Percolation model for the superconductor-insulator transition in granular films

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(Received 31 October 2007; published 28 December 2007)

We study the temperature dependence of the superconductor-insulator transition in granular superconductors. Empirically, these systems are characterized by very broad resistance tails, which depend exponentially on the temperature, and the normal state resistance. We model these systems by two-dimensional random resistor percolation networks in which the resistance between two grains is governed either by Josephson junction coupling (Cooper pair’s tunneling) or by quasiparticle tunneling. Our numerical simulations as well as an effective medium evaluation explain the experimental results over a wide range of temperatures and resistances. Using effective medium approximation we find an analytical expression for the effective resistance of the system and the value of the critical resistance separating conducting from insulating branches.

DOI: 10.1103/PhysRevB.76.224528

PACS number(s): 74.81.Bd, 74.25.Fy, 74.50.+r, 74.81.Fa

I. INTRODUCTION

The disorder driven superconductor-insulator transition (SIT) in thin films has gained revived attention over the past few years mainly because of the possibility that it represents a quantum phase transition.1–3 Disordered superconductors can be categorically divided into two groups, granular and homogeneous.8 Experimentally, homogeneous samples are found to exhibit sharp superconducting transitions and the insulator and the superconductor phases seems to be much less universal.2–7,9–11 In these samples it was found that the temperature dependence of the sheet resistance, R(T), below the critical temperature, Tc, can be described by an inverse Arrhenius law as follows:

\[ R(T) \sim \exp(T/T_c) \]

where Tc is a constant.5 The R(T) curves following Eq. (1) have been observed for a large variety of granular superconductors and over a wide range of temperatures (down to temperatures below Tc/10). A typical example of the granular film’s microstructure is shown in Fig. 1 and a typical example for a set of measurements7 of R(T) for quench condensed Pb granular films is shown in Fig. 2(a). The latter figure shows that the considered system can be driven through the SIT as a function of the film’s mean thickness.

It is important to distinguish between local and global superconductivities. When cooling the sample, each grain of the array becomes separately and independently superconducting at the same critical temperature Tc (approximately equal to Bardeen-Cooper-Schrieffer’s critical temperature, Tc, in bulk samples), for both insulating and superconducting sides of the SIT. This is in contrast to high-Tc granular superconductors in which the grains overcome to the superconducting state over some wide temperature range.12 Therefore, the global superconductivity and SIT in the granular film is a result of a competition between Josephson coupling and phase fluctuations. There are two types of these fluctuations: thermal and quantum. The latter are due to the charging energy, which pins the charge, which is quantum conjugate to the phase. Thus, via the uncertainty principle, the phase fluctuates.

Each grain is described by a condensate wave function (a complex superconducting order parameter13,14) \( \Psi_i = \sqrt{\rho} \exp(i \phi_i) \). The density \( \rho \) of Cooper pairs on each grain is assumed to be a constant and only the phases \( \phi_i \) are allowed to fluctuate. To describe these fluctuations, one typically employs a Hamiltonian15–17

\[
\hat{H} = \frac{1}{2} \sum_{ij} E_{ij}^{(c)} n_i n_j + \sum_{ij} V_{ij} + \sum_{i,j \neq i\text{like}} \left[ 1 - \cos(\phi_i - \phi_j) \right] + \sum_{ij} (\phi_i - \phi_j) X_{ij} + \sum_{ij} h_{ij} X_{ij},
\]

where the first term is the charging energy, the third term

FIG. 1. A typical atomic force micrography photography (reproduced from Ref. 11) of Pb grains deposited on the insulating SiO2 plate. Each grain is individually superconducting with its own phase. The mean size of the grain is of the order of 20 nm, while the intergrain distance is 1 order smaller, i.e., is about 2 nm.
describing the coupling associated with the shunt resistor across the junction $(ij)$. By neglecting the dissipation terms in Eq. (2), it is possible to obtain some expressions for determination of the critical value of the Josephson coupling energy, below which the superconducting bond becomes disconnected, as a result of the combined effect of the thermal and the charging energy. Finally, Ref. 15 obtains a phase diagram (in terms of $E_c/E_J$, where $E_c$ is the energy of Coulomb blockade) and a condition when two neighboring grains become “phase locked.”

$$E_c/E_J = 1/\coth(E_c/E_J k_B T)^{1/2} = 1.$$  

We also propose a simplified version of phase dynamics (see Refs. 18 and 19 and results of de Gennes in Ref. 20 derived in the mean-field approach). The criterion for the two neighboring grains to become phase locked is

$$z E_c \geq k_B T + E_c,$$  \hspace{1cm} (3)

where $z$ is a parameter of the order of the number of the nearest-neighbor grains. That is, Eq. (3) conveniently puts thermal and quantum fluctuations on equal footing and just clarifies that if $E_c$ is larger than $k_B T + E_c$, the phase is locked, while in the opposite case it is not locked.

In this paper we present a model based on percolation to account for the observed temperature dependence of the resistance in granular superconductors. Our percolation approach is based on a two-dimensional (2D) random disordered array of grains; each neighboring pair represents a superconducting junction in which transport can be achieved either by Josephson tunneling or by quasiparticle tunneling, depending on the intergrain coupling and temperature. Our numerical simulations of such a system exhibit an exponential-like dependence of $R(T)$ over a large range of temperatures which is in good agreement with recent experiments. We find that the critical resistance that separates insulating from superconducting branches depends on the distribution of disorder and on the nature of the percolation network of the current trajectories.

The remainder of this paper is arranged as follows. In Sec. II, we describe our model and numerical scheme of simulations as well as results of simulations. In Sec. III, we present analytical evaluation of SIT in the framework of effective medium approximation (EMA), followed by a brief discussion in Sec. IV.

**II. MODEL AND NUMERICAL SIMULATIONS**

When the film is enough thin, it breaks into a set of spatially separated grains. The thinner is the film, the larger is the mean distance between the grains. Therefore, the different thickness of the film can be represented in our percolation model by different distributions of intergrain mean distances.

If two grains are sufficiently decoupled, the resistance between two neighboring sites, $i$ and $j$, is given by

$$R_{ij} = R_0 \exp(r_{ij}/r_0 + e_{ij}/k_B T),$$  \hspace{1cm} (4)

where $R_0 = T k_B / (e^2 \gamma_{ij}^0)$, $\gamma_{ij}^0$ is a rate constant related to the electron-phonon interaction [usually of the order $10^{12}$ s$^{-1}$, 

FIG. 2. (Color online) (a) Experimental plots of log$_{10} R$ vs $T$ for quench condensed Pb films (Ref. 7) with grain sizes of the order $\sim 5$–10 nm, having different intergrain coupling. The higher curves correspond to the thinner films, i.e., to films with larger intergrain distance and, therefore, to the stronger disorder which is determined by $\kappa$. (b) Theoretical plots of log$_{10} R$ vs $T$ for the systems with different disorder strengths [see Eq. (7)], $\kappa = 20, 19, 18, 17, 16, 15, 14, 13.5, 13, 12, 11, 10, 9$ (from top to bottom, respectively). Squares are the results of our numerical simulations, while the dashed lines are the plots of our analytical prediction (12). The sample size in simulations is $40 \times 40$ resistors, $T_c = 7$, $\alpha = 3.25 \times 10^3$, $z = 10$, and $R_0 = T$ (in dimensionless units as is defined in the text). The coefficient $\beta r_{ij} / E_c E_{10} k_B$ [see Eq. (8)] was taken in our calculations to be equal to 0.5 K.
enabling us to assume in our further consideration that \( k_B/\epsilon^2 r_j^0 \approx 1 \) and \( R_0 = T \) (in dimensionless units), \( r_{ij} \) is the distance between the two sites, \( r_0 \) is the scale over which the wave function decays outside the grain, and \( \epsilon_{ij} = (|E_j| + |E_i| + |E_j - E_i|)/2 \) is the zero field activation energy, which can be determined from physical principals. In the case of superconducting grains this is a nontrivial problem, but in general \( \epsilon_{ij} \) is related to the superconducting gap \( \Delta_j(T) \) and Coulomb energy \( E_{ij} \sim \epsilon^2/2C \) (where \( C \) is capacitance and \( \epsilon \) is the electron charge).\(^{19,27} \) \( \epsilon_{ij} = \Delta_j(T) + E_{ij} \). Since the grains are assumed to be large enough to sustain bulk superconductivity, we assume \( \Delta(T) \) is the same for all grains.

It is known that dissipation tends to suppress the quantum fluctuations.\(^{16,28,29} \) Consequently, there can be some changes in the phase diagram to take place in the presence of dissipation in the junctions.\(^{30} \) Therefore, in general, Eq. (3) should be modified accordingly. However, for simplicity, we do not consider this effect in this paper.

According to Ambegaokar-Baratoff formula, the Josephson coupling constant in Eqs. (2) and (3) can be written as\(^{31-33} \)

\[
E_J = a \left[ \Delta(T)/R_j^{(N)} \right] \tanh \left[ \Delta(T)/2k_BT \right],
\]

where \( a = \pi \hbar/4e^2 \approx 6.5/2 = 3.25 \) k\( \Omega \) and \( R^{(N)}_j \) is the local normal resistance (local resistance at \( \Delta = 0 \)) between the grains. The Josephson energy \( E_j \) is related to the Josephson current \( J_j \) as follows: \( E_j = (\hbar/2e)J_j \).

To perform numerical simulations of this model, we assume that the random distance between grains is \( r_{ij} = 2\tilde{t} \cdot \xi_{ij} \), where \( \xi_{ij} \) is a random number taken from a uniform distribution in the range \((0,1)\), i.e., \( 0 \leq \xi_{ij} \leq 1 \), and \( \tilde{t} \) is the mean distance between metallic grains.\(^{34} \) Therefore the term \( r_j/r_0 \) can be expressed as \( \kappa \xi_{ij} \), where \( \kappa = 2\tilde{t}/r_0 \) can be interpreted as the dimensionless mean hopping distance or as the degree of disorder\(^{35} \) (the lower density of the deposited grains represents larger \( \kappa \)). Similarly, the charging energy, \( E_c \), can be expressed through the same factor \( \kappa \xi_{ij} \) as follows:

\[
E_c = \beta \frac{2e^2}{4\pi \epsilon_0 \epsilon d} R_{ij} = \beta \left( \frac{r_0}{d} \right) E_c^{(0)} (\kappa \xi_{ij}),
\]

where \( d \) and \( \epsilon \) are the mean size of the grains and the dielectric constant, of the substrate, respectively.\(^{18,36} \) The value \( E_c^{(0)} = 2e^2/(4\pi \epsilon_0 \epsilon d) \) is a mean charging energy of a single grain, and \( \beta \approx 0.1 \) is the effective coefficient which was invoked as a result of the influence of the surrounding grains.\(^{18,37} \)

Finally we can rewrite expression (4) for the local net resistor mimicking the local hopping resistance between the grains in the convenient form as follows:

\[
R_{ij} = R_0 \exp \left[ \kappa \xi_{ij} + \Delta(T)/k_BT \right],
\]

where

\[
\kappa = \kappa \left[ 1 + \beta(r_j/d)E_c^{(0)}/k_BT \right].
\]

The superconducting gap \( \Delta(T) \) is the solution of the integral equation\(^{13,31} \)

\[
\ln(\Delta(0)/\Delta) = 2[I(\Delta(T)/T)], \quad I(u) = \int_0^\infty \left\{ \sqrt{x^2 + u^2} \exp \left( \sqrt{x^2 + u^2} + 1 \right) \right\}^{-1} dx.
\]

For temperatures near the critical value \( T \approx T_c \), the gap \( \Delta(T) \) can be approximated by the analytical form\(^{13,38} \)

\[
\Delta(T) = 3.06k_BT_c(1 - T/T_c)^{1/2}.
\]

Since the Coulomb energy can be expressed through the parameter \( \kappa \) [see Eq. (6)], we can write a self-consistency equation for the upper value of \( R_{ij}^{(N)} \) for which Eq. (3) is fulfilled,

\[
R_{jc} = -\frac{z\kappa \left[ \Delta(0)/k_BT \right] \tanh \left[ \Delta(0)/2k_BT \right]}{1 + \beta(r_j/d)E_c^{(0)}/k_BT} \ln \left( R_{jc}/R_0 \right)
\]

Here \( R_{jc} \) is the local critical parameter where the subscripts \( ij \) have been omitted for simplicity. When \( R_{ij}(\Delta = 0) \approx R_{jc} \) the neighboring \( i \)th and \( j \)th grains are Josephson coupled. Here we have used the relation \( \kappa \xi_{ij} = \ln \left( R_{ij}^{(N)}/R_0 \right) / \left[ 1 + \beta(r_j/d)E_c^{(0)}/k_BT \right] \). The solution of Eq. (9) (i.e., the dependence of the critical resistance \( R_{jc} \) on \( T \)) for different \( \beta(r_j/d)E_c^{(0)} \) is shown in Fig. 3. In the case of small \( E_c \) (when \( E_j/E_c \rightarrow \infty \), i.e., in the case of classical SIT), inequality (3) can be reduced to a simple intrinsic condition:\(^{19,32,33} \)

\[
R_{ij} \approx \kappa \left[ 1 + \beta(r_j/d)E_c^{(0)}/k_BT \right] \ln \left( R_{jc}/R_0 \right)
\]

Next we aim to evaluate the total resistance of the network system. Our numerical simulations were performed considering a 2D bond-percolating resistor network where \( R_{ij} \) of each resistor is zero if Eq. (3) is fulfilled, otherwise it is given by Eq. (7). We solve the obtained system of linear Kirchhoff equations\(^{34,39} \) and calculate the total effective resistance, \( R(T) \), of the 2D network. The results are shown in Fig. 2(b). These results are in good agreement with the experimental data shown in Fig. 2(a). From Fig. 2 we can also see that the SIT is a result of an interplay between quasiparticle tunneling, which tends to turn the curves up [i.e., to increase resistance, \( R \), with decreasing the temperature, \( T \), see Eq. (7)] and Josephson coupling mechanism, which tend to turn curves down [i.e., to decrease the resistance, \( R \), due to increase the total number of the Josephson junctions, which is proportional to superconducting gap \( \Delta(T) \), see Eqs. (5)–(9)]. To qualitatively understand the behavior obtained in our simulations [which are very similar to the experiments (Fig. 2)], we describe the percolation mechanism leading to
this behavior. For large $\kappa$ (strong disorder), the current flows along a single path of the percolation cluster which is the path with minimal total resistance\(^{41}\) and the total resistance of the path is determined by few critical (red bonds)\(^{42}\) resistors.\(^{41,43}\) The tail decrease of $R$ in Fig. 2 can be understood when $T$ decreases (below $T_c$), $R_{JC}$ increases and more critical resistors become superconductors.

It is seen that an inverse Arrhenius dependence of $R(T)$ is obtained over a wide range of temperatures. Our agreement with experiments is further demonstrated in Fig. 4 which shows the dependence of $T_0$ [the slope of the tails, see Eq. (1)] as a function of the normal state resistance (resistance at $T=T_c$) of the sample, $R_N$, for both experimental and simulation results.

We conclude this section by estimation of the charging energy in SIT’s experiments\(^ {1-6,9}\) and its role in our calculations. A simple estimation (using the formula $\varepsilon^2/2C$ and typical grain’s sizes $\sim 20\text{ nm}$) gives the values of the order hundreds of Kelvin which is much larger than the critical temperature $T_c\sim 7\text{ K}$, and makes it not relevant to the SIT [does not fulfill condition (3)]. This problem is already discussed in literature. For example, in Ref. 44 it was assumed that the dielectric constant is larger than 10 leading to islands of grains. According to Abeles et al.,\(^ {36}\) to estimate the charging energy is not a trivial problem because of its complicated dependence on the grain size, separation, and the dielectric constant of the oxide coating. Using the results of Ref. 36, we presented the charging energy in Eqs. (6)–(9) as a product of the value usually used for Coulomb blockade estimation $E_c^{(0)}$ by a prefactor $\beta(r_o/d)$, which describes the features of the sample microstructure. So, even if $E_c^{(0)}$ can be of the order $\sim 10^{-10}-10^{2}$, the prefactor $\beta(r_o/d)$ is of the order $\sim 10^{-3}-10^{-2}$ and, therefore, their product $\beta(r_o/d)E_c^{(0)}/k_B$ written in Eq. (9) should be in the range $10^{-2}-1\text{ K}$. The curves shown in Fig. 2(b) are calculated for the case $\beta(r_o/d)E_c^{(0)}/k_B=0.5\text{ K}$. From Eq. (9) it is clear (see also Fig. 3) that the charging energy decreases the value $R_{JC}$ that suppresses the SIT. As a result the critical resistance (separating the metal-like branches from the insulatinglike) $R_{crit}$ appears at a smaller value compared to the case $E_c=0$. In order to bring it back to consistency with the experimental data [see Fig. 2(a)] one needs, e.g., to increase the value of the neighbors $z$, which requires special justification.

III. EFFECTIVE MEDIUM APPROXIMATION

For further understanding of this complex transition, we have also calculated the total effective resistance, $R$, of such a network using the symmetric self-consistency EMA\(^ {34,45}\). The effective resistance $R$ of the random conductance network [the local resistivities, $\rho$, of which are distributed continuously according to some distribution function $f(\rho)$] can be found as a solution of the integral equation

$$
\int f(\rho) \left( \frac{\rho - R}{\alpha \rho + R} \right) d\rho = 0.
$$

If $\xi$ in Eq. (7) is uniformly distributed between 0 and 1, then $f(\rho)=1/\kappa\rho$, and $\rho$ is varied in the range $R_0 e^{\Delta/k_B T} \leq \rho \leq R_0 e^{\Delta/k_B T + \kappa}$ [see Eq. (7)],

$$
\int_{R_0 e^{\Delta/k_B T}}^{R_0 e^{\Delta/k_B T + \kappa}} \theta(\rho e^{-\Delta/k_B T} - R_{JC}) \frac{1}{\kappa \rho} \left( \frac{\rho - R}{\alpha \rho + R} \right) d\rho - \int_{R_0 e^{\Delta/k_B T}}^{R_0 e^{\Delta/k_B T + \kappa}} \theta(R_{JC} - \rho e^{-\Delta/k_B T}) \frac{1}{\kappa \rho} d\rho = 0,
$$

where $R_{JC}$ is a critical resistance value determined by Eq. (9), $a=\pi z/2-1$, and $z$ is the number of bonds at each node of the network. Here we split the integral into two parts in order to take the Josephson coupling into account in accordance with condition (9), and have used the fact that $\rho_0=\rho_0 e^{\Delta/k_B T}$ [see Eq. (7)]. In the first integral we calculate the cases in which $\rho$ is larger than necessary for the Josephson coupling $\rho > R_{JC}$ [i.e., when $\theta(\rho - R_{JC})=1$, where $\theta$ is the Heaviside function]. In the second integral we consider the opposite situation, i.e., when $\theta(R_{JC} - \rho)=1$. In this case Josephson coupling exists and $\rho$ in the brackets should be taken as zero ($\rho \to 0$).

From Eq. (11) we can find the solution (for $R_0 e^{\Delta/k_B T} < R_{JC} \leq R_0 e^{\Delta/k_B T + \kappa}$)

$$
R = \frac{1-p_c}{p_c} \frac{(R_0 e^{\Delta/k_B T} - R_{JC}) e^{\Delta/k_B T}}{1 - e^{-k_B T(1-p_c)}},
$$

where $R_0$ is given by Eq. (9), and $p_c=1/(1+a)=2/z$ (see also Ref. 34).

Equation (12) can be understood qualitatively as follows: The total resistance of the system at $T>T_c$ [when $\Delta(T)=0$] and for large $\kappa$ is equal to $R_0 e^{\kappa \xi} [R(\beta_{0}+\Delta(T)) e^{\Delta/k_B T}$ (where $z=4$, i.e., $p_c=0.5$). As discussed above, if the system is strongly disordered, then its total resistance is determined by few resistors\(^ {34,43}\) along a path on the spanning cluster\(^ {26,34,35}\). At $T<T_c$, some of the grains along this path have $R_j(\Delta=0)$ smaller than $R_{JC}$ and, according to Eq. (9), these will be in the superconducting state. Therefore this resistance, proportional to $R_{JC}$, should be subtracted from the total resistance: $R=R_0 e^{\kappa \xi} - R_{JC} e^{\Delta/k_B T}$.

We expand Eq. (12) linearly near the critical temperature ($T-T_c$), from which we get an expression for $R_c$ linear in terms ($T_c-T$) as follows:
In the same approximation we get
\[ \ln[\frac{R}{R(T_c)}] = -(3.062 z \alpha/2T_c)(T_c - T). \] (14)

shown in Fig. 4 in comparison to experimental and numerical results. It should be noted that the system with exponents \( T_c/T \) and critical value of the effective resistance \( R_{cr} \) is the system resistance at \( T=T_c \). Note that the inverse Arrhenius law (1) follows immediately from the latter expression with
\[ T_0 = (2T_c/3.062 z \alpha)R_N, \] (15)

Theoretical results can also be used in order to find the critical value of the effective resistance \( R_{cr} \) separating the metal-like (\( \partial R/\partial T > 0 \)) and insulator-like (\( \partial R/\partial T < 0 \)) behaviors. By taking the derivative and solving the equation \( \partial R/\partial T = 0 \), we get a self-consistency equation, which determines the critical value separating metal-like behavior (at \( R \leq R_{cr} \)) from insulator-like (at \( R > R_{cr} \)). The value of \( R_{cr} \) as well as \( R \), see Eq. (12) depends on two main factors: number of neighboring grains \( z \) and charging energy \( E_c \). For small \( E_c \) we get a simple expression
\[ R_c = z \alpha(1 - p_c/p_c). \] (16)

IV. SUMMARY

In summary we have modeled and studied the temperature dependence of the superconductor-insulator transition in granular superconductors. Our numerical simulations explain well the experimental results over a wide range of temperatures and resistances. Calculations of effective medium approximation also show excellent agreement with the experiments for temperatures close to \( T_c \). These calculations also enable us to determine the critical resistance value, separating the superconducting and insulating branches.

ACKNOWLEDGMENTS

This research was supported by grants from the US-Israel Binational Science Foundation, the Israel Science Foundation (Grant No. 249/05) and the European Research NEST Project No. DYSONET 012911. We thank V. Sandomirsky, B. Shapiro, I. Shlimak, L. Burlachkov, E. Kogan, D. Stauffer, Y. Avishai, and P. Sheng for valuable conversations. We thank J. M. Valles for providing the data for Fig. 2(a).


8. Though some granularity can exist even in “homogeneous” superconductors.


