Two-Dimensional Phase Transition in Granular NbN Films

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The resistance of several granular NbN films has been measured as a function of temperature as they approach the superconducting state. The lowest-resistance data deviate from earlier reported power-law behavior above a phase-ordering transition T_{cj}, but are consistent with the exponential expression describing the resistance above a two-dimensional phase-transition temperature, T_{2D}, of the type proposed by Kosterlitz and Thouless. At T_{2D} a finite supercurrent appears.

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There has been considerable recent interest in two-dimensional (2D) phase transitions. Until the pioneering work of Kosterlitz and Thouless (KT)[1] and Berezinskii[2] predicting the existence of a 2D topological transition it was believed that the absence of long-range order precluded the occurrence of a phase transition in a 2D system with unbroken symmetry.

Very recently 2D topological phase transitions have been observed in superfluid helium and there is increasing evidence that a similar transition involving the condensation of vortex-antivortex pairs is present in 2D superconductors of finite size.[3] Indirect evidence for the existence of such a transition has been reported by Gubser and Wolf where current-voltage (I-V) and critical current (I_c) anomalies in granular NbN consistent with a KT transition were observed.[4] More recently, Hebard and Fiory,[5] analyzing the complex ac impedance of granular aluminum films, Voss, Knoedler, and Horn,[6] using a noise spectrum of granular Al, and Epstein, Goldman, and Kadin,[7] analyzing current-voltage characteristics of Hg-Xe films, also reported data indicative of a KT transition. In this Letter we report evidence for a 2D transition with unusually large critical parameters in granular films of NbN by comparing theoretically predicted resistances above T_{2D} with newly acquired experimental results.

The samples measured were prepared by rf reactive sputtering a 200-Å-thick NbN film and subsequently anodizing the film until the nominal metal thickness remaining was about 20–30 Å. This procedure produces a granular material consisting of 50-Å-thick NbN grains approximately 80 Å in diameter, surrounded by a niobium oxide matrix.[8] The sheet resistance of our samples could be varied over a wide range but the range for samples reported here is between 10 000 and 20 000 Ω/□ at 30 K.

Resistances were measured using a four-lead ac technique with ± 0.015-Ω sensitivity at 100 nA of sample current. This low current, $I < I_M(T)$, was required to stay in a linear voltage-current regime where I_M marks the onset of non-Ohmic behavior and $I_M \to 0$ as $T \to T_{2D}$. The data were taken in a rf-shielded environment as rf noise would affect the low-resistance data. A calibrated Speer carbon resistor was used as the thermometer.

In Fig. 1(a) we show a power-law fit to the data plotted as $\ln R$ vs $\ln(T - T_{cj})$, where T_{cj} is an adjustable parameter previously associated with a phase-ordering transition temperature between the individual NbN grains in the film. We had earlier reported[9] similar power-law dependences $[R \propto (T - T_{cj})^{3.7}]$ for $R > 1$. The new data agree with the previous results in the region of overlap, but show deviations from power-law behavior below 1 Ω. Figure 1(b) shows the same data plotted on a semilogarithmic scale vs $(T - T_{2D})^{-1/2}$. As seen from this plot, all the data are fitted very well by an exponential expression

$$R = a \exp[b(T - T_{2D})^{-1/2}], \tag{1}$$

where a, b, and T_{2D} are constants determined by the fitting procedure. This exponential dependence was predicted by Halperin and Nelson (HN)[10] and

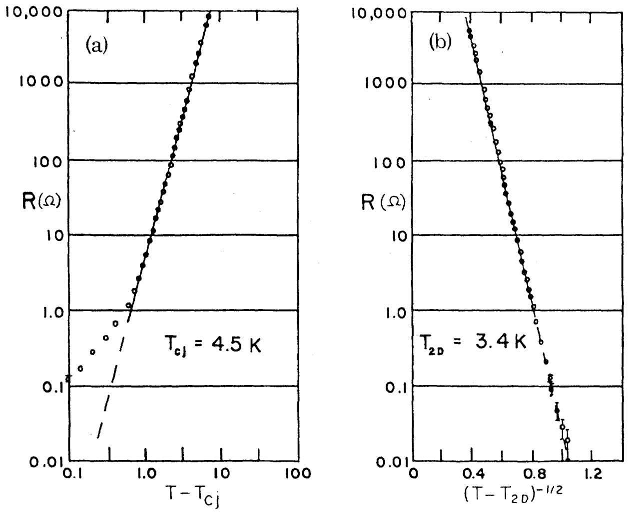

FIG. 1. (a) $\ln R$ vs $\ln(T-T_{cj})$ for granular NbN with T_{cj} of 4.52 K. (b) $\ln R$ vs $(T-T_{2D})^{-1/2}$ for granular NbN with T_{2D} of 3.42 K.

Doniach and Huberman (DH)[11] for free-vortex resistance above the 2D vortex-antivortex pairing transition. Computer fits to the data gave values of $a = 2\times10^7\ \Omega/\square$, $b = 21.3$, and $T_{2D} = 3.42$ K.

Quantitative interpretation of these new data remains speculative, however, because of unexpectedly large values for the various parameters in the fitted equation. The constants a and b in Eq. (1) are nearly 2 orders of magnitude larger than theoretical estimates. Also, there is reason to wonder why Eq. (1) agrees with the data over such a wide temperature range— specifically, in the region above T_{cj} where the power law also fits the data. Direct attempts to resolve these questions are difficult since in the range where the power law and exponential both work, the two mathematical expressions are approximately equal numerically over 4 orders of magnitude. This numerical coincidence is a result of the large value for b in expression (1) and the large exponent (3.7) in the power-law expression. Likewise, attempts to uniquely fit Eq. (1) using only data below T_{cj} were not meaningful since the data in this region have larger relative error and a three-parameter fit does not permit significant determination of the constants. A resolution of the question as to the correct temperature dependence above T_{cj} will require more theoretical and experimental input on 2D phase transitions in granular systems.

Notwithstanding the above mentioned difficulties there are several significant relationships that are realized in this granular 2D system. First the constants a and b are supposed to be related by the expression[10]

$$b = 2[(a/10.8R_n)(T_c' - T_{2D})]^{1/2}, \qquad (2)$$

where R_n is the normal-state sheet resistance of the sample and T_c' is the Ginzburg-Landau (GL) transition temperature for a Josephson-coupled, granular 2D superconductor. To check this relation between the constants, we used the maximum resistance of our films $R = 20\,000\ \Omega/\square$ and took T_c' to be T_{cj} (=4.52 K) which was obtained from the power-law fit of the higher-resistance data ($R > 1\ \Omega$) (see discussion below). Using these values, Eq. (2) is satisfied to 2%, well within experimental uncertainties. Secondly, the temperature T_{2D} corresponds to a characteristic temperature in our films where the current-voltage (I-V) characteristics changed pronouncedly.

Figure 2(a) shows I-V characteristics at temperatures above, slightly below, and well below T_{2D}. A characteristic step appears at T_{2D} and becomes rather pronounced below T_{2D}. Figure 2(b) shows dV/dI data for the same films, indicating how the step evolves with temperature. The temperature dependence of this step is identical to that of the critical current for a Josephson junction using T_{cj} as the transition temperature.[9] Likewise, the product I_cR_s, where R_s is the differential resistance above the voltage step, is equal to 2Δ $[=3.5kT_{cj}]$, indicating ideal Josephson behavior. The absolute magnitude of the critical current $I_c(0)$, however, is several orders of magnitude smaller than that predicted in KT theory. The nature of I_c in granular films also needs further clarification, but appears consistent with a model associating I_c with localized phase-slip excitations.[12]

There are also a number of significant relationships in the power-law region of the data ($R > 1$). We had reported[9] the existence of universal scaling behavior of V with current, temperature, and magnetic field about T_{cj}. This involved both a single universal equation with several exponents and several limiting forms each with a characteristic power-law dependence. In particular,

$$V \propto I^x \chi((T - T_{cj})/I^\lambda) \qquad (3)$$

with limiting forms $V \propto I^3$ at $T = T_{cj}$ and $R \propto (T - T_{cj})^{3.7}$ as $I \to 0$. Here χ is a universal function of argument $\Delta T/I^\lambda$ and x and λ are constants. Similar scaling results have also been observed by Epstein, Goldman, and Kadin.[13] In fact, Hurault[14] has predicted a cubic power-law depen-

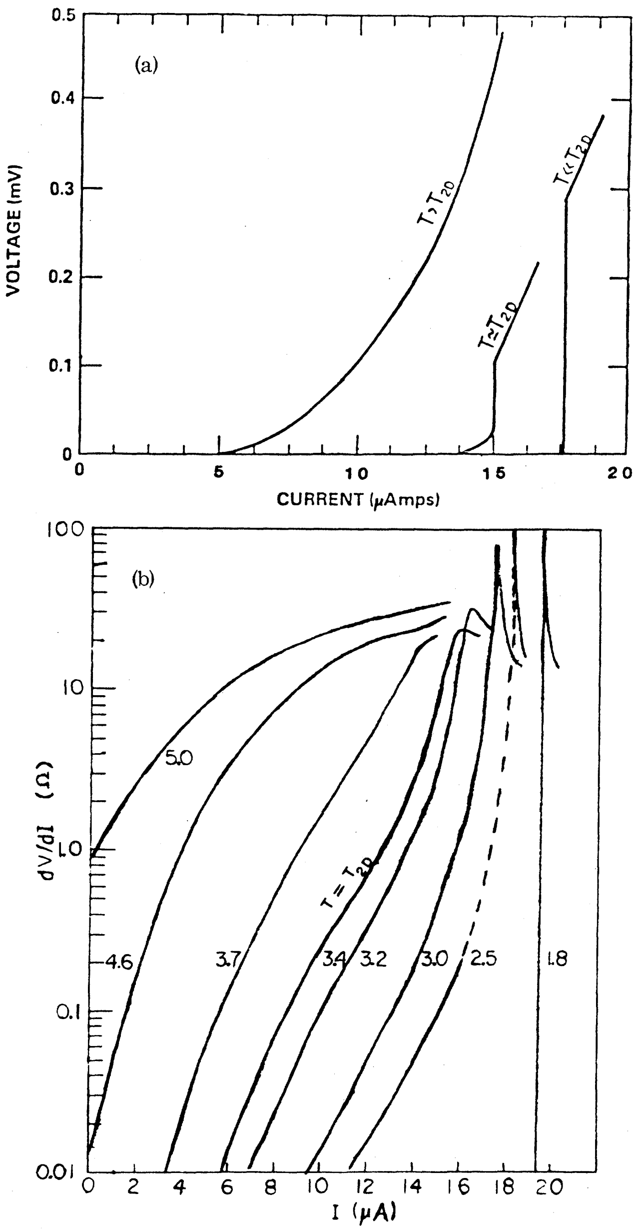

FIG. 2. (a) Voltage vs current for granular NbN at temperatures above, just below, and far below T_{2D}. (b) dV/dI vs I for granular NbN showing the temperature evolution of the structure beginning at $T_{2D} = 3.4$ K.

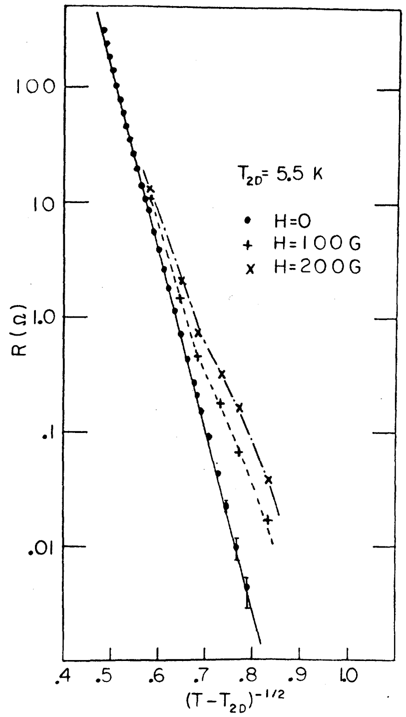

FIG. 3. $\ln R$ vs $(T - T_{2D})^{-1/2}$ for granular NbN film in zero field, 100 G, and 200 G.

dence $V \propto I^3$, for a 2D superconductor in the mean-field regime. Maekawa, Fukuyama, and Kobayashi[15] have also discussed a mean-field T_c in granular films. We therefore believe the power law and scaling relations previously reported have significance in a mean-field sense for 2D granular samples. We associate T_{cj} extracted from the data with the mean-field GL transition temperature for a Josephson-coupled, granular 2D superconductor.

The ratio T_{2D}/T_{cj} depends on R_n of the film according to the approximate relation,[3,16] $T_{2D}/T_{cj} = (1 + R_n/R_0)^{-1}$, where R_0 is a constant. Theoretically, R_0 has a value of 24 000 Ω/□ to 34 000 Ω/□ depending on strong-coupling effects. Our data, however, give $R_0 = 70\,000$ Ω/□ which, although large, is not inconsistent with other published reports.[3,17]

These remarkable resistance, I-V, and I_c dependences have not been reported in other granular systems. We believe the distinction lies within the nature of the material itself. First, according to the HN theory the size of the critical region τ_c and the number of vortex excitations n_f are given by

$$\tau_c \propto n_f(\tau) \propto [\xi^2(\tau_c)]^{-1}, \qquad (4)$$

where $\tau \equiv T - T_{2D}$; $\tau_c \equiv T_{cj} - T_{2D}$; and $\xi(\tau_c)$ is the GL coherence length for the material evaluated at τ_c. The ratio of the square of coherence lengths for different materials represents the relative size of the critical region and the relative number of thermally excited vortices in films of each material. Comparing granular NbN to granular Al, one finds this ratio[18] to be 10^2–10^3. Since the resistivity above T_{2D} is dependent on the number of vortices, the temperature region over which thermally excited vortices dominate the resistance will be much larger in NbN than in the other granular materials. By similar reasoning, deliterious effects due to residual magnetic fields, excessive measuring currents, or sample inhomogeneities will be much less pronounced in NbN films.

Figure 3 illustrates these facts by showing the relatively small effects of a perpendicular magnetic field on the temperature dependence of the resistance for a planar sample. At fields of 10 G or less, the resistance change was negligible, i.e., $B=0$ limit, in sharp contrast to the observations by Masker, Marcelja, and Parks[19] for granular Al. Similarly, to avoid finite-current effects (non-Ohmic behavior) one must be in the $I\rightarrow 0$ limit, i.e., $I<I_M$ where $I_M \propto n_f$. In the NbN films reported here, I_M was approximately 0.2 μA as the sample resistance approached zero, i.e., $R \lesssim 0.1\ \Omega$. For other granular systems with larger $\xi(\tau_c)$, I_M will be several orders of magnitude smaller, thereby making voltage detection in the $I\rightarrow 0$ limit difficult. Likewise, variations in grain size d (sample inhomogeneities) are relatively less important for materials having ξ of comparable size since $\Delta\psi/\psi \propto (\xi/d)(\Delta d/d)$, where ψ is the superconducting order parameter of a grain. In our NbN films $\xi/d \sim 1$, while the distribution of grain size is small, $\Delta d/d \approx 20\%$. No sample inhomogeneity effects have been observed in our NbN samples.

In conclusion, we speculate that a 2D topological phase transition is occurring in our granular samples at T_{2D}, below a mean-field GL transition temperature T_{cj}. Theoretical explanation of the large values for the various parameters is needed. The small coherence length ξ of NbN greatly facilitates these observations and significantly lessens many experimental difficulties.

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