Superconductor-Insulator Transition in a Parallel Magnetic Field

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(Received 11 April 1994; revised manuscript received 25 July 1994)

We report the use of a parallel magnetic field to suppress the superconducting gap and intergrain Josephson coupling in granular Al films. The critical field $H_{c\|}$ is spin paramagnetically limited, and the field induced superconductor-insulator (S-I) transition is found to go from second order to first order at 250 mK. In the vicinity of $H_{c\|}$, the field reproduces the quasireentrance and super-resistive behavior seen in the zero-field S-I transition. We show that the super-resistive phase is dominated by Coulomb blockade.

PACS numbers: 74.20.Mn, 74.25.–q, 74.50.+r, 74.80.Bj

The interplay between disorder and superconductivity in low-dimensional systems continues to be of great current interest. In particular, studies of the superconductor-insulator (S-I) transition in ultrathin metal films [1,2] have yielded a number of fascinating manifestations of the inherently quantum nature of these systems. Both nominally uniform [1] and granular [2] thin films tend to lose their zero resistance state at a normal state sheet resistance R_N that is close to the quantum pair resistance $R_Q = h/(2e)^2$. Uniform films, in fact, undergo a sharp S-I transition near R_Q, while a truly insulating state is not seen in granular films until $R_N \gg R_Q$. In addition, granular films display quasireentrance which is not observed in uniform films [2,3]. A detailed understanding of the morphological origins of these differences continues to be an outstanding problem in the field.

In contrast to uniform films, it is believed that the granular S-I transition is mediated by competition between grain charging and Josephson coupling. Granular systems are, in principle, susceptible to charging due to the fact that to put an electron on an isolated grain of capacitance C costs an electrostatic energy $E_c = e^2/2C$ which can become very large if the grains are small. This energy barrier is commonly known as Coulomb blockade and will be detrimental to Josephson tunneling [4] if E_c is comparable to or larger than the Josephson energy E_J. Recent theoretical work [5,6] has supported this conjecture, but there has been little direct experimental evidence as to the exact role of grain charging in the granular S-I transition. In this Letter, we present a study of the parallel magnetic field tuned S-I transition in granular Al films. Parallel field $H_\|$ was chosen to avoid complications associated with vortex dynamics [7], thus allowing us to straightforwardly vary the relative magnitudes of the superconducting gap Δ and Josephson coupling energy E_J with respect to E_C. In addition, as we will show below, the parallel field transition offers a unique opportunity to study a new first-order S-I transition.

Most studies of the S-I transition in ultrathin superconductors have been on films quenched-condensed new liquid helium temperature [1,2]. Film morphology has not been studied directly due to obvious technical difficulties. We have developed a novel technique for fabricating thin granular Al films at room temperature whose morphology can be directly imaged by scanning force microscopy. Our films were made by a standard electrochemical anodization process which reduced, in a controlled fashion, the thickness of 20 nm thick Al films evaporated onto glass substrates [8]. By careful control of the anodizing voltage we could increase the resistance of a film from an initial value of 2 Ω/sq to as high as 10^6 Ω/sq. Scanning force microscopy images of films anodized to a few kΩ/sq showed a surprisingly narrow distribution of grain sizes of the order of 30 nm [8].

The film area was 1.3 mm $\times$ 6 mm. A multilead pattern was used to test for 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 a few percent were used. From perpendicular critical field measurements, the elastic mean-free path [9] was estimated to be of the order of 15 nm for a 10 kΩ/sq film near 2 K. Resistances were measured using a lock-in amplifier operating at 27 Hz. Probe currents of 10 nA were used on films with $R_N <$ 50 kΩ/sq and 20 pA on higher resistance samples. We also measured dc I-V's to check the consistency of our ac measurements. The data presented below was taken in either a ^{3}He refrigerator or a dilution refrigerator, giving a temperature range of $0.04 < T < 10$ K with magnetic fields up to 9 T applied parallel to the film plane. A superconducting transverse trim coil was used to compensate for sample misalignment. Results were found to be independent of the orientation of the probe current.

Shown in Fig. 1 is resistance versus temperature in zero field for several films of varying R_N. There are two important features of this figure that are generally believed to be generic to granular superconducting films [2]. First, the transition temperature, $T_c \approx 1.8$ K, is not sensitive to R_N. Second, films with $R_N \sim 100$ kΩ/sq display quasireentrant behavior. It has been well established that even in insulating granular films there is a well-defined superconducting gap Δ on the grains [10] for $T < T_c$. Hence,

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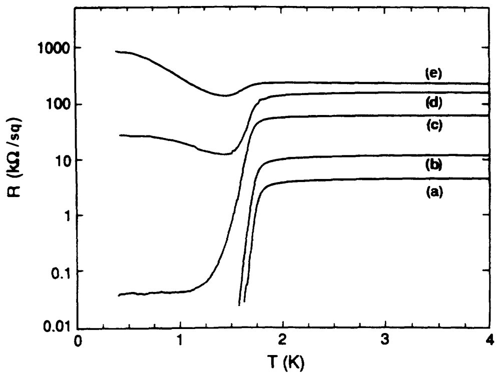

FIG. 1. Resistance versus temperature for several Al films of varying R_N in zero magnetic field. Curves are labeled from (a) to (e) with increasing R_N

transport properties of the films are dominated by the nature of the intergrain coupling. In low resistance films, $R_N < R_Q$, it is believed that global superconductivity is established via intergrain Josephson tunneling. However, as R_N is increased above R_Q, the Josephson coupling energy $E_J = \Delta(R_Q/2R_N)$ becomes small compared with the characteristic intergrain dephasing energy, e.g., the charging energy E_c, and global superconductivity is lost [2]. This is reflected in the finite resistance tail in curves (c) of Fig. 1. Finally, at high enough sheet resistance, $R_N > 100$ kΩ/sq, 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 super-resistive behavior [2,11] as can be seen in curve (e) of Fig. 1. The rounding at low temperatures, seen in curve (c), has also been observed by other groups [2]. However, if we look very closely, the resistance tail for curve (c) still has a positive dR/dT at the lowest temperatures. We have tried commercial rf filters with all the electronics running on dc batteries, but the rounding was not affected.

In order to understand to what extent grain charging determines the observed behavior, we have studied, for the first time, the normal state behavior in a high parallel magnetic field well below T_c. To that end, we will first show that the normal state can be reached via a first-order transition in $H_{\parallel}$. The upper parallel critical field due to the orbital motion of electrons in a thin film varies with film thickness t as $t^{-3/2}$ and the transition is second order [12]. As t is reduced, the orbital critical field can become so large that the Zeeman splitting of the Cooper pairs will be the dominant pair breaking mechanism. If the spin-orbital scattering rate is small, a first-order transition will occur at low temperatures when the Zeeman splitting is comparable to Δ. This is the spin-paramagnetic limit [13] with a critical field $H_{c\parallel} \sim (18.6\ \mathrm{kG/K})T_c$. Previous studies of the superconducting fluctuations in Al films down to 0.4 K suggested that the spin-paramagnetic transition became first order [14] near 0.6 K. However, no significant hysteresis was ever found. We show in the inset of Fig. 2 a giant hysteresis loop measured in one of our films during a 10 G/s field sweep at 80 mK. The temperature evolution of the hysteretic region is shown in Fig. 2 by plotting the critical fields defined at the midpoint of the transitions in field-up and field-down sweeps. Though not a true thermodynamic measurement, Fig. 2 suggests a tricritical point, $T_{\mathrm{tri}} \sim 250$ mK. With increasing R_N, the width of the hysteresis loop did not vary much for $R_N > 1$ kΩ/sq, and the temperature at which the hysteresis appeared was not changed. Details of the origin of the giant hysteresis and the unusual dynamics observed in the hysteric region will be discussed elsewhere [15].

Figure 2 provides convincing evidence that a true normal state is reached via a first-order transition for parallel field $H_{\parallel} \sim 50$ kG and $T < 250$ mK. In searching for the evolution of the charging energy with increasing R_N, we have measured ac I-V's as function of dc bias voltage V_b in the high-field normal state. Though films with $R_N \leq R_Q$ were found to be only weakly nonohmic, higher resistance films displayed significant zero bias anomalies. In Figs. 3(a) and 3(b) we plot ac conductance dI/dV versus V_b in $H_{\parallel} = 70$ kG for a 60 kΩ/sq film and a 180 kΩ/sq film at several temperatures, respectively. Since bulk N-I-N tunneling has a linear I-V [16], we believe that the anomalies observed in the normal state of our films are due to grain charging. Similar zero-bias anomalies were reported in ac I-V studies of small Sn particles imbedded in an oxide layer and were associated with grain charging [17]. Notice that the percentage of change of the zero-bias anomaly is same for the two films in Fig. 3. However, the difference in temperature scales indicates that the charac-

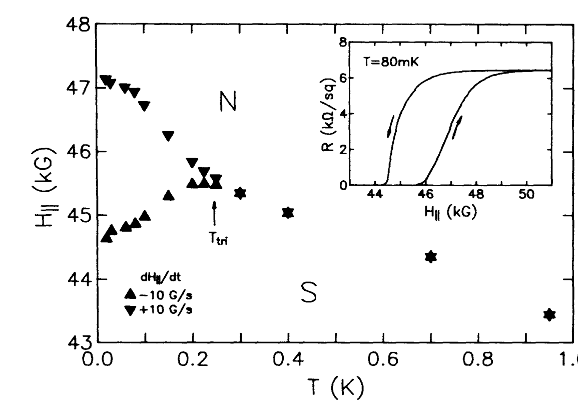

FIG. 2. Parallel critical fields versus temperature measured from field sweeps at fixed temperatures for a film with $R_N = 6$ kΩ/sq. $dH_{\parallel}/dt$ is the field sweep rate. S: superconducting state; N: normal state. Inset: a typical hysteresis loop. Hysteresis was not seen in a perpendicular magnetic field.

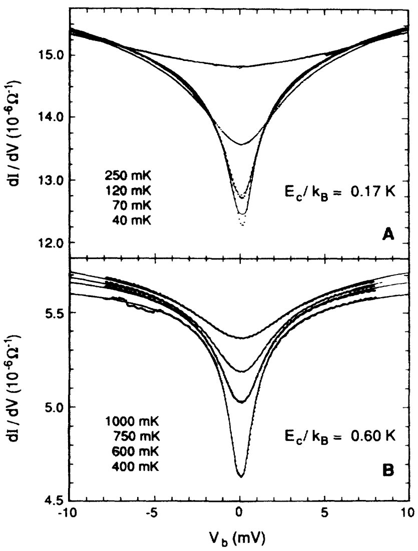

FIG. 3. ac I-V's in the normal state for a 60 kΩ/sq film (a) and a 180 kΩ/sq film (b) at several temperatures. Temperature labels from top to bottom correspond to anomalies from top to bottom. Solid lines are least-squares fits by Eq. (2).

teristic charging in energy associated with the anomaly in the 180 kΩ/sq film is much larger than that of the 60 kΩ/sq film. Correspondingly, the zero-field residual resistance of the 180 kΩ/sq film was of order 10 kΩ/sq, whereas the residual resistance of the 60 kΩ/sq film was of order 1 Ω/sq.

For a more quantitative understanding of the I-V anomalies, we have studied a simple model based on the fact that once an electron moves from a neutral gain to another nearby neutral grain, it effectively creates a charge-anticharge pair [18]. We assume that transport is mediated via ionization of such charge-anticharge pairs, which can occur by either thermal activation or electric field induced tunneling [19]. To estimate these contributions we consider the potential of a 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, $\kappa = \varepsilon[1 + d/2s]$ is the effective dielectric constant for the system [19] with d, s, and ε being the size of the grains, the spacing of neighboring grains, and the dielectric constant for Al_2O_3, respectively. The first term in Eq. (1) is the charging energy that must be overcome in order to create one nearest-neighboring charge-anticharge pair, the second term is the Coulomb energy between the pair members (screening effects are neglected), and the last term is due to the applied electric field $E = V/L$ with V the applied voltage and L the sample length. This potential has a maximum at $r_c = \gamma/E^{1/2}$ with the barrier height $U(r_c) = E_c - 2e\gamma E^{1/2}$, where $\gamma = (e/4\pi\varepsilon_0\kappa)^{1/2}$. Thermal activation over this barrier leads to a charge carrier density $\sim \exp[-U(r_c)/k_BT] = \exp[-(E_c - \alpha V^{1/2})/k_BT]$, where k_B is the Boltzmann constant and $\alpha/k_B = 2e\gamma/k_BL^{1/2} \sim 1$ K$/\sqrt{\text{V}}$. The other source of pair ionization is the field induced tunneling through the barrier. If E is small, it is easy to show that the width of the barrier ; $r_w \propto E^{-1}$. The tunneling probability through a barrier of height $U(r_c)$ and width r_w is proportional to $\exp[-r_w\sqrt{(2m/\hbar^2)U(r_c)}] \sim \exp(-\beta/V)$, where m is the electron mass and β is a constant. Combining these two ionization processes, this model predicts a conductivity of the form

$$\sigma(T,V) = \sigma_B + \sigma_a \exp\left[-\frac{E_c - \alpha V^{1/2}}{k_BT}\right] + \sigma_t \exp\left(-\frac{\beta}{V}\right), \qquad (2)$$

where 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.

To compare this model with data in Fig. 3, we first fit the temperature dependence of the ac conductance at zero bias to an activated form $\sigma(T,0) = \sigma_B + \sigma_a \exp(-E_c/k_BT)$ in order to determine σ_B, σ_a, and E_c. Taking $\alpha/k_B = 1$ K$/\sqrt{\text{V}}$ and leaving σ_t and β to be determined, Eq. (2) gives excellent fits to the ac I-V's at all temperatures, as shown by the solid line in Fig. 3. This model, though naive, captures the essential physics of the normal state of the films. The characteristic charging energy E_c/k_B was found to increase from 0.17 K for the 60 kΩ/sq film to 0.6 K for the 180 kΩ/sq film. For sample (e) in Fig. 1, E_c/k_B was about 1.4 K, which is of the order of Δ/k_B. These values are close to the expected value [19] of $E_c = e^2/4\pi\varepsilon_0\kappa d \sim 1$ K with $\varepsilon \sim 10$, $d \sim 30$ nm, and $s \sim 1$ nm.

Comparison of E_c obtained in the fits to the data in Fig. 1 strongly suggests that when E_c is of order Δ a granular film will display quasireentrance and super-resistive behavior. This conclusion is supported by the following simple argument considering two nearby superconducting grains. The energy cost for a quasiparticle to tunnel from one grain to the other is $2\Delta + E_c$, while a Cooper pair tunneling cost $4E_c$. Quasiparticle tunneling will become favorable if $4E_c > 2\Delta + E_c$, or if E_c is of the order of Δ. Haviland *et al.* [20] have also found in a number of quench-condensed metal films that the characteristic activation energies of conduction above T_c, as determined by hopping models, were close to Δ when quasireentrance and super-resistive behavior appeared. Most recently, this condition was also found to be relevant to the physics in single electron tunnel junctions [21].

In zero field, the condition $E_c \sim \Delta$ is reached by increasing E_c via R_N. A natural test of the above discussion is to lower Δ in a low-resistance film by applying $H_\parallel$. Films of $R_N \sim 10$ kΩ/sq have a small characteristic E_c and quasireentrance and super-resistive behavior should appear. This is indeed observed in Fig. 4(a) where we

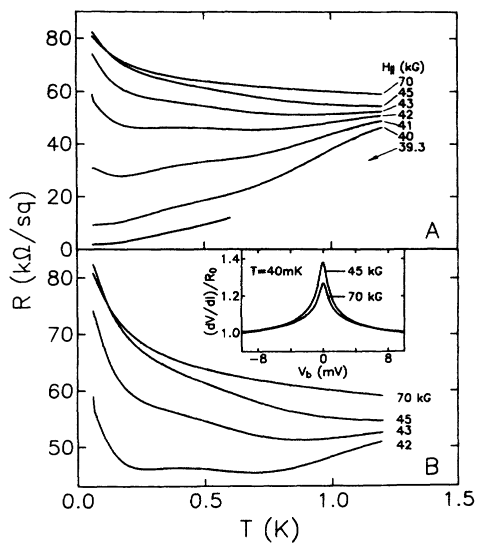

FIG. 4. (a) Resistances versus temperature at several $H_\parallel$ close to $H_{c\parallel}$. (b) Data from (a) plotted on a different scale to show the super-resistive behavior at low T. Inset: ac I-V's scaled by $R_0 = dV/dI$ at $V_b = 10$ mV, showing the suppression of the gap with increasing $H_\parallel$.

plot the temperature dependence of resistances at several $H_\parallel$ for a film with $R_N \sim 60$ kΩ/sq. For $H_\parallel > 50$ kG, the background insulating behavior no longer changed with $H_\parallel$. Although seen on somewhat different temperature scale, the $H_\parallel \sim 42$ kG data in Fig. 4(a) exhibits the character similar to those seen in curve (e) of Fig. 1. In particular, the transition is quasireentrance to a super-resistive phase below 250 mK. It is evident that a finite gap $\Delta(H_\parallel)$ exists [14] even at 45 kG. Since the gap is not completely suppressed at this field, the $H_\parallel = 45$ kG curve actually crosses the normal state curve of $H_\parallel = 70$ kG, see Fig. 4(b). In the inset to 4(b), we plot the ac I-V's for $H_\parallel = 45$ kG and 70 kG at 40 mK. The decrease in the magnitude of the anomaly with increasing field is due to the suppression of Δ. The fact that the magnitudes of the anomalies due to charging (in the 70 kG curve) and due to Δ are of the same order implies that E_c is of the order of Δ. It is very interesting that, below 250 mK, films with R_N ranging from a few kΩ/sq to 100 kΩ/sq displayed a thresholdlike sheet resistance very close to R_Q that separated curves of positive temperature coefficient from those of negative temperature coefficient, see the $H_\parallel = 40$ kG curve in Fig. 4.

In summary, anodized Al films appear to be a compelling alternative to quench condensed granular systems for studying the interplay between grain charging and Josephson tunneling in the S-I transition. We observed a unique first-order S-I transition in $H_\parallel$ below 250 mK. In the vicinity of this transition, the important characteristics of the zero-field transition, including quasireentrance and a super-resistive phase, were reproduced. We conclude that Coulomb blockade is the dominant factor in both the zero field and finite field super-resistive phases.

We thank Dana Browne, Steve Girvin, Heinrich Jaeger, Robert Meservey, Paul Tedrow, and especially James Valles for valuable discussions. We are grateful to Robin McCarley for the scanning force microscopy characterization of our films. This work was supported by NSF Grant No. DMR 9258271 and Grant No. 9204206.

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