Army Research Office. A more complete report will be published elsewhere.

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Universal Current Scaling in the Critical Region of a Two-Dimensional Superconducting Phase Transition

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The transition to a superconducting state in ultrathin (~30 Å) granular niobium nitride is found to obey scaling laws characteristic of critical regions near phase transitions. Dynamic scaling of voltage with current is theoretically suggested in analogy with magnetic phase transitions and is experimentally demonstrated. A universal function of current and temperature involving two independent critical exponents is generated.

Critical phenomena have been of great interest for many years. Unfortunately, methods developed for studying the critical region of a phase transition are not readily applicable to the usual bulk superconducting transition because of experimental difficulties in exploring the very narrow critical region ($< 10^{-6}$ K). Recently, unusually broad transitions to the superconducting state have been observed in granular- and ultrathinfilm systems, suggesting that a superconducting critical region may be experimentally accessible in these materials.¹⁻⁶ This Letter reports the first demonstration of universal current scaling in the critical region for a superconducting transition of two-dimensioanl (2D) granular NbN films. It is shown that the dynamic variable voltage Vcan be expressed in terms of a universal function of current I and temperature T involving two independent critical exponents.

The recognition that voltage near a superconducting transition should scale with current and temperature arises by analogy to other critical phenomena.⁷⁻⁹ The zero-field magnetic susceptibility χ of an Ising ferromagnet, for example, diverges as T approaches the critical temperature T_c from above like $\chi \equiv [\partial M/\partial H]_{H=0} \propto (T - T_c)^{\gamma}$, where M is the magnetization and H is the magnetic field. While the zero-field χ is infinite at T_c , the finite-field χ is not. Thus at T_c , M(H) is not a linear function, i.e., $H \propto M^{\delta}$ where $\delta > 1$. More generally, $M(H, T - T_c)$ satisfies a wellknown scaling equation of state. By analogy, the electrical conductivity $\sigma \equiv [\partial j / \partial E]_{E=0}$ (j is the current density and E the electric field) is infinite at the superconducting T_c only for E = 0, e.g., σ $\propto (T - T_c)^{-\mu}$

Similarly, $E \propto j^x$ with x > 1 at T_c and $E(j, T - T_c)$ is expected to obey a scaling equation for $T \ge T_c$. On a microscopic level, scaling of $M(H, T - T_c)$ follows from the fact that both H and $T - T_c$ $(\equiv \Delta T)$ scale, with different exponents, with changes in the length scale. Since current density would also, by dimensionality arguments, scale like dimensionality-dependent powers of the length and time scales, one would also expect scaling behavior in this case. By analogy to formalism developed for other critical phenomena, as T approaches T_c from above, one can write

$$E(j,t) = Bj^{x}\chi(Dt/j^{\lambda}) = Bj^{x}\chi(z)$$
(1a)

 \mathbf{or}

$$V(I,\Delta T) = bI^{x}\chi(d\Delta T/I^{\lambda}) = bI^{x}\chi(z), \qquad (1b)$$

where $t = (T - T_c)/T_c$, *B* (*b*), and *D* (*d*) are sample-(and geometry-) dependent constants, *x* and λ are sample-independent critical exponents, and $\chi(z)$ is a universal scaling function. Since $V \propto I^x$ at T_c and $R \propto \Delta T^{\mu}$ as *I*-0, one finds

$$\chi(z) \rightarrow 1 \text{ as } z \rightarrow 0, \qquad (2)$$

$$\chi(z) - z^{\mu} \text{ as } z - \infty,$$

thus

 $x - \mu \lambda = 1,$

which provides a relationship between the critical exponents x, μ , and λ and illustrates the manner in which the parameters b' and d' are experimentally determined.

Below T_c , there exists a critical current density j_c where superconductivity is destroyed. As $T + T_c$ from below, $j_c + 0$. This vanishing of the scale of currents on which superconductivity persists, is consistent with Eq. (1). Indeed one expects that at small but finite current density σ_{j+0} $\propto [T_c(j) - T]^{-\tilde{\mu}}$ and $T_c - T_c(j) \propto j^{\theta}$, with $\theta < 1$. Using arguments similar to those used to relate the critical exponents above T_c , one finds that if universality is obeyed, i.e., $\mu = \tilde{\mu}$, then $\theta = \lambda$. Hence, λ obtained from the critical exponents above T_c should describe the critical form of j_c below T_{c^*}

The formalism just presented relating voltage and current is applicable in the critical region of a phase transition and is expected to apply to the broad transitions in granular- and ultrathin-film superconductors if these broad transitions are, in fact, due to critical fluctuations associated with a superconducting transition. For the 2D granular NbN system investigated in this Letter, one expects the critical region to be large since the resistance per square, R_{\Box} , is large. According to the results of Ref. 3 (see also Ref. 7), modified for the 2D case, the critical region is large when R_{\Box} is of the order $10^4 \Omega/\Box$. This is indeed the range in which the granular NbN samples fall as indicated in Table I.

The samples were prepared from reactively sputtered cylindrical niobium nitride films 250-350 Å thick that were subsequently thinned by anodization to nominally 20-30 Å.² Both ends of the samples were protected against anodization so that electrical connections could be soldered to the films.

When a Nb or NbN film is thinned by anodization to less than the grain size (~ 100 Å for our films) fairly uniform regions of mixed oxides of niobium form between the grains in the last remaining metallic layer, separating them from one another by a resistive barrier. As the anodization proceeds further, the grains become more isolated since the mixed-oxide region separating the grains grows thicker and achieves a higher state of oxidation. The temperature dependence of the resistance of these films changes quite drastically from a clearly metallic behavior $[R_{300}/$ $R_{30} > 1$ when the anodized film thickness d is greater than 60 Å to a semiconducting or hoppingconductivity behavior $[R_{300}/R_{30} < 1]$ when d < 40 Å. Above the grains, the niobium nitride has been completely anodized to form Nb₂O₅-N, a good insulator.

Previously reported experiments have shown that similar films, but with smaller intergranular separations, exhibit a paraconductivity associated with zero-dimensional superconductivity above a temperature T_{cg} (about 10 K) and a zero-resistance (superconducting) state with a critical current characteristic of a Josephson junction at temperatures below a lower temperature T_{cJ} . It is around this lower critical point T_{cJ} that the voltage universally scales with current and T $-T_{cI}$.

Extreme care was required in performing these measurements since the voltage-current charac-

TABLE I. Sample characteristics.

Film	Thickness (Å)	$\begin{array}{c} R \square \\ (\Omega/\Box) \end{array}$	Т _с ј (К)	μ	x	λ
1	30±3	6600	6.75	3.6-4.2	2.8-3.2	0.4-0.6
2	30 ± 3	9090	5.50	3.6-4.2	2.8 - 3.2	0.4-0.6
3	23±3	7800	3.60	3.6-4.2	2.8-3.2	0.4-0.6

teristics were very sensitive to the presence of external rf interference. Each lead was individually filtered and the experiments were performed inside a shielded room.

 $T_{c\,\rm J}$ was experimentally defined as the *T*-axis intercept of a continuous voltage-versus-temperature plot at a constant current of 400 nA. Values of $T_{c\,\rm J}$ for the three samples are indicated in Table I. The nominal thicknesses of the niobium nitride samples, estimated from the anodization voltage, are also shown in the table.

Figure 1 shows log-log plots of $R_{I=400 \text{ nA}}$ vs ΔT and $V_{T=T_{cJ}}$ vs I. The parallel straight lines for all samples demonstrates a consistent agreement with the two limiting cases of the universal function $\chi(z)$ and immediately defines the two critical exponents x and μ and the two experimental constants b and d. With use of the relation between exponents, $x - \lambda \mu = 1$, λ can be calculated. The values of these exponents are given in Table I. The uncertainty in the exponents indicated in the table is primarily due to the difficulty in precisely determining T_{cJ} . For the very narrow range of T_{cJ} 's consistent with the data, the corresponding range of exponents is reported.

Figure 2 is a plot of the universal scaling function $\chi(z) \equiv V/bI^x$ vs $z \equiv d\Delta T/I^{\lambda}$, using x = 3 and $\lambda = 0.54$. *b* and *d* were experimentally determined constants for each sample. The points represent values obtained from voltage measurements taken at various ΔT and *I* values covering the entire range of data. Thus all *I-V* curves for all three samples are accurately described by Eq. (1).



FIG. 1. (a) R vs ΔT plotted on a log-log scale for samples 1, 2, and 3. The lines all have a slope of 3.7. (b) V vs I at $T = T_c$; plotted on a log-log scale for all three samples. The lines have a slope of 3.

Maximum ΔT 's used were about 1.3 K; hence, the critical region for these granular films is at least 1.3 K in width.

The value of 3 for the critical exponent x reported here is consistent with several experiments reporting nonlinear electric-field effects near T_c .¹⁰ In fact, theories based on a modified time-dependent Ginzburg-Landau equation predict a current density proportional to $E^{1/3}$ ($V \propto I^3$) at high electric fields, close to T_c .^{10,11}

The value of the critical exponent μ listed in Table I is quite different from those previously reported for other superconducting systems. For most granular systems reported, the μ values are near 1 which are characteristic of values expected for a transition described by random percolation theory.³⁻⁵ Chien and Glover¹² have reported values for μ of 2 for amorphous bismuth films. In the case of a random percolation transition there are theoretical questions as to whether this transition is, in fact, a critical phenomenon,



FIG. 2. Scaling function $\chi(z)$ vs z plotted on a loglog scale for samples 1, 2, and 3. The solid line has a slope of 3.7.

and for the amorphous bismuth film, the critical region was only a few millikelvin wide, making an exact determination of μ difficult. The data of Strongin $et \ al.^7$ show a critical region 0.2 K wide due to their large R_{\Box} ; when these data are reanalyzed to obtain a value for μ , the result is consistent with the values reported here on granular NbN films. It is believed that a rather uniform separation of the individual grains is produced by our anodization technique and by the air oxidation technique of Strongin et al. This uniform coupling of the grains violates the assumptions of random percolation theory and permits the observation of critical exponents which, we believe, are characteristic of a 2D superconducting transition. We emphasize that, although the existence of a superfluid transition in the neutral 2D case has now been fairly well established,¹³ the same is not true for the charged superfluid. Our results, particularly the scaling-law behavior, strongly support the existence of such a superconducting state.

While the role of current density as a scaling variable is demonstrated in this Letter for a 2D superconducting transition only, it is suggested that current scaling is a more general phenomenon and will apply to phase transitions where conductivity becomes zero as well as infinite, i.e., $\sigma \propto (T - T_c)^y$, where |y| > 1. The scaling form of E as a function of j and the parameter driving the transition should remain the same as Eq. (1) which describes one case where the critical-point analogy holds for a nonequilibrium system.⁶

In conclusion, critical behavior analogous to that seen for other phase transitions has been observed for the first time at a two-dimensional superconducting transition. The unique characteristics of the samples, i.e., large R_{\Box} and uniform granular coupling, broaden the critical region and made it experimentally accessible. Universal scaling of voltage with current and ΔT has been demonstrated and the critical exponents have been determined. It is suggested that current scaling may be a general property of transitions where the conductivity is critical.

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