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Coherence and Disorder in Arrays of Point Contacts

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By measuring the penetration depth and the critical current of three-dimensional assemblies of weakly coupled superconducting grains as a function of temperature, two critical exponents, β and ν', were determined characterizing the transition to coherence of the system. As in calculations on disordered systems, strict universality does not hold, but the exponents are shown to be compatible with the hypothesis of weak universality.

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Since the early work of London[1] supercurrents and the penetration depth of superconductors appear as manifestations of phase coherence, or order in momentum space, among the superconducting electrons. Similarly Josephson currents and penetration depth result from phase correlations between superconductors separated by a barrier. By the same token, one may expect intuitively that an array of a great number of junctions (such as a granular superconductor) will display supercurrents and static screening of external fields provided long-range correlations exist between superconducting phases in different electrodes. We report here on measurements of critical current and penetration depth of three-dimensional (3D) assemblies of bulk grains (diameter $a \lesssim 50$ μm) weakly coupled through Josephson point contacts.

Our samples are made by simply pressing together in an epoxy resin slightly oxidized superconducting Nb grains. We thus obtain arrays of about 10^6 point contacts which can be molded to the desired shape, in this case cylinders 3 mm in diameter and 13 mm long. Resistivities ρ_n in the range 0.1–1 Ω cm are easily obtained by controlling the pressure on the system during hardening of the resin. On the basis of resistive measurements we have pointed out[2,3] that a phase transition from a high-temperature "paracoherent" state to a low-temperature coherent one takes place at a temperature $T_0 < T_c$. This is due to the fact that superconducting phases in individual grains can undergo thermal fluctuations that destroy coherence down to $k_B T_0 \simeq 2F_J/z$, where F_J is the coupling energy per junction and z the number of first neighbors.

The existence of a second (coherence) transition as distinct from a first (superconducting) one in individual grains has been discussed by Deutscher, Imry, and Gunther,[4] and shown to require weakly coupled large grains, i.e., $\rho_n \gg \rho_2 \simeq \rho_1(a/a_0)^{3/2} \gg \rho_1(\Omega\ \mathrm{cm}) \simeq 10^{-8}/[a(\mu\mathrm{m})]^2$, where $a_0 \simeq 30$ Å. Both conditions are obviously satisfied by our samples. Furthermore, they are expected to display large critical temperature shifts[4,5] $(T_c - T_0)/T_c \simeq 10^{-3}\rho_n(\Omega\ \mathrm{cm})/a(\mathrm{cm})$ and extended critical regions.[3,4] Existing evidence on (single-transition) granular Al films[6] give a Gorter-Casimir $[1-(T/T_0)^4]^{-1/2}$ law for the penetration depth, while the critical current j_c of some 2D NbN granular films[7] with evidence of a double transition show a kind of crossover from a $j_c \sim t^{1.7}$ $[t = (T_0 - T)/T_0]$ dependence, close to the mean-field $t^{3/2}$ expression, to a Josephson-like behavior at low temperature. The latter have $a > \xi_0$, the superconducting coherence length in the grains, in common with our samples.

It may be easily shown[8] that the transition at T_0 in our case is described by an X-Y model with order parameter $\Psi = \langle e^{i\varphi}\rangle = |\Psi| e^{i\tilde{\varphi}}$, where φ is the superconducting phase of a grain and the angular brackets denote thermal average. The questions we would like to answer then are the following: (i) What are the critical exponents (if any) of the coherent penetration depth λ_c and maximum supercurrent density j_c and how are they related to the X-Y–model predictions? (ii) What are the effects of disorder (in coupling strength, in grain size, in number of first neighbors, in junction orientation,...) inevitably present in our samples?[9] Does it result in a smeared transition with no well-defined transition temperature or does it bring about a renormalization of T_0 and possibly of critical exponents?[9,10]

Before discussing the experiments let us make the relationship between penetration depth and supercurrents somewhat more precise. When an external field $\vec{H}$ is applied, a "microscopic" field $\vec{h}$ sneaks around and into the grains down to a superconducting penetration depth λ_s. An average over a volume element containing many grains gives $\vec{B}(\vec{r}) = \langle \vec{h}(\vec{r})\rangle = \nabla \times \vec{A}$. If there were no Josephson coupling between grains, the only effect would be the almost perfect $(a \gg \lambda_s)$ diamagnetism of the grains, i.e., $\vec{B} = \mu\vec{H} \simeq (1-f)\vec{H}$, where f is the metal filling factor of the sample (typically $f \simeq 0.5$). On the other hand, in a coherent state due to Josephson interaction, screening currents will develop in a region of the order of λ_c. In the presence of a field the gauge-invariant gradient of the phase $\tilde{\varphi}$ is $\vec{k} = \nabla\tilde{\varphi} - 2\pi\vec{A}/\Phi_0$, where Φ_0 is the flux quantum. Using now Maxwell's equations we easily obtain, neglecting time derivatives and dissipative terms,

$$\nabla^2\vec{k} = (8\pi^2\mu/\Phi_0 c)\vec{j}(\vec{k}, T), \tag{1}$$

where $\vec{j}$ is the supercurrent density.

Equation (1) is the analog of the Ferrell-Prange equation[11] in our multiple-junction system, once the functional dependence of $\vec{j}$ upon $\vec{k}$ is known or obtained from a model.[12] Actually, we shall need this dependence only in two limiting cases, namely $k \to 0$ and $k \to \infty$ (or a physical cutoff $k \sim 2\pi/a$). When $k \to 0$ it may be shown[13] that $\vec{j} = F_J(z\pi c/\Phi_0 a) \times |\Psi|^2\vec{k}$, and Eq. (1) becomes $\nabla^2\vec{k} = \vec{k}/\lambda_c^2$ with

$$\lambda_c = \frac{\Phi_0}{2\pi}\left(\frac{a}{2z\pi\mu F_J|\Psi|^2}\right)^{1/2} \tag{2}$$

the coherent penetration depth. For $T_0 < T_c/2$ we expect $F_J(T < T_0)$ to be independent of temperature and therefore $\lambda_c \sim t^{-\beta}$ $(T \to T_0^-)$, where β is the critical exponent characterizing the growth of the order parameter $|\Psi|$ below T_0. The limit $k \to \infty$ should apply for large fields over most of the sample. We then expect that $j \to 0$ because of destructive interference effects due to orientational disorder in the junctions. This may be easily seen[12] by computing the total current due to contributions from randomly oriented junctions, provided that individual currents are assumed to be odd bounded periodic functions of the phase difference across each barrier.

The experimental setup is classical: Mumetal and superconducting shields reduce the external field on the sample to less than 1 mG. All leads have low-pass filters to prevent perturbation by spurious rf fields. Since the passage to finite voltages in the $V(I)$ characteristics is not sharp, we have instead measured the dynamical resistance by conventional four-point and lockin techniques. The critical current is then defined by the appearance of a finite resistance, typically of the order of 10^{-3} the normal resistance of the sample.

In the penetration-depth measurement, a pickup coil closely wound around the cylindrical sample gives a signal proportional to the ac flux seen by the sample and produced by a larger coil in the He bath. This flux can be related to the coherent penetration depth [Eq. (2)] by solving Eq.

(1) in the limit $k \to 0$:

$$\Phi_c = 2\pi R \lambda_c [I_1(R/\lambda_c)/I_0(R/\lambda_c)]\mu H \quad (H \to 0), \qquad (3)$$

where R is the radius of the sample and I_1 and I_0 are the modified Bessel functions of order 1 and 0, respectively. In strong fields the flux is

$$\Phi_s \simeq \pi R^2 \mu H \quad (H \to \infty). \qquad (4)$$

Equation (3) is valid only if there are no vortices in the sample, while Eq. (4) is valid in particular when the vortex density is such that $B \sim \mu H$. Now the field of first vortex penetration H_{c_1} can be estimated by linearizing Eq. (1), which takes us to the same problem as in type-II superconductors.[14] By pursuing this analogy one obtains, in order of magnitude, $H_{c_1} \simeq \Phi_0/4\pi\lambda_c^2 \simeq 10^{-5}$ Oe, and a surface barrier field opposing the entrance of new vortices of the order of the thermodynamic critical field, $H_s \simeq 1$ mOe.

In view of these estimates, we decided to apply a purely ac field of varying amplitude H_m. We then expect that, in a range where $H_m < H_s$, Eq. (3) would apply, while for $H_m \gg H_s$ vortices would be swept in and out of the sample, giving $B \simeq \mu H$ over most of the ac cycle, i.e., Eq. (4). Then by plotting the quantity (proportional to the average permeability) $v = V/H_m$, where V is the voltage from the pickup coil, as a function of H_m, one should find two regions where $v =$ const, $v = v_c$ and $v = v_s$ for small and large fields, respectively. This can be seen in the typical data of Fig. 1, where it is worth noticing that v_s is obtained for flux densities of about one flux quantum per cross section of the smallest grains in the sample. Furthermore, v_s should be practically independent of temperature in the range studied, which is actually the case. The coherent penetration depth is obtained by solving the transcendental equation

$$\frac{\lambda_c}{R}\frac{I_1(R/\lambda_c)}{I_0(R/\lambda_c)} = \frac{1}{2}\frac{v_c}{v_s} \qquad (5)$$

for each temperature. The critical exponent β was then found by plotting $\lambda_c^{-1/\beta}$ as a function of temperature and varying β until the data fell on a straight line, corresponding to the law $\lambda_c \simeq \lambda_c(0) \times t^{-\beta}$. The critical temperature T_0 results from linear extrapolation. The value $\beta = 0.7 \pm 0.1$ fits all samples studied (Fig. 2). No acceptable fit was found for a Gorter-Casimir or mean-field law. The extrapolated $\lambda_c(0)$'s are in reasonable agreement with estimates that can be obtained from Eq. (2) with $zF_J/2 \sim k_B T_0$ (~0.4 mm for the data of Fig. 2).

Maximum supercurrent data have been analyzed in a similar way, giving a critical exponent $\xi = 2.5 \pm 0.3$, as shown in Fig. 3. For most of the temperature range in Fig. 3, $\lambda_c(T) \gtrsim R$, which implies a practically uniform current distribution. The exponent ξ is then that of critical current density, $j_c \sim t^{\xi}$. It is striking that the value of β we find is about twice the generally accepted value for the X-Y model.[15] Actually, recent calculations on a random 2D Ising model[16] result in doubling the exponent of the boundary order parameter

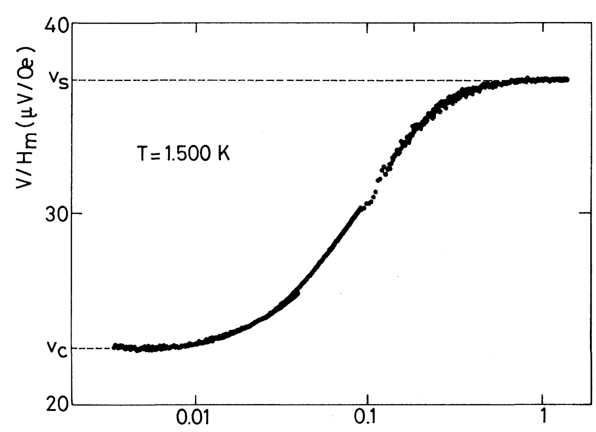

FIG. 1. Pickup-coil output divided by amplitude H_m of ac applied field as a function of H_m in a typical experiment. Two limiting behaviors, for low and high fields (v_c and v_s), are displayed.

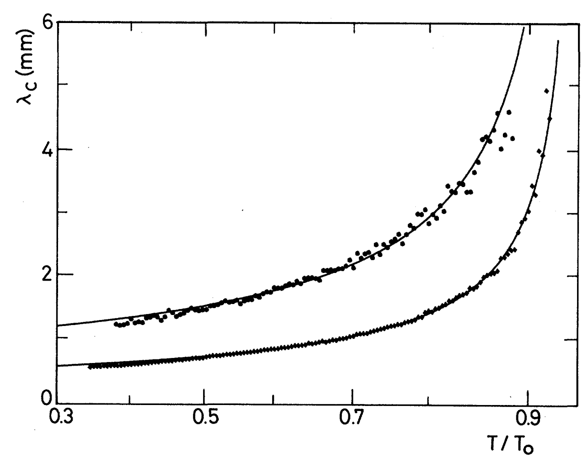

FIG. 2. Measured penetration depth as a function of reduced temperature for two different samples. Full lines are fits by the law $\lambda_c(T) = \lambda_c(0)(1 - T/T_0)^{-\beta}$, with $\beta = 0.7$. These particular samples have $\lambda_c(0) = 0.97$ mm, $T_0 = 3.95$ K and $\lambda_c(0) = 0.46$ mm, $T_0 = 4.35$ K.

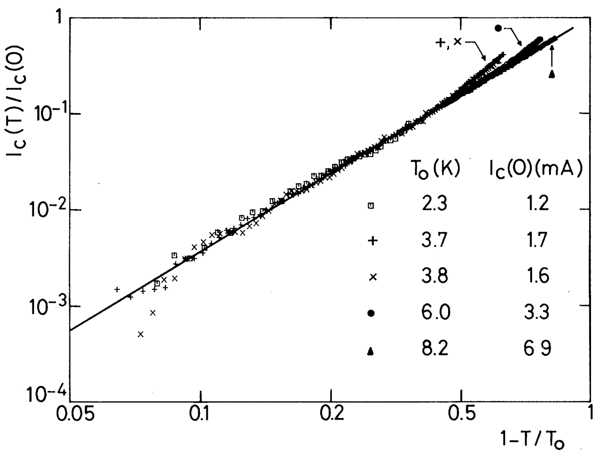

FIG. 3. Log-log plot of measured critical current against reduced temperature. Data for five different samples are shown, with coherence temperatures ranging from 2.3 to 8.2 K. The straight line has slope 2.5.

with respect to the ordered case. Similar results are obtained from a renormalization-group calculation.[17] Significant increases in the correlation-length and susceptibility exponent ν and γ have been found from high-temperature series expansions in dilute Ising models.[9,18] These calculations support the hypothesis of weak universality,[19] which is interesting to check in our case.

According to Ref. 19, exponents like β, γ', Δ, ... may change (for example, with disorder; notation is as in Fisher's paper[20]), but the ratios β/ν', γ'/ν', Δ/ν',..., as well as the exponents δ and η, should be universal; i.e., dependent only on space and order parameter dimensionalities. The exponent $\zeta \simeq 2\nu'$ if, as has been argued for percolation problems,[21] $j_c \sim \xi_c^{-(d-1)}$, where ξ_c is the correlation length and d is the number of space dimensions. On the other hand, a Ginzburg-Landau analysis would give[14] $j_c \sim |\Psi|^2 k_c$, where k_c varies as ξ_c^{-1}, in which case $\zeta = 2\beta + \nu'$. Both statements are simultaneously true if the scaling equation

$$(d-2+\eta)\nu' = 2\beta, \qquad (6)$$

where $\eta \simeq 0$ for the X-Y model,[15] holds. We obtain the prediction $\zeta = 2\nu' = 4\beta = 2.8 \pm 0.4$ in good agreement with the value found. Alternatively, we may take $\nu' = \zeta/2 = 1.25 \pm 0.15$ and obtain from the scaling relation $\gamma' = (2-\eta)\nu' = \beta(\delta - 1)$ the value $\delta \simeq 4.6 \pm 0.3$, which is close to $\delta \simeq 5$ for the classical 3D X-Y model. This is just what is predicted by weak universality.

An argument by Harris,[22] as well as renormalization-group calculations,[10] suggests that weak disorder should be relevant; i.e., it would either smear the transition or renormalize critical temperature and/or exponents of the ordered system if the latter has a specific-heat exponent $\alpha_0 > 0$. Estimates for the classical X-Y model give[23] $\alpha_0 \simeq 0 \pm 0.01$. Our data seem to indicate therefore that α_0 is nonnegative. Furthermore, the disordered transition may be expected to be sharp if the renormalized exponent $\alpha = 2 - d\nu < 0$. In other words, the specific-heat singularity may be washed out,[17] but the transition remains sharp in the sense that a critical region exists where physical quantities obey power laws in $|T - T_0|$ with a well-defined critical temperature T_0. In our samples the transitions remain sharp in the above sense. The fact that β increases by a factor of about 2, as it does in disordered–Ising-model calculations,[16,17] may be something more than a simple coincidence.

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Superconducting Tunneling in the Amorphous Transition Metals Mo and Nb

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The results of a tunneling experiment in thin films of amorphous Mo and Nb stabilized with N_2 are presented. The data were analyzed by several methods to obtain the Eliashberg function, $\alpha^2F(\omega)$. The resulting spectra are qualitatively different from $\alpha^2F(\omega)$ of amorphous simple metals, and in good agreement with computer model simulation of Rehr and Alben of the phonon spectrum of amorphous transition metals.

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The electron-phonon interaction in amorphous nontransition metals has been studied for some time by the method of tunneling spectroscopy.[1] In the present work we have extended the method to the amorphous transition metals (TM) Nb and Mo stabilized with nitrogen. Two striking results were observed: (1) The phonon density-of-states function, $F(\omega)$, which is qualitatively reflected in the Eliashberg function, $\alpha^2F(\omega)$, has no apparent signature of either longitudinal or transverse peaks—in contrast to crystalline metals and simple amorphous metals. The normalized spectrum $g(\overline{\omega}) \equiv \alpha^2F(\omega/\omega_{\max})/\alpha^2F_{\max}$ is essentially identical in both Nb and Mo; this shape is in excellent agreement with computer model simulations of the vibrational spectrum of amorphous TM.[2] (2) There is no significant enhancement of the low-energy end of the electron-phonon-coupling function, $\alpha^2(\omega) = \alpha^2F(\omega)/F(\omega)$. Lack of enhancement will result in the weak-coupling behavior observed in many amorphous TM.[3] Again, this is in contrast to amorphous simple metals where $\alpha^2(\omega)$ is inversely proportional to energy and strong coupling is common.[1] The Eliashberg equations, modified for a thin proximity effect,[4] provide an adequate description of superconductivity in those materials—to within the accuracy of the measurements.

Amorphous films of pure transition metals are highly unstable and tend to anneal at very low temperature.[3] In order to perform the tunneling experiment it was necessary to achieve high film homogeneity. By depositing the films when dry high-purity nitrogen was introduced into the system, it was possible to get very homogeneous films characterized by a narrow superconducting transition, $\Delta T_c < 0.1$ °K. T_c and ΔT_c determined by both the film and the junction resistive transitions were in agreement.

The junctions were of the Al-Al_2O_3-M type, where the Al was kept normal by maintaining the temperature above its T_c and M a TM; the advantage of this arrangement has been discussed by Robinson.[5] The aluminum film was predeposited on a quartz substrate and exposed to room air. The substrate was then mounted in an ultra high-vacuum system with a cryostat and an electron gun. During deposition the pressure was raised from a background of 1×10^{-10} to 5×10^{-7} Torr for Mo and to 5×10^{-8} Torr for Nb by leaking N_2 and the substrate temperature lowered to ~ 55 °K (lower temperature resulted in the for-

