Evidence of Collective Charge Behavior in the Insulating State of Ultrathin Films of Superconducting Metals

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Nonlinear I-V characteristics have been observed in insulating quench-condensed films which are locally superconducting. We suggest an interpretation in terms of the enhancement of conduction by the depinning of a Cooper pair charge density wave, Cooper pair crystal, or Cooper pair glass that may characterize this insulating regime. We propose that this is a more likely description than the Coulomb blockade or charge-anticharge unbinding phenomena.

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In the context of the Bose-Hubbard model, which is equivalent to the model of a Josephson junction array with charging, zero-temperature superconductor-insulator transitions in two dimensions (2D), tuned by disorder or magnetic field, are believed to be direct, with metallic behavior only at the quantum critical point [1]. Recent experiments have suggested the existence of a significant metallic phase between the superconductor and insulator [2]. The Stanford group reported this for MoGe films in moderate magnetic fields, and conjectured that it was due to dissipation [3]. They later found "true" superconductivity in low fields [4]. Long ago, metallic behavior was reported in quench-condensed granular films over a range of thicknesses intermediate between those for which films were insulating and superconducting [5], and it was found more recently in Josephson junction arrays [6]. Reexamination of the theory has involved the inclusion of aspects of percolation [7], elaboration of the Bose-Hubbard model [8,9], and consideration of dissipative Bose systems [10]. Das and Doniach [8] explained aspects of Ref. [4] by extending the Bose-Hubbard model to high filling and including nearest-neighbor as well as on-site Coulomb interactions. Their phase diagram contains the possibility of an intervening Bose metal phase, or a direct transition, depending upon the relative magnitudes of the various Coulomb and Josephson coupling energies. The Bose metal is prevented from Bose condensing by dissipation. The insulator is a condensate of vortices, or in other language, a Cooper pair charge density wave (CDW). Dalidovich and Phillips [9], using the original form of the Bose-Hubbard model, also demonstrated that there was a Bose metallic phase, with the insulator resulting from a dissipative process such as coupling to a heat bath or from a high enough level of disorder. Ng and Lee [10] asserted that a Bose metal did not exist at zero temperature, but noted a crossover regime at finite temperature that could be identified as a metallic phase.

In this Letter we shift the focus to the study of the insulating regime of granular or clustered quench-condensed films. In the past the insulator was not studied, as the issue was the onset of superconductivity. Since tunneling has been used to demonstrate that there is a nonvanishing order parameter amplitude, in such insulating films [11], they should be good candidates for the application of the Bose-Hubbard model. We have studied Ga films, which earlier exhibited the most striking metallic regime [5]. We propose that the nonlinear I-V characteristics found in the insulating regime may be evidence of a Bose insulating state, which could be either a Cooper pair CDW or Cooper pair crystal. For sufficient disorder it would be a Cooper pair glass. The nonlinearities appear to be similar to those associated with the depinning of charge density waves [12]. The behavior of these clustered insulating films is different from that of nominally homogeneous films, where the observation of an orbital magnetoresistance linear in field was interpreted as evidence of vortices [13].

Gallium films were grown on glazed alumina substrates held at liquid helium temperatures. This was done using an ultrahigh vacuum apparatus which permitted cycles of film evaporation at 12 K and *in situ* transport measurements [14] down to 150 mK. Thicknesses are nominal, being derived from a calibrated thickness monitor. All electrical leads entering the vacuum chamber for four-probe measurements were filtered with low-pass interference filters in series with 20-nH inductors. This combination exhibited voltage attenuations of 15 dB at 1 MHz, increasing to 75 dB beyond 100 MHz. Magnetic fields could be applied up to 12 kG perpendicular and 20 kG parallel to the plane of the sample. This apparatus is unique in that the evaporants are derived from commercial Knudsen cells, and the geometry of the cells and the source-to-substrate distance (60 cm) is such that the flux density at the substrate is uniform to better that one part in 10^4. This permits the observation of the dramatic effect of very tiny changes in thickness, which might be averaged over in film growth systems with less uniformity of flux density.

To set the stage, we show in Fig. 1 the evolution of $R(T)$ with thickness for a series of a-Ga films, adapted from Ref. [5]. In the thinnest films $R(T)$ is a monotonically increasing function of decreasing temperature. In thicker films, R drops near the bulk T_c of a-Ga, providing evidence

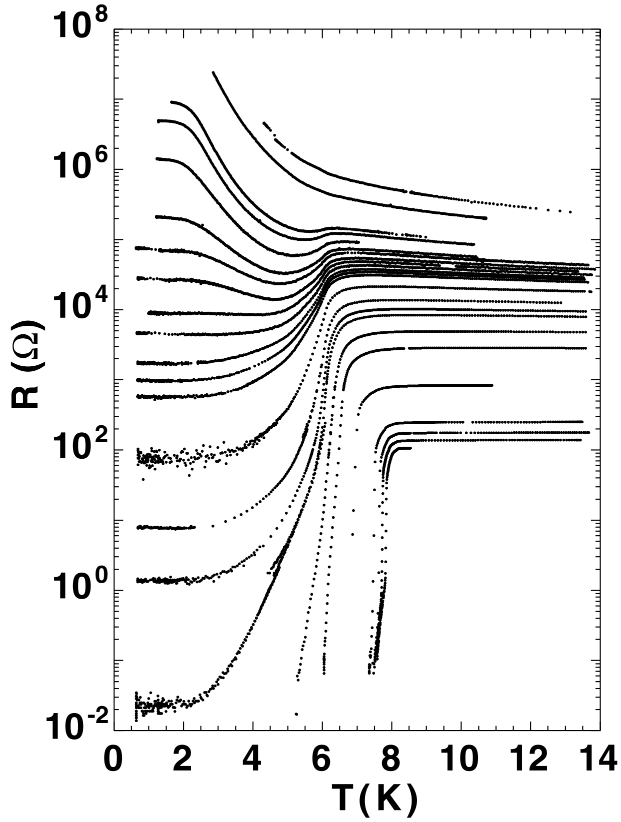

FIG. 1. Evolution of $R(T)$ with thickness for a series of Ga film, adapted from Ref. [4]. Film thicknesses range from 12.75 to 26.67 Å and increase from top to bottom. Thickness was changed in increments of 0.05 Å. Some data have been omitted for clarity. For this sequence the limiting temperature was 0.5 K.

for local superconductivity. At lower temperatures it increases exponentially with decreasing T, until about 2 K, where it levels off. Further increase in thickness results in metallic behavior at low temperatures. For somewhat thicker films $R(T) \sim \exp(\frac{T}{T_0})$, which has been interpreted as evidence of quantum tunneling of vortices [15]. At the lowest temperatures for these films, $R(T)$ is also temperature independent. Further increase in thickness results in superconductivity. Metallic films have linear I-V characteristics, whereas all of the other films have nonlinear ones. (For the thinnest films at the lowest temperatures, the I-V characteristics are not known as long time constants precluded accurate measurements.) From the onset of local superconductivity to full superconductivity, nominal film thickness increased from 12.75 to 14.50 Å.

In discussing nonlinear I-V characteristics, we will consider a second sequence of films that were studied over a range of temperatures that extended far below that available for the first sequence. The films of this second sequence were grown with substrate temperatures slightly higher than those of the first. Although not changing the qualitative evolution of behavior with increasing thickness, this causes the onset of full superconductivity to manifest itself at a greater thickness.

The leveling off of $R(T)$ of high resistance films was current dependent. The lower the current, the higher the value of $R(T)$ that would be attained with decreasing temperature. This is shown in Fig. 2, which displays representative data from a film in the second sequence. The differential conductance, $G(V)$, also shown in Fig. 2, exhibits a threshold voltage, V_T, which is independent of temperature, and which decreases with increasing thickness, both of which are shown in Fig. 3.

Heating cannot account for the observed effects for several reasons. Considering the film to be a thermometer, one can define an "effective temperature," $T_{\rm eff}$, based on $R(T)$. For a range of dissipated power spanning 3 orders of magnitude, from 10^{-13} to 10^{-10} W, $T_{\rm eff}$ is well above the thermometer temperature. However, it did not increase with applied power, and in some cases even decreased, implying that the changes in $R(T)$ are not due to heating and that the film is not acting as a thermometer. Furthermore, the power at which the resistance leveled off as films became thicker decreased even when the fractional change in thickness was small. Actually, the power needed to heat to a particular temperature should increase rather than decrease with increasing film thickness, as both heat capacity and thermal conductance become larger. All of this implies that the nonlinearities are intrinsic. In addition, much higher resistances were measured in the same apparatus

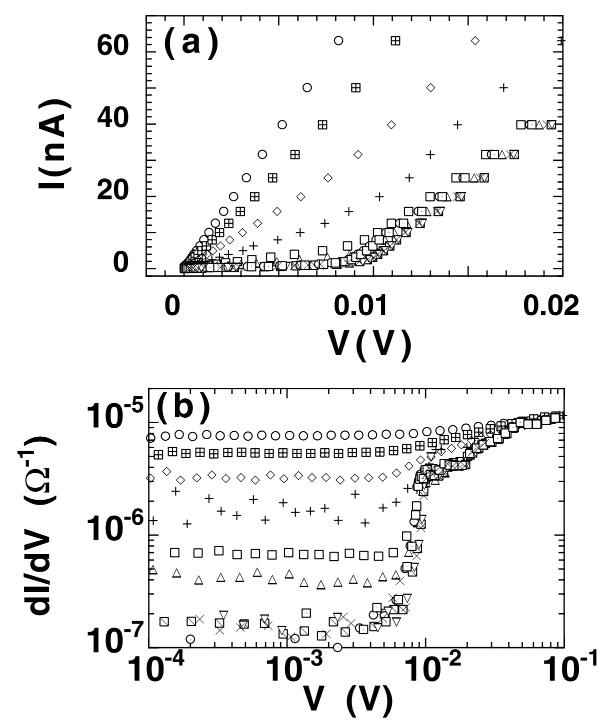

FIG. 2. (a) The evolution of the I-V characteristics with temperature in a 18.07 Å thick Ga film from a sequence different from that shown in Fig. 1. These data are representative of what is found in a number of insulating films. The temperatures are 8, 7, 6, 5, 4, 3, 2, 1.5, 1, 0.5, and 0.15 K from top to bottom. (b) dI/dV vs V at the same temperatures for the same film. A temperature independent threshold voltage of about 6 mV can be seen where the conductance suddenly increases.

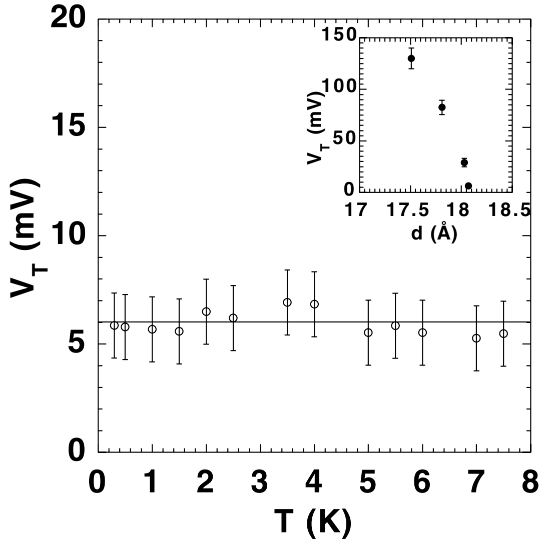

FIG. 3. Threshold voltage as a function of temperature of an 18.07 Å thick Ga film. Inset: Threshold voltage as a function of thickness for four Ga films in a series.

at lower temperatures for other films not exhibiting either superconductivity or flattening [16]. Finally, the temperatures at which $R(T)$ became temperature independent were well above 1 K, making a scenario of not cooling the electrons unlikely [17].

Quasiparticle tunneling involving superconductor-insulator-superconductor junctions in the film [5] can be ruled out because V_T is temperature independent as shown in Fig. 3. If V_T were a signature of the superconducting gap, it would increase with decreasing temperature. Not shown is the magnetic field *independence* of the threshold, which is also consistent with it not being attributable to quasiparticle tunneling. However, the subthreshold conductivity does increase with increasing temperature and magnetic field suggesting the existence of quasiparticle transport channels.

A phenomenological picture of charge unbinding has been used to explain nonlinear I-V characteristics in very much thicker granular Al films, driven insulating by a magnetic field [18]. This model, which includes five free parameters that cannot be independently measured, can be fit to our data. The fit, which is not shown, does not reproduce the sharpness of the conductance increase at V_T.

In Josephson junction arrays with ultrasmall junctions [19] and Pd films [20], nonlinear behavior at high resistances has been attributed to effects related to the charge Kosterlitz-Thouless-Berezinskii (KTB) transition. In this picture, nonlinear I-V characteristics would be expected, with $I \propto V^a$, and a exhibiting a jump from 1 to 3 at the transition temperature. There should also be a square root cusp in $R(T)$ above the KTB transition temperature. These effects were not seen. This model is most likely irrelevant because the screening length in these films is too short for the interaction to be logarithmic over a substantial distance, a requirement for this scenario [21].

Nonlinearities in the I-V characteristics could be the signature of a Coulomb blockade [22]. The fact that the high-voltage current asymptote of the I-V characteristic, at low temperatures, extrapolates back through the threshold voltage V_T, rather than through the origin, is consistent with this picture. The macroscopic I-V characteristics of a film would then be dominated by those associated with tunneling in and out of a single "representative island," according to the arguments of Ambegaokar, Halperin, and Langer [23]. The threshold voltage would be given by the energy necessary to overcome the Coulomb energy E_C in adding charge to this island, as $V_T = E_C/e = e/2C$, where C is the capacitance of the island. The central argument against this picture is that nonlinearities are not seen until the temperature corresponding to the onset of local superconductivity. Values of V_T ranging from 6 to 130 mV in films of different thicknesses, correspond to Coulomb energies much greater than k_BT at 8.3 K, where the threshold appears.

We now turn to a picture of the insulating state in which it is a CDW-like configuration of Cooper pairs or a two-dimensional Cooper pair crystal. This idea is mentioned in theoretical papers, but is never treated in detail [1,8–10]. We suggest that the nonlinearities may be associated with the depinning of this putative CDW, crystal, or glass state. Features of the depinning that qualitatively resemble the data are the sharply enhanced conductance at V_T and the fact that at low temperatures, all the I-V curves come together at high bias with the asymptotic limit of the current extrapolating back to V_T [12].

Atomic force microscope (AFM) pictures of the film sequence discussed in detail here, which are not shown, were obtained at a thickness of 18.24 Å, and after the film was warmed up and removed from vacuum. These pictures showed clusters of 200 Å diameter, which should be representative of the structure at low temperatures. Similar studies of a thinner film from a different series, revealed smaller clusters, suggesting a correlation between cluster size and thickness. These results were very similar to *in situ* scanning tunneling microscopy pictures obtained for quench-deposited Pb films [24]. Although on warming, annealing of the microstructure takes place, the mesoscale structure should be largely unchanged, because room temperature, or 20 °C, is not high enough to result in atomic diffusion over distances sufficient to change the mesoscale structure. Clusters form during growth, with the kinetic energy of the impinging atoms making them mobile on the substrate surface, even though it is attached to a holder cooled to helium temperature.

It may be possible to consider the film structure of a random array of nearly identical clusters to be the origin of the pinning of the Cooper pair CDW or crystal. The decrease of the threshold voltage with film thickness, shown in the inset of Fig. 3, would then result from a reduction of the areal density of pinning centers with increasing thickness and physical cluster size. Scaling of the I-V characteristic near V_T, which is found in many CDW systems could not be tested. This was a consequence of our measurements being current biased. At V_T the voltage increased very rapidly, limiting the data in a manner that made a definitive test of scaling impossible. What is most important is that the nonlinear behavior manifests itself only when a local minimum in $R(T)$ is present. Thus the phenomenon must be associated with Cooper pairing.

An applied magnetic field of up to 12 kG perpendicular and 20 kG parallel to the film left the threshold in $G(V)$ unchanged, increased $G(V)$ for $V < V_T$, and lowered the temperature at which the local minimum in $R(T)$ was found, by up to 1 K. Neither maximum value of field was high enough to completely obliterate the threshold. The fact that V_T is field independent suggests that the pinning mechanism is associated with phenomena unaffected by field, i.e., defects. The effect of a parallel field on the subgap conductance is smaller than that of a perpendicular field, but is of the same order of magnitude. This suggests that there could be some role for spin degrees of freedom. The subthreshold conductance, as mentioned, most likely comes from quasiparticles. The application of a magnetic field would increase the number of such quasiparticles participating in transport.

The picture we have presented here as well as the Coulomb blockade and charge-unbinding effects all involve Coulomb interactions. The difference between charge-structure depinning and these other phenomena is whether the behavior is collective, or whether charges individually overcome a Coulomb energy. Further support for collective behavior is the similarity and even electrical duality of $G(V)$ characteristics of "high" resistance films with $R(I)$ characteristics of low resistance films. In the latter, the current threshold is a consequence of the depinning of vortices [25]. The saturation of $G(V)$ and $R(I)$ at low temperatures would imply that both types of depinning may be quantum phenomena. If collective charge behavior indeed governs the insulating phase of these 2D superconducting systems, then it is a member of a larger class of problems including vortex lattices, charge density waves, Wigner crystals, and magnetic bubbles, in which it is necessary to understand the dynamics of a periodic structure in the presence of static substrate disorder. It would then be appropriate to bring to the problem the ideas used to study such solids [26].

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