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Evolution towards superconductivity in granular films of bismuth

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The thickness-tuned two-dimensional insulator-superconductor transition has been investigated in ultrathin, granular films of amorphous bismuth. "Double reentrance" was observed in the resistance vs temperature. We suggest that the reentrance into the insulating state results from the opening of the energy gap on superconducting grains or clusters coupled by quasiparticle tunneling, and that the reentrance into the superconducting state is a consequence of the onset of intercluster Josephson coupling leading to global phase coherence. Measurements of voltage fluctuations have also been carried out. For insulators, the first power spectra exhibit $1/f^2$ frequency dependence and the second spectra are white, which is consistent with uncorrelated fluctuations. The first power spectra gradually change from having a $1/f^2$ frequency dependence in the insulating state to being independent of frequency in the superconducting state. There is no specific feature of the noise that can be associated with the onset of superconductivity

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The sequence of ground states in two-dimensional "superconductor-insulator" (SI) transitions is an open issue that has received much attention recently. A model by Fisher described a transition between superconducting and insulating ground states with a resistive "metallic" state separating them precisely at the critical value of the tuning parameter.[1] Various groups have reported such "direct" transitions. However, there have been reports of intermediate metallic regimes that span extended ranges of tuning parameters in granular films,[2,3] Josephson junction arrays,[4] and nominally homogeneous films.[5,6] Various theoretical works have attempted to explain these regimes.[7–10]

While these regimes appear to be intrinsic, there is always a possibility that temperature-independent resistances observed at low temperatures are the consequence of a failure to adequately cool the electrons of the film or array as effectively as the thermometer. We have studied "hot electron effects" in homogeneous films of amorphous bismuth (*a*-Bi) and were able to attribute temperature-independent resistances below 100 mK in these films as being a consequence of the failure to cool the electrons due to weakening of the electron-phonon coupling with decreasing temperature.[11] While the flattening of resistance that occurs in *a*-Bi homogeneous films can be attributed to experimental artifacts, we find the temperature independence of resistances in granular films that occur as high as 2–4 K to be more difficult to attribute to these effects, as the electron-phonon coupling is robust at these higher temperatures.

Here, we report measurements of the thickness-tuned insulator-superconductor transition of ultrathin granular films carried out to lower temperatures than in previous works and accompanied by low-frequency noise measurements that in principle might serve to distinguish between different regimes. Instead of a metallic regime, or even a direct transition, double reentrance in $R(T)$ was observed over a small range of thicknesses around the nominal critical thickness. The reentrance to the insulating phase may result from the opening of the energy gap on superconducting grains or clusters coupled by quasiparticle tunneling, with the reentrance into the superconducting state being a consequence of the onset of Josephson coupling and global phase coherence. In the most insulating films, an analysis of voltage fluctuations revealed a $1/f^2$ frequency dependence of the first power spectra. "Second spectra," which are almost independent of frequency, imply that the fluctuators are uncorrelated. The noise gradually changed from a $1/f^2$ spectral density to white as the phase coherence became global.

Films were grown on epipolished (100) $SrTiO_3$ substrates cooled to 8 K in a UHV chamber, and measured in an attached dilution refrigerator.[12] Depositions were alternated with measurements without removing the films from UHV or warming them above 10 K. The four-wire measurement geometry was a square of area $(0.5\ \text{mm})^2$. The dilution refrigerator was shielded with 60 Hz *RC* filters and kHz π-section filters at 300 K and with GHz thermocoax filters[13] at the mixing chamber.

There have been recent studies of $R(T)$ vs thickness of quench-condensed Bi films grown on similarly bare substrates. The results have been interpreted as evidence of an amorphous to granular transition at an early growth stage,[14] and of the presence of mesoscale thickness inhomogeneities usually interpreted as evidence of granularity.[15] There have also been *in situ* STM studies of quench-condensed Au and Pb films from which similar conclusions have been drawn.[16] Because of differences in growth conditions specific to each of these investigations, which are all different from ours, it is not possible to make generalizations about morphology from these works. The films of the present work are all in the granular regime. The results are from a particular sequence of *a*-Bi films. The same features were observed in one other sequence, not investigated in detail. These features were not found in five sequences of films grown under *identical* conditions exhibiting regular transitions, only differing in that their substrates were pre-coated with a monolayer of amorphous Sb, which is believed to yield homogeneous rather than granular films.[17]

In Fig. 1, the temperature dependence of the sheet resistance, $R(T)$, is shown for film thicknesses between 21.11 and 27.56 Å. In the two thinnest films, strong insulating behavior was observed. In slightly thicker films, local minima in $R(T)$

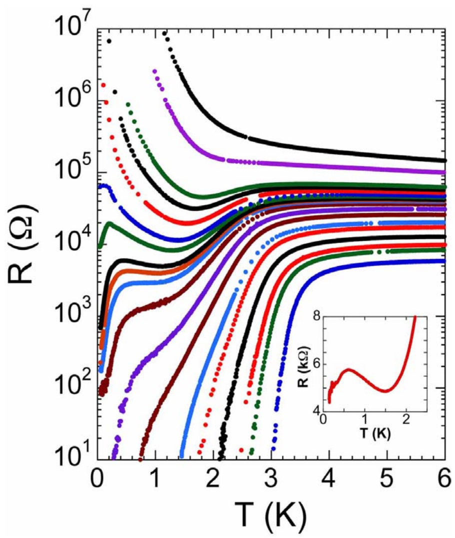

FIG. 1. (Color online) Sheet resistance vs temperature for the thickness-tuned superconductor-insulator transition. The thicknesses are 21.11 (top), 21.41, 21.75, 21.83, 21.94, 22.04, 22.10, 22.14, 22.17, 22.23, 22.38, 22.63, 22.95, 23.37, 23.66, 24.52, 25.40 26.07, and 27.56 Å (bottom). Glassy effects were found for films with $R > 10^7\ \Omega$. In this glassy regime R was obtained from the linear regime of *I-V* characteristics, after waiting hours to achieve equilibrium. Inset: Data from a separate sequence of a film with a nominal thickness of 25.10 Å.

are apparent. This “reentrant insulating behavior” is believed to be caused by two effects: (i) a drop in resistance as superconductivity develops on isolated grains, and (ii) an increase in resistance at lower temperatures as the energy gap opens in the grains. In the thickest films, global superconductivity occurs, presumably because the superconducting clusters become Josephson coupled.

In films having thicknesses between 22.04 and 22.63 Å, a regime of “double reentrance” is observed in $R(T)$. In this regime, the insulating behavior that starts below ~1.5 K is overcome by a reentrance to superconductivity at temperatures below ~500 mK. The reentrance to superconductivity occurred at ~100 mK in the 22.0 Å thick film and at progressively higher temperatures as the thickness is increased. When superconductivity is robust with $T_c \sim 1.5$ K, there is no kink in R vs T.

Double reentrance in a SI transition is unusual. As discussed earlier, most SI transitions are direct, or exhibit an intermediate metallic regime. The failure to observe double reentrance in granular films in previous studies results from measurements not being carried out to sufficiently low temperatures. In addition, the range of thickness for which the effect is found is tiny and would be missed if thickness increments were too large. Although our observations of double reentrance are the most extensive to date, Hadacek *et al.* observed a weak version of the effect near the critical magnetic field in some sputtered TiN and NbN microcrystalline superconducting films,[18] and Van der Zant *et al.* found a weak effect in Al Josephson junction arrays whose geometry set the ratio of the charging energy of a grain to the Josephson coupling energy, E_C/E_J, closest to the critical value, but

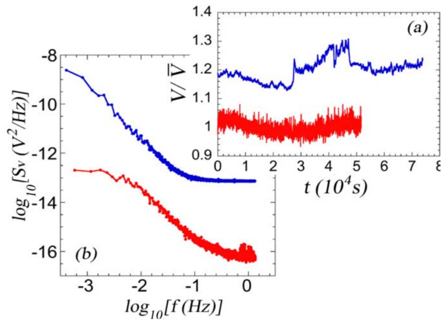

FIG. 2. (Color online) (a) Normalized V vs t for the 21.41 Å thick film at 500 mK (top) and 1 K (bottom). The 500 mK data are offset. (b) The first power spectrum for the data in (a), at 500 mK (top) and 1 K (bottom). $1/f^2$ frequency dependence below ~100 mHz and white noise above ~100 mHz are observed.

not in arrays with larger or smaller values of this ratio.[4] Although there are no detailed theories for granular films, Dalidovich and Phillips have predicted that near the critical point of Josephson junction arrays that dissipation can produce a metallic regime at intermediate temperatures, which suddenly changes to the superconducting state as $T \rightarrow 0$.[19] Junction arrays have long been used to model the behavior of granular films.

In addition to $R(T)$ measurements, noise spectral densities were studied, with the expectation that they would yield information about the various regimes. For each thickness, the capacitance C was determined at 1 K, the *I-V* characteristics and resistance were measured at low temperature, and dc currents at the pA or nA level in the linear regime were applied for a time greater than 5 RC at which point transients disappeared. Voltage was then sampled at 2.5 Hz with a Keithley nanovoltmeter for times between 8 and 48 h.

Fluctuation measurements revealed $1/f^2$ frequency dependent first power spectra for strongly insulating films. In Fig. 2(a), normalized voltage V vs time t is shown for the 21.41 Å thick film at 500 mK and 1 K. The voltage was observed to meander nonmonotonically in time. The resistance was $2 \times 10^8\ \Omega$ at 500 mK and 240 kΩ at 1 K. In Fig. 2(b), the Fourier transform of $V^2(t)$, i.e., the first power spectrum, is shown for both temperatures. Here, a $1/f^2$ frequency dependence was found between 0.5 and 65 mHz at 500 mK and between 10 and 130 mHz at 1 K. Above 65 and 130 mHz, respectively, the spectra were white, which presumably resulted from thermal noise and shot noise.

This $1/f^2$ dependence only extended down to a knee frequency f_k below which the dependence was typically $1/f$ or weaker. In Fig. 2(b), f_k was ~10 mHz at 1 K and ~1 mHz at 500 mK. This frequency decreased as the resistance increased, either due to decreasing temperature or increasing disorder. The temperature dependence was observed in the 21.41 Å thick film at 500 mK, 750 mK, and 1 K, and the thickness dependence was observed in the 20.90, 21.11, and 21.41 Å thick films at 1 K.

The second power spectra are almost independent of frequency for insulating films that have $1/f^2$ first power spectra, which implies that the fluctuators are uncorrelated.[20] The most extensive data were for the 20.90 Å thick film at

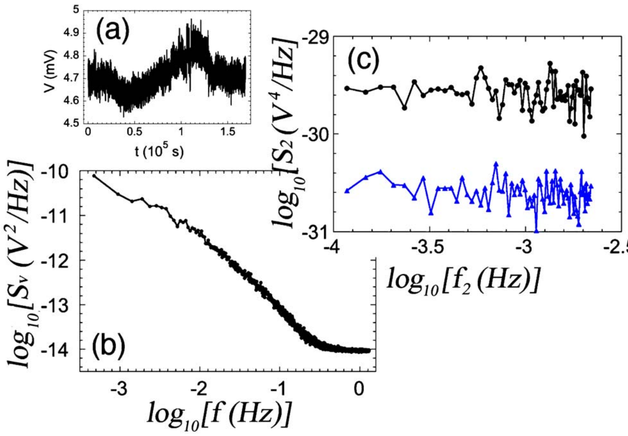

FIG. 3. (Color online) (a) $V(t)$ for the 20.90 Å thick film at 100 mK. (b) First power spectrum for this film. (c) Almost frequency-dependent second power spectra at 50 mHz (top) and 100 mHz (bottom).

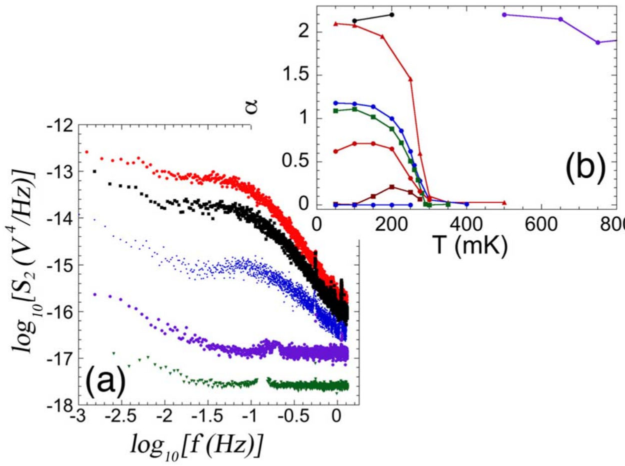

FIG. 4. (Color online) (a) The first power spectrum for the 21.94 Å thick film at 100 (top), 175, 250, 350, and 500 mK. (b) Slope of the first power spectrum as a function of temperature for the 21.41 (top right), 21.83, 21.94, 22.04, 22.10, 22.17, 22.38, and 22.95 Å (bottom) thick films.

100 mK, which is discussed here. However, shorter $V(t)$ traces at other temperatures and at different thicknesses show the same qualitative results. In Fig. 3(a), $V(t)$ is shown. In Fig. 3(b), the first power spectrum is shown, which is $1/f^2$ over a wide range. In this case, the second power spectrum was obtained by dividing V vs t into 150 equal-length segments, calculating the first power spectrum $V^2(f)$ for each segment, integrating the power P, from f to $2f$ for a given frequency, and taking the Fourier transform of this $P(t)$ trace. If fluctuations are uncorrelated, then the fluctuations in P should be independent of frequency. In Fig. 3(c), we show the second power spectra for this film at 50 and 100 mHz, which are almost independent of frequency.

In strongly insulating films, $1/f^2$ behavior was found at all temperatures for which measurements were made, but in less resistive films, the first power spectra were dependent on temperature. For instance, in the 21.94 Å thick film, $1/f^2$ noise was also observed below 175 mK, white noise was found above 300 mK, and a fairly abrupt transition occurred at 300 mK. In Fig. 4(a), these power spectra are shown at several temperatures.

In the regime of double reentrance, as insulating behavior gradually gave way to superconducting behavior, the frequency dependence of the first power spectra gradually switched from $1/f^2$ to white. In Fig. 4(b), we plot the exponent of the first power spectrum α from $S_V \sim 1/f^{\alpha}$, vs T for various film thicknesses. In the superconducting state, the fluctuations are white over a wide range of frequencies. One interesting issue that arises from the data in Fig. 4(b) that we cannot explain is why the noise changes from white to nonwhite at about 300 mK over a range of film thicknesses.

The $1/f^2$ noise in the insulating state is most likely caused by trapping and release of charge carriers. In a system that has trapped charges, releases that occur at various rates will cause voltage fluctuations that have a $1/f^2$ frequency dependence.[21–23] If the rates vary between two characteristic frequencies f_A and f_B, for which $f_A < f_B$, fluctuations yield white noise below f_A, $1/f$ noise between f_A and f_B, and $1/f^2$ noise above f_B. When $f \gg f_B$, $1/f^2$ noise is smaller than white thermal and shot noise. This model, which is often used as an explanation for $1/f$ noise, could explain our data if the relevant frequencies were low, i.e., if f_k corresponded to f_B. A possible scenario would be that the average voltage is set by the charges on the connected path through the percolation network. Noise would result from charges that tunnel between this path and semi-isolated clusters. This would explain our observation that f_k is smaller in thinner films, as the average distances would be larger.

While the previous picture for $1/f^2$ noise in our insulating films is plausible, a classical two-dimensional (2D) model of interacting electrons may also explain our observed dependences on thickness and temperature.[24] In this model, electrons evolve with Langevin dynamics and are attracted to defect sites. The frequency dependence of the power spectrum switches fairly abruptly from white to $1/f^{1.4}$ as either temperature or electron density are decreased. This qualitative dependence on temperature is very similar to our results, as is the dependence on density, if our density is taken as the density of mobile carriers. There is a quantitative difference in that our noise varies as $1/f^2$ rather than $1/f^{1.4}$.

Noise with roughly $1/f^2$ dependence has been observed in the insulating phases of other two-dimensional strongly correlated electron systems, though significant differences exist between these observations and ours. Bielejec and Wu observed glassiness and slow relaxation in the normal state of superconducting granular Al films, in large magnetic fields in excess of the spin-paramagnetic limit.[25] Hysteretic behavior in R vs T, stretched-exponential relaxations, and $1/f^{1.7}$ noise were found at low temperatures and were taken as evidence of a correlated electron glass. Our results are similar to theirs in that decreasing temperature causes an increase in noise at low frequencies. However, the temperature dependence is different and they do not report a dependence on disorder. More important, our almost-white second spectra suggest that our fluctuations are uncorrelated.

The present results also have qualitative similarities to those reported for dilute Si-metal-oxide-semiconductor field-effect transistors by Popovic *et al.*[26] The first power spectra in their insulating state had $1/f^{1.8}$ frequency dependence that changed to $1/f$ by either changing density towards the metallic phase or by increasing the temperature. However, their second spectra were frequency dependent, suggesting that fluctuations in their system were correlated.

Our observation of a regime of double reentrance instead of metallic behavior raises the question of whether an intermediate metallic regime exists in granular films. In granular films, the onset temperature of metallic behavior can be as high as 2 or 4 K,[2,3] temperatures at which the electron-phonon coupling should be robust. Thus it is possible that a metallic phase simply does not occur in granular Bi due to material properties, while it does occur in other granular films. However, we cannot rule out the possibility that the earlier observations of extended metallic regimes are artifacts of the failure to cool the electrons in the film to low enough temperatures either because of limitations of refrigeration or the weakness of the electron-phonon coupling in the face of thermal loads. The present measurements were carried out in a refrigerator[12] that is filtered better than that used for previous experiments.[2] In the case of homogeneous *a*-Bi films, the removal of room temperature filters has been shown to affect the shape of resistive transitions.[11]

In conclusion, the thickness-tuned 2D superconductor-insulator transition of a granular superconductor has been investigated. A regime of double reentrance in $R(T)$ was observed near the nominal critical thickness, possibly explained by a theory related to that of Ref. 19. Measurements of voltage noise in the insulating state revealed $1/f^2$ frequency dependencies of the first power spectra and nearly frequency-independent second spectra. As the film is made thicker, these first power spectra gradually become "white" as superconducting coherence is achieved. Noise studies do not appear to be an incisive probe of the onset of superconductivity. The precise mechanism of the evolution of the spectral density from a $1/f^2$ frequency dependency to white is not known and must be considered to be a challenge to theory.

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