# Universal transport in two-dimensional granular superconductors

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The transport properties of quench condensed granular superconductors (Pb, Sn, and Pb-Ag films) are presented and analyzed. These systems exhibit transitions from insulating to superconducting behavior as a function of intergrain spacing. Superconductivity is characterized by broad transitions in which the resistance drops exponentially with reducing temperature. The slope of the log $R$ versus $T$ curves turns out to be universally dependent on the normal state film resistance for all measured granular systems. It does not depend on the material, critical temperature, geometry, or experimental setup. We discuss possible physical scenarios to explain these findings.

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Two-dimensional granular superconductors, i.e., systems of superconducting grains embedded in an insulating matrix, exhibit a superconductor to insulator transition (SIT) as a function of the intergrain distance. Such systems are of interest since they can serve as model systems for many dirty superconductors. In particular it has been suggested that a granular superconductor can mimic the behavior of high-$T_C$ superconductors and its properties can be used to explain some of the features observed in the cuprates.[1]

An established technique to study the properties of granular superconductors is quench condensation.[2–6] In this method one performs sequential evaporations on a cryogenically cold substrate under UHV conditions using the following scheme. Metallic leads for four-terminal measurements are prepared on an insulating substrate which is then mounted onto an evacuated measurement probe and immersed in a liquid He bath. The low temperature of the probe causes cryopumping and hence the substrate is situated in UHV conditions and, at the same time, at cryogenic temperatures. This enables evaporation of ultraclean thin superconducting layers on a substrate held at temperatures lower than 10 K while continuously monitoring the film resistance and thickness. Once a desired resistance (or film thickness) is achieved, the evaporation is terminated and the transport properties are measured. Incremental layers of material are then added *in situ* and further measurements are taken at different film resistances. Using this method one can study the properties of a single sample throughout the entire transition from an insulator to a superconductor as a function of the amount of deposited material while keeping the sample at low temperatures and in a UHV environment without having to thermally cycle it (risking metallurgical or structural changes due to annealing) or to expose it to atmosphere (thus oxidizing the surface).

If the samples are quench condensed on a passivated substrate (such as $SiO_2$), they grow in a granular manner so that the film begins its growth as disconnected islands with diameters of 100–200 Å.[6–8] The average distance between the islands decreases upon adding material. Beyond a percolation threshold, the grains connect, forming a continuous conducting layer. In these samples there is a critical nominal thickness $d_C$ below which no conductivity can be measured (the sheet resistance $R_\square$ is larger than $10^{10}\Omega$). Once the thickness, d, of the sample is larger than $d_C$, $R_\square$ drops exponentially with thickness until, for $R_\square \lesssim 6k\Omega$, it crosses over to a normal Ohmic behavior ($R_\square \propto 1/d$).[8]

Varying the thickness of the film causes a transition from an insulating behavior for the thinnest films to a superconducting behavior for thick films. Figure 1 demonstrates examples for this transition in three different granular superconductors: a Pb film (critical temperature $T_C \approx 7.2$ K), a Sn film ($T_C \approx 4.5$ K), and a Pb/Ag bilayer. The latter is a system in which a thin layer of insulating granular Pb is quench condensed on a $SiO_2$ substrate followed by sequential evaporations of Ag ultrathin layers.[1,9] The curves were measured in a shielded room using standard four probe techniques and assuring, for each point on the curve, that the $I$-$V$ characteristics are in the linear regime. The three systems show a transition from an insulator for thin films to a superconductor at thicker films. In all 2D granular systems these transitions are characterized by broad resistance tails and critical temperatures which are not very well defined until the resistance of the film becomes low. The transitions become sharper as material is added to the film. In this paper we define $T_C$ as the temperature at which the resistance starts dropping exponentially with lowering the temperature. We justify this by observing that this is the temperature that the individual grains become superconducting even in the insulating case as discussed below. In the Pb and Sn films [Figs. 1(a) and 1(b)] $T_C$ has bulk value and barely changes throughout the entire transition. Moreover, even on the insulating side of the SIT the curve changes its slope at $T=T_C$ reflecting the presence of superconductivity even in the thinnest measurable samples. The fact that the grains are superconducting with bulk properties throughout the entire transition has been demonstrated by tunneling measurements.[10] A bulk energy gap was observed in the grains even when the film was on the insulating side of the SIT.

In the Pb/Ag film the $T_C$ decreases as material is added to the film. This is due to suppression of superconductivity in the Pb grains by the silver overlayer because of proximity effects. Nevertheless, the general behavior of the insulator to superconductor transition is very similar to that of the pure superconductors.

 

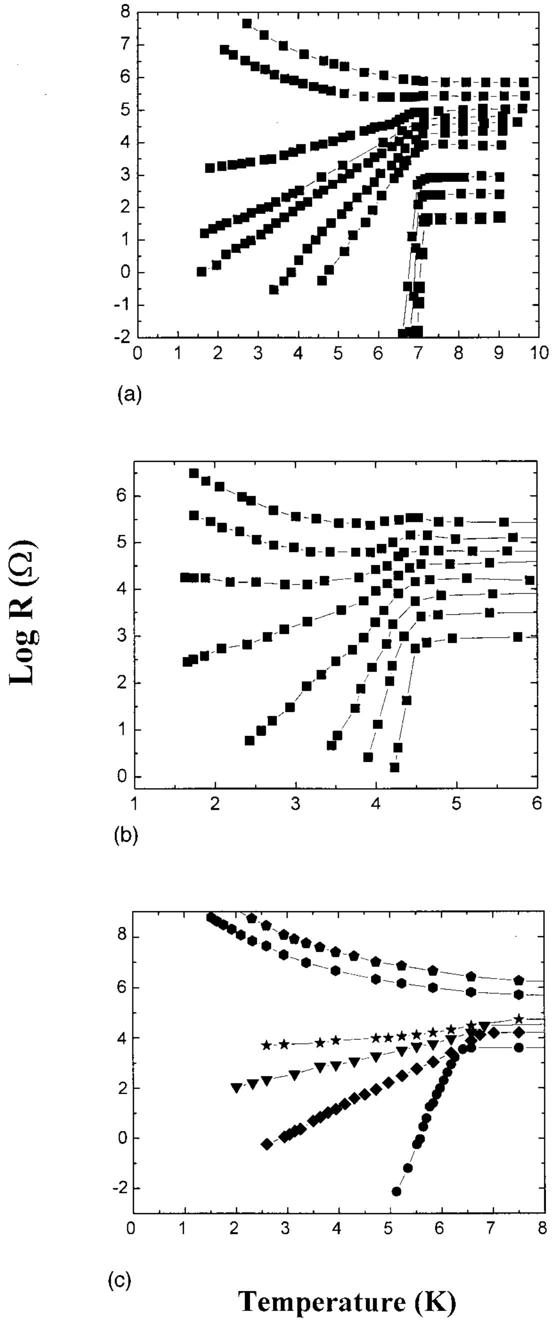


FIG. 1. Resistance versus temperature for sequential layers of quench-condensed granular Pb (Top), Sn (middle), and Pb/Ag (bottom). Different curves correspond to different nominal thickness. The nominal thickness of the top curves in the three graphs are 76, 52, and 78 Å, respectively. The lower curves in each graph are the results for sequential additional layers of thickness 0.5–2 Å.

These transport curves are attributed to the unique nature of the SIT in a granular structure.[11] In these systems each grain sustains superconductivity with bulk properties. However, for the high resistance samples, there are rapid, thermally activated phase fluctuations between the grains leading to an insulating $R$-$T$ curve. As the grains become closer the inter-grain Josephson coupling increases until phase coherence is achieved throughout the film.

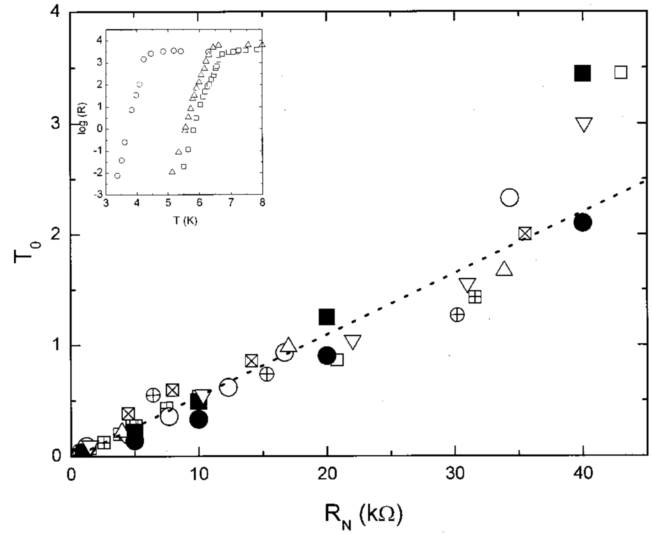


FIG. 2. The inverse slope of the $R$-$T$ curves ($T_0$ of Eq. (1)] as a function of the normal-state sheet resistance for different granular samples. The open symbols are the experimental data. Squares are results for various Pb samples, circles are for Sn samples and triangles for Pb/Ag proximity samples. Single symbols are used for the same quench-condensed films at different thicknesses (and hence $R_N$'s). Full symbols are the slopes extracted from the calculated samples of Fig. 3 (squares: Pb and circles: Sn). The line is a guide to the eye. Inset: Resistance versus temperature for granular Pb (squares), Sn (circles), and Pb/Ag (triangles) samples having a normal state resistance of 4 kΩ.

A striking feature common to the quench condensed granular superconductors is that at temperatures below $T_C$ the sheet resistance decreases exponentially with decreasing temperature and can be described approximately by an "inverse Arrenius" law

$$R = R_0 e^{T/T_0}. \tag{1}$$

It is important to note that we see no flattening of the $R$-$T$ curve ( resistance approaching a flat temperature dependence at low temperatures) in any of our samples and in no regime of thicknesses.[12] Nor does the $R$-$T$ change its trend and begin dropping to zero resistance at low temperature. The dependence described in Eq. (1) is observed in all of our granular samples using different materials and different measurement apparatuses. It spans many orders of magnitude in $R$, persists to temperatures below 100 mK (Ref. 13) and is observed for the different steps of the sequential evaporations providing the resistance is smaller than a few tens of $k\Omega$. Moreover, the slope of the log $R$ versus $T$ curves, $1/T_0$ of Eq. (1), turns out to be universal for all of our samples. It depends only on the normal state sheet resistance $R_N$ and does not depend on the material, the critical temperature, or sample geometry. We illustrate this in Fig. 2 where we show the dependence of the inverse slope $T_0$ on $R_N$ for a large number of granular superconductors (different materials, geometries and prepared in various quench condensation evaporators). It is seen that all the slopes fall on a master plot having the form of

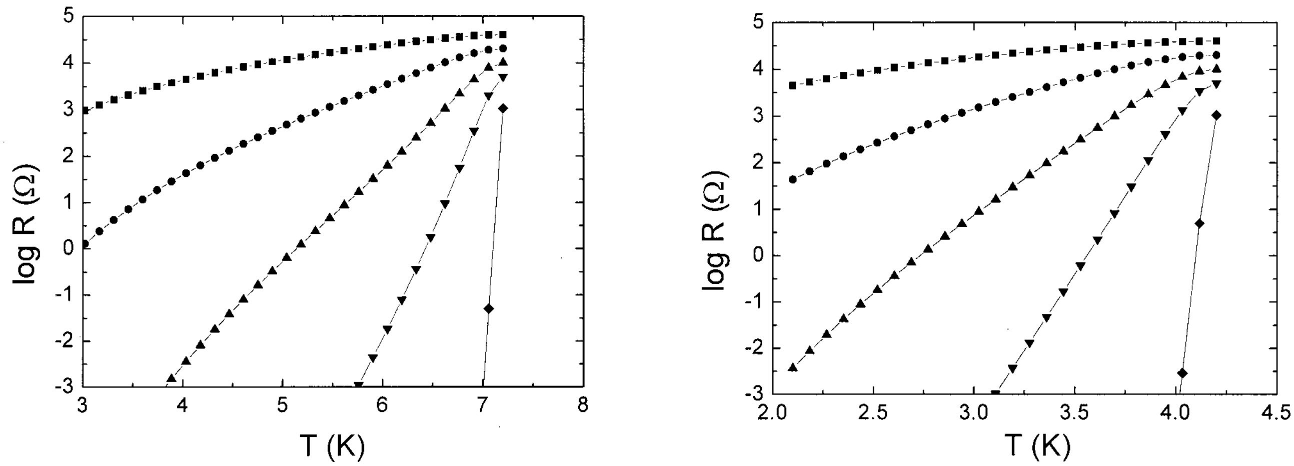


FIG. 3. Numeric simulations of the resistance of arrays of ten junctions in series based on Eq. (5). The right graph is for Sn and the left for Pb.

$$T_0 \approx C^* R_N, \tag{2}$$

where C* is a constant of approximately 0.05 K/$k\Omega$. Note that we have included samples having different critical temperatures as well as the Pb/Ag system in which $T_C$ varies with the thickness (and hence with $R_N$).

This observation indicates that the behavior of the superconducting tails does not depend on the properties of the superconducting grains themselves but only on their geometrical arrangement (density, intergrain spacing, morphology configuration, etc.) which determines the tunneling percolation network and $R_N$. Two superconductors having the same $R_N$ but different critical temperatures $T_{C1}$ and $T_{C2}$ will have parallel superconducting R-T tails, at temperatures lower than the respective $T_C$, shifted by $T_{C1}-T_{C2}$. An example for such behavior is shown in the inset of Fig. 2.

We can now combine this observation with Eqs. (1) and (2) and extract the expression for $R_0$ of Eq. (1):

$$R_0 = R_N e^{-T_C/T_0} = R_N e^{-T_C/C^* R_N}. \tag{3}$$

$R_0$ is the value of the resistance obtained by extrapolating the $R$-$T$ curves to $T=0$. This means that if the observed exponential $R$-$T$ curves persist to zero temperature the samples will exhibit a finite zero-temperature resistance which will depend both on $R_N$ and on $T_C$.

In an attempt to examine possible intuitive understanding of the observed phenomena we note that at temperatures below bulk $T_C$ the grains have been shown to be fully superconducting.[10] Hence, each two grains are expected to be Josephson coupled with a Josephson binding energy of the form

$$Ej = \frac{\pi\hbar}{4e^2}\frac{\Delta(T)}{R_N}\tanh\frac{\Delta(T)}{2K_BT}, \tag{4}$$

where $\Delta(T)$ is the temperature-dependent superconducting gap and $R_N$ is the normal resistance between the grains. One can expect that the ratio $Ej/K_BT$ would determine whether the grains are phase coupled or not. As the temperature of a granular system is lowered $Ej$ becomes larger than $K_BT$ in an increasing number of pairs of grains thus increasing the effective superconducting regions which are phase coherent. This leads to a characteristic "phase-coupled" length which grows with decreasing temperature until superconducting percolation is achieved.

However, the naive model described above can not explain the observed broad superconducting tails in our films. Since the samples are two dimensional, scaling considerations imply that as long as superconducting percolation is not achieved throughout the sample, the resistance of the granular system should be independent on the size of the superconducting clusters. The growth of the phase coherent clusters with decreasing temperature would not change the overall 2D resistance. One can expect to see a flat resistance versus temperature curve below the percolation threshold and a sharp superconducting transition at the percolation threshold. The fact that the experimental transitions are broad imply that the resistance of the individual junctions is temperature dependence. This urges us to consider superconductor fluctuation effects as fluctuations introduce a broad temperature dependence in a Josephson junction resistance rather than a sharp transition at $Ej=K_BT$.

The temperature dependence of the zero-bias resistance of each pair of grains due to thermal phase fluctuations is expected to take the Ambegaokar-Halperin form[14,15]

$$R(T) = \frac{R_N}{[I_O(Ej(T)/K_BT)]^2}, \tag{5}$$

where $I_0(X)$ is the modified bessel function of order 0. Using this model we have calculated the temperature dependence of 1D arrays of junctions subject to thermal fluctuations. The results of the simulations for Pb and Sn samples having different $R_N$ are shown in Fig. 3. It is seen that $R(T)$ may qualitatively mimic an exponential dependence for temperatures larger than about $T_C/2$. As $R_N$ is reduced the $R$-$T$ curves become sharper in a similar way to that seen in the experiments. While the curves are not truly $e^T$ dependent for purposes of illustration we can extract a slope from each curve. The extracted slopes are plotted on the master-plot of

Fig. 2 showing relatively good agreement with the experimental results. We can not expect better than qualitative agreement as this is a simple 1D chain model.

Hence, the thermal-fluctuation picture can provide some insight to the physics of the 2D granular films. However, two major points in which the simple model deviates from the experimental data have to be noted: In the first place the theoretical model predicts that the slopes should depend on $T/T_C$. Our simulations fail to reproduce the $T_C$ independence and the ''universality'' of the experimental transition tails. The calculated curves for Sn and Pb samples having the same $R_N$ yield Sn slopes which are consistently sharper than those of Pb. Secondly, the thermal fluctuations can imitate the experimental behavior only for a limited range of temperatures. At low temperatures the thermal fluctuations are expected to freeze out causing the resistance to drop much more rapidly, until, for zero temperature, the resistance reaches zero ($R_0=0$). We do not see signs for such a tendency in the experiments even at very low temperatures. We have considered the possibility that at low temperatures quantum phase fluctuations dominate the behavior and induce a finite resistance even at $T=0$. The presence of such effects is consistent with the observed experimental dependence of $R_0$ on $T_C$ as shown in Eq. (3) since the quantum fluctuations are expected to decay exponentially with $Ej$. However, we are not able to reproduce the experimental exponential *R-T* curves over the entire temperature range using a model which combines thermal fluctuations and quantum fluctuations. Such a model would suggest that at low temperature the *R-T* curve would flatten out and saturate at a constant value. As noted above, we do not observe such behavior in any of our samples. Clearly, further theoretical treatment is required in order to shed more light on the origin of the findings presented in this paper.

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