relative difference becomes more significant. This fact has a pronounced effect over the quasireentrant superconductivity.

For a true two- or three-dimensional system we can assume the probability p to be proportional to the nominal thickness, but for a real (2+1)-dimensional film the relation between p and the nominal thickness may be rather nonlinear. In Fig. 3 the fitted values of p versus nominal thickness of films (assuming constant increments of 0.18 Å between consecutive films in set B of Ref. 2) are plotted. Two different linear regions are present with a breakdown near p_c. This breakdown may suggest that the percolating backbone at p_c is not in the basal plane.

Finally we want to make some remarks about nonlinearity in current-voltage (I-V) characteristics. At low temperature ($T < T_c$) the experimental I-V characteristics[2] show a ''quasi-particle-like'' behavior for very thin (high-resistance) samples with an upturn in current. Thicker samples have ''Josephson-like'' behavior with an upturn in voltage. If the current-voltage characteristic of each component (metallic bond) contains a small nonlinear contribution, the nonlinearity is amplified because of the tenuous structure of the infinite percolating cluster at $p \gtrsim p_c$ and the sample as a whole shows similar nonlinearity.[22] This is the case for the ''Josephson-like'' I-V characteristics. Nonlinear behavior for $p < p_c$ usually arouses the picture of some sort of tunneling phenomenon.[23] This behavior is associated with ''quasi-particle-like'' I-V characteristics. At low temperature the thermal activation mechanism of the tunneling junctions is frozen and only in the presence of a sufficiently strong local

field may the junction become conducting. At higher voltage the ''quasi-particle-like'' characteristic would be Ohmic.

In our model we have not included Josephson coupling between metallic grains. In consequence, we cannot describe the I-V characteristics in the first stages of global superconductivity.[32] Moreover we cannot accomplish the description for the ''quasi-particle-like'' characteristics within the framework of linear response theory upon which Eq. (1.1) is based. From Eq. (1.3) we only obtain the linear relation $I=(1/CT)\omega_{\rm eff}V$. However, we can predict the threshold voltage V_c for tunneling conduction at low temperature. If E is the tunnel energy barrier, we have $V_c=E/e$ and this can be evaluated if we assume $E=k_BU$. Using the fitted value $U=12$ K results in $V_c\approx 1$ mV. This value is according to that suggested by the experimental results (see Fig. 2 in Ref. 2).

VI. CONCLUSION

In summary, we have presented an analytical solution of a tunneling percolation model for metallic granular films. Using the effective-medium approximation we have obtained explicit expressions for the resistance as a function of grain concentration p and temperature. Our results are in good agreement with the expected behavior from experiments. The agreement is excellent near p_c in spite of the oversimplified assumption taken in the model [see Eqs. (4.1)]. The fits suggest the importance of the film thickness in the dimensionality of the conduction process.

In our model we have omitted the Josephson coupling between superconducting grains. We have considered only tunneling of single particles between metallic grains. An important question that might be asked is at which point in the film evolution do Josephson junctions span in the sample and drive the superconductivity.[1] By working out our phenomenological model we have obtained a strong indication about the percolation mechanism that governs the quasireentrant superconducting regime, i.e., thermally activated hopping tunneling. The Josephson junctions must percolate in our framework for $p\gtrsim p_c$ and give the signature of the first stages of global superconductivity. The present work seems to strengthen the importance for the geometrical interpretation of quasireentrance based on the interplay between metallic and tunneling bonds, instead of a quantum collective effect supported by a Josephson network.

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