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Electric-field tuning of the superconductor-insulator transition in granular Al films

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The superresistive phase of thin granular films near the superconductor-insulator transition is believed to result from the suppression of Josephson tunneling by the large charging energy, E_c, of the grains. Our ac I-V studies of granular Al films suggest that a small dc bias voltage can be used to overcome this Coulomb barrier and to recover Josephson tunneling. This bias, in fact, drives a critically disordered film from the superresistive phase to a quasisuperconducting phase.

It is generally agreed that superconductivity in ultrathin granular films is mediated by intergrain coupling. The order parameter of the system can be written[1] as $\Delta^{1/2}e^{i\phi}$, where the amplitude, Δ, is the superconducting gap and ϕ reflects the phase coherence between the grains. Tunneling experiments[2] have found robust superconductivity on the grains, with the gap and the superconducting transition temperatures, T_c, being close to the values of the bulk material even in insulating granular films. Hence, at low temperatures, there is always local superconductivity on the grains. Global superconductivity is established via Josephson tunneling if there is sufficient long-range phase coherence.[3,4]

It was proposed[5] many years ago that the dephasing mechanism that destroys long-range phase coherence in granular system is grain charging. This is the electrostatic energy, $E_c = e^2/2C$, that must be overcome in order to move an electron onto an isolated grain of capacitance C. E_c could be quite large if the grains are small and sufficiently isolated. This charge barrier, which is also known as the Coulomb blockade,[6] suppresses charge fluctuations. This leads to fluctuations in the phase variable due to the number-phase uncertainty relation for the quantum condensate.[7] This model predicts the destruction of global superconductivity if E_c is larger than the Josephson coupling energy E_J.[5,8] More sophisticated theories[9,10] found that dissipative degrees of freedom reduced fluctuation effects, leading to a threshold normal-state sheet resistance R_N, of the order of $R_Q = h/4e^2 \approx 6.45\ \mathrm{k}\Omega/\square$, that characterized the onset of global superconductivity. However, truly insulating behavior is only observed in films with $R_N > 50\ \mathrm{k}\Omega/\square$, which is much larger than R_Q. In such films, the characteristic E_c is of the order of Δ.[11] When Cooper pair tunneling, which costs an energy of $4E_c$, is no longer favorable compared to quasiparticle tunneling whose energy cost is $2\Delta + E_c$, the superresistive behavior[3,4,11,12] is seen in insulating granular films.

In this paper, we present data which suggest that a longitudinal dc bias voltage, V_{bias}, applied to a granular film in the superresistive state will tilt the Coulomb barrier potential and reestablish charge flow. This increases charge fluctuations which lead to a suppression of the phase fluctuations,[7] and recovers, at least partially, the Josephson tunneling. Our data, in fact, show that, upon biasing, the ac resistance of a superresistive granular film will drop to a value well below its *normal-state* resistance R_N, indicating the existence of superconducting coupling. This electric-field-tuned superconductor-insulator (S-I) transition is essentially in accord with the I-V characteristics of small superconducting single electron tunnel junctions recently reported by Iansiti *et al.*[13] and by Geerligs *et al.*[14]

Ultrathin granular Al films were made by a standard electrochemical anodization process which reduced, in a controlled fashion, the thickness of 20-nm thick Al films evaporated at room temperature. The area of the films was 1.3 mm $\times$6 mm. A multilead pattern was used in order to check large-scale homogeneity by measuring four-probe resistances over three different sections of a film. Only films with section resistances differing by no more than 10% were used. More details about film preparation, scanning force microscopy images, and large-scale homogeneity are discussed elsewhere.[15] Resistances and ac I-V characteristics were measured in a four-probe configuration using a lock-in amplifier operating at 27 Hz. Probe currents of 10 nA were used on films with $R_N < 50\ \mathrm{k}\Omega/\square$ and 20 pA on higher resistance samples. We have also measured dc I-V's and found that the ac I-V's agreed with the numerical derivative of the dc I-V's. Data was taken in the temperature range 0.4 K$<T<$15 K with magnetic fields, $H_\perp$, up to 9 T applied perpendicular to the film plane.

Shown in Fig. 1 is the temperature dependence of sheet resistance, R, in zero field for films of varying R_N. The behavior seen here is typical to granular superconducting films.[3,4,12] First, the transition temperature, $T_c \approx 1.8$ K is not sensitive to R_N. Secondly, samples with $R_N \sim 100\ \mathrm{k}\Omega/\square$ display quasireentrance and superresistive behavior. It is known that in low resistance films, $R_N < R_Q$, global superconductivity is established via Josephson tunneling between the grains.[3,4] However, as R_N is increased above R_Q, the Josephson coupling energy, $E_J = (R_Q/2R_N)\Delta$, becomes small compared with E_c and global superconductivity is lost. This is reflected in the finite resistance tails at low temperature, such as seen in curve (c) of Fig. 1. Finally, at high enough sheet resistance, $R_N > 50\ \mathrm{k}\Omega/\square$, E_J is diminished to the point that what were formally Josephson junctions become more characteristic of S-I-S tunnel junctions with the current being primarily carried via quasiparticle tunneling. Since quasiparticle tunneling is exponentially attenuated below T_c, a sufficiently disordered film will display superresistive behavior[3,4,12] as can be seen in curve (e) of Fig. 1.

Of particular interest is the differential resistance, dV/dI, as function of the dc bias voltage, V_{bias}, which displayed

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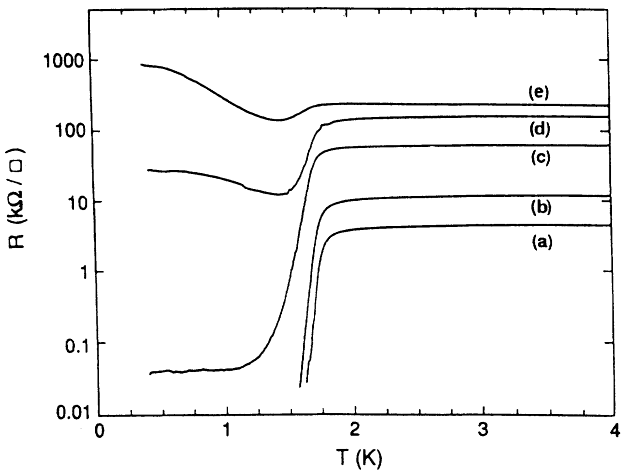

FIG. 1. Sheet resistances versus temperature for several Al films of varying R_N. Low resistance films display robust superconductivity, while quasireentrance and superresistive behavior are seen in high resistance films. Curves are labeled from (a) to (e) with increasing R_N.

unusual zero-bias anomalies, as shown in Fig. 2 for film (e) at 0.9 K. Similar anomalies were also found in earlier studies of dc I-V's of quench condensed granular films in the superconducting state,[4,12] and were explained as S-I-S tunneling between the superconducting grains.[12] In order to distinguish the anomalies associated with the normal state from those of the superconducting state, it is imperative that studies of the normal state be made at low temperatures, $T \ll T_c$. This can be done by applying a large enough magnetic field to suppress superconductivity. The zero-bias anomaly in the $H_\perp = 7$ T curve in Fig. 2 is typically seen in our high resistance films ($R_N > 10$ kΩ/□). The maximum upper critical field in thin Al films is known to be limited to 4–5 T by the

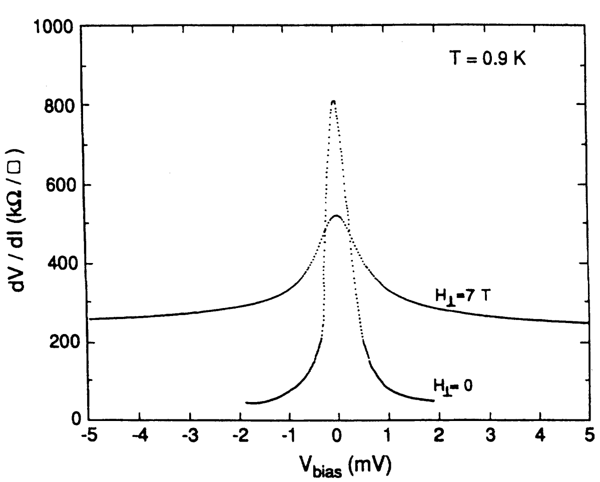

FIG. 2. Differential resistance, dV/dI, as a function of dc bias voltage, V_{bias}, for sample (e) in Fig. 1. For the $H_\perp = 7$ T curve, superconductivity is suppressed. For the $H_\perp = 0$ curve, ac resistance falls well below R_N (~ 250 kΩ/□) with increasing V_{bias}. The difference in the magnitudes of the zero-bias anomalies in the two curves is due to the superconducting gap in the quasiparticle density of states.

spin paramagnetic effect.[11,16] Since this limit is determined by the Cooper pair spin coupling to the applied field, it is independent of grain size. The perpendicular critical field is always lower than the spin paramagnetic limit and in our case $H_{c2} \sim 1.5$ T. We have made extensive measurements of the upper parallel critical field in our samples[11] and have found that their spin paramagnetic limit is 4 T. Therefore the $H_\perp = 7$ T curve in Fig. 2 represents the true normal state behavior. Thus even in the normal state there is a strong zero-bias anomaly. This anomaly is similar to the charging anomaly discussed by Giaever and Zeller.[17] We believe that the zero-bias anomaly in the normal state I-V curve is due to a Coulomb blockade effect.

This conjecture is supported by the fact that our films are granular. In the normal state, transport is dominated by normal metal-insulator-normal metal (N-I-N) tunneling between the grains. It is known that bulk N-I-N tunneling is Ohmic[18] for bias voltages much less than U_{barrier}/e, the barrier height for the insulator. In our case the tunnel barrier is Al_2O_3 with $U_{\mathrm{barrier}}/e > 1$ V. Therefore the normal state zero-bias anomaly is certainly a grain charging effect. A crude estimate of the effective grain charging energy,[19] $E_c = e^2/(4\pi\varepsilon_0 d\kappa)$, gives a value of order 1 K, where $\kappa = \varepsilon[1 + d/(2s)] \sim 160$ with ε_0, $d \sim 30$ nm, $s \sim 1$ nm, and $\varepsilon \sim 10$ being the vacuum permittivity, the size of the grains, the typical grain separation, and the dielectric constant for Al_2O_3, respectively. However, the width of the zero-bias anomaly in the $H_\perp = 7$ T curve is much smaller than the product of the effective grain charging barrier, E_c/e, and the number of grains, $N_s \sim 10^5$, in series along a transport channel. From our film homogeneity tests,[15] we can rule out the possibility of transport dominated by a few extremely high resistance junctions. In addition to the narrow width, the normal state I-V curve in Fig. 2 has a peculiar curvature that is not consistent with a simple Coulomb blockade voltage threshold for conduction.

For a better understanding of the normal state I-V's, we have applied a simple dipole ionization model to our data. In this model, if an electron tunnels from a neutral grain to another nearby neutral grain, it effectively creates a charge-anticharge pair. Such pairs have been studied extensively in Josephson junction arrays and are known as soliton-antisoliton pairs.[20] At $T=0$ there can be no conduction unless free solitons are produced by an applied electric field. For simplicity we have considered the energy of an unscreened charge-anticharge pair:

$$U(r) = E_c - e^2/(4\pi\varepsilon_0\kappa r) - e(V/L)r, \qquad (1)$$

where r is the pair separation, V is the applied bias voltage, and L is the sample length. The first term is the charging energy required to create a nearest-neighbor pair, the second term is the Coulomb interaction energy between the pair members, and the last term is due to the applied electric field $E = V/L$. It can be shown that $U(r)$ has a local maximum at $r_c = \gamma/E^{1/2}$ with a barrier height $U(r_c) = E_c - 2e\gamma E^{1/2}$ where $\gamma = (e/4\pi\varepsilon_0\kappa)^{1/2}$. The width of the barrier is proportional to $1/E$. Conduction is mediated by the ionization of the pairs, which can occur either by thermal activation over this barrier or by electric field induced tunneling through the barrier. Therefore, conductivity is given by

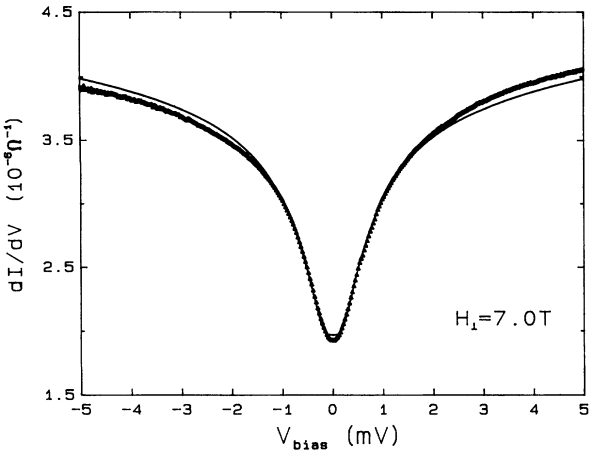

FIG. 3. Differential conductance versus bias voltage at $T=0.9$ K for sample (e) in Fig. 1. The solid line is a least-squares fit by Eq. (2).

$$\sigma(T,V)=\sigma_B+\sigma_A\exp\left(\frac{E_c-\alpha V^{1/2}}{k_BT}\right)+\sigma_T\exp\left(-\frac{\beta}{V}\right), \quad (2)$$

where $\alpha=2e\gamma/L^{1/2}$ and β are constants, σ_A and σ_T are the conductivity prefactors for thermal activation and tunneling, respectively, and σ_B is a temperature and field independent background conductivity.

Shown in Fig. 3 as points is the differential conductance corresponding to the $H_\perp=7$ T curve in Fig. 2. To compare Eq. (2) with data in Fig. 3, we first fit the temperature dependence of the conductance at zero bias, $V=0$, to determine σ_A,σ_B, and E_c. Taking $\alpha/k_B\sim 1K/V^{1/2}$, this leaves β and σ_T to be determined from fits to data in Fig. 3. The solid line is a least-squares fit by Eq. (2) with $\sigma_A=5.8\times10^{-7}\ \Omega^{-1}$, $\sigma_B=1.9\times10^{-6}\ \Omega^{-1}$, $\sigma_T=2.1\times10^{-6}\ \Omega^{-1}$, $\beta=0.74$ mV, and $E_c/k_B=1.4$ K. Though the data has a slight asymmetry that is not accounted for in the model, the fit is quite good. Not only does Eq. (2) correctly predict the curvature of the I-V but, in fact, gives a value for $E_c/k_B\sim1.4$ K which is in good agreement with the estimate given earlier. More extensive analysis of the normal state I-V's at various temperatures and sheet resistances are reported elsewhere.[11]

Notwithstanding the quality of the fit in Fig. 3, there remains a puzzle as to why the normal state zero-bias anomaly is so narrow. Naively one would expect a width of order $N_sE_c/e\sim$ volts. It is useful to compare our normal state I-V data with data taken from nanofabricated arrays of small normal tunnel junctions. I-V studies of two-dimensional arrays of normal tunnel junctions reveal[20] a somewhat more complex behavior than suggested by the simple addition of local charging energies. At low temperatures, I-V characteristics of normal arrays typically display[20] a very well-defined low voltage conduction threshold, V_t, that can be more than an order of magnitude lower than N_sE_c/e. Recent theoretical work[21] on the low-temperature transport properties of arrays has suggested that V_t is a threshold for soliton-antisoliton ionization at the array-electrode interface. In this process, a charge is injected into the array and subsequently interacts with its image charge in the electrode. Since this free charge production mechanism is an edge effect, it does not scale with N_s. In fact, it only depends upon the geometry of the device, $V_t\sim(E_c/2e)(C/C_0)^{1/2}$, where C is the junction capacitance and C_0 is the stray capacitance of the electrode. Due to the disordered nature of our samples, it is not possible for us to measure C and C_0 directly. However, as in the junction arrays, C/C_0 could be the order of 10^2 to 10^3 which would suggest a threshold voltage of a few mV. Allowing for rounding due to the distribution of grain sizes and junction resistances, this is about the width of the curve in Fig. 3. Our data seem to indicate that the carriers are being produced by field ionization of charge-anticharge pairs at the contact electrodes.

We now will turn our attention to the $H_\perp=0$ curve in Fig. 2. At zero bias, the differential resistance is higher than in the $H_\perp=7$ T curve. This is due to an additional attenuation of conductivity as a result of the superconducting gap opening up on the grains. Very interestingly though, when the sample was biased the differential resistance decreased a factor of 50 below the unbiased value. More significantly, the differential resistance falls a factor of 6 below the *normal state* sheet resistance R_N. At higher dc bias (not shown in Fig. 2), the ac I-V displayed a critical current of about 0.2 μA at which the resistance increased sharply to R_N. Superficially, this behavior is similar to that of a S-I-S tunnel junction in that the resistance is high at zero bias and lower at high bias. In fact, the differential resistance of a S-I-S function at $V_{\rm bias}=2\Delta/e$ can be an order of magnitude lower than its normal state resistance. At higher bias, the resistance of a S-I-S tunnel junction approaches its normal state value.[17] Despite the similarities, we do not feel that the data in Fig. 2 can be explained by an array of S-I-S tunnel functions. The first problem is that the width of the $H_\perp=0$ curve is much less than $N_s(2\Delta/e)$. In fact, it is very close to the width of the normal state curve. Secondly, we did not see a local minimum in the differential resistance that can be associated with the usual S-I-S tunneling conductance peak at $V_{\rm bias}=2\Delta/e$. Finally, at much higher bias than is shown in Fig. 2 we saw what appeared to be critical current behavior.

The fact that the $H_\perp=0$ curve in Fig. 2 has a width that is the same as in the $H_\perp=7$ T curve indicates that the bias

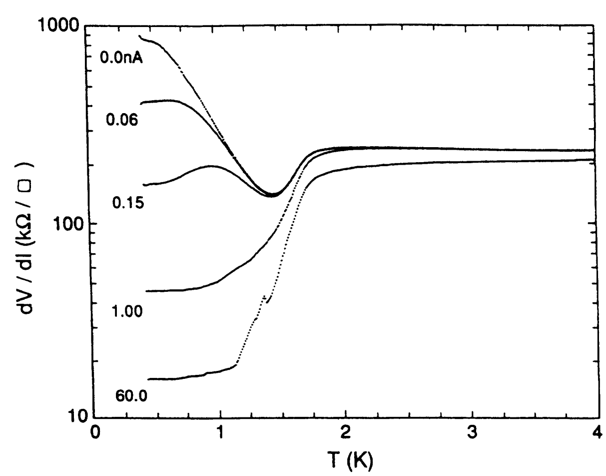

FIG. 4. Differential resistances versus temperature for several dc bias currents for sample (e) in Fig. 1. The values of the dc bias current for each curve are shown.

voltage scale in the $H_{\perp}=0$ curve is also set by the charging barrier seen in normal state curve. This suggests that the $H_{\perp}=0$ curve in Fig. 2 represents a suppression of the Coulomb blockade by the dc bias voltage which, in turn, partially reestablishes Josephson tunneling and phase coherence. This picture is well supported by experiments of Iansiti *et al.*[13] on small single junctions in which they observed a zero-bias charging anomaly, characterized by an extremely high zero bias resistance. However, upon applying a bias voltage of order E_c/e they found that Josephson coupling was restored with critical current behavior clearly evident in their I-V's. In I-V studies of small arrays of superconducting tunneling junctions, Geerligs *et al.*[14] also observed a supercurrent due to Cooper pair tunneling when the bias voltage was increased to above the Coulomb gap. Our data is also in qualitative agreement with dc I-V curves reported by Jaeger *et al.*[4] in Ga films and by Barber and Glover[12] in Pb films. We analyzed the numerical derivative of their dc I-V's and found that under bias the resistance of their superresistive samples decreased a factor of 2 to 10 *below* the normal state resistance.

Shown in Fig. 4 are resistances versus temperature at different dc bias currents for film (e). Figure 4 is complicated by the fact that a constant dc bias current was used instead of constant dc bias voltage. Nevertheless, these curves show characteristics similar to those of Fig. 1. The fact that the low-temperature residual resistance in the curve under the highest dc bias current (60 nA) is still not zero tells us that the length scale of the phase coherence is still much shorter than sample dimension.

In summary, our ac I-V studies of granular Al films demonstrate that a small dc bias voltage can be used to overcome the charge barrier in marginally superresistive samples and thereby partially recovering Josephson coupling between the grains.

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