[6]It should be noted that significant oscillation of the laminae is not to be expected under the conditions of the present experiment, as is also the case for the results of other workers to be quoted later, since in all these instances the frequency used is sufficiently high that the normal-state skin depth $\delta \ll a$. In this limit, the length of the curved phase interface within a skin depth of the surface may be calculated from Eq. (2) to be $\sim (a\delta 2)^{1/3}$. The oscillation amplitude of this section of interface is limited by eddy-current damping in the adjacent normal domain [A. B. Pippard, Phil. Mag. 41, 243 (1950)], and is expected to be $\sim \delta H_{rf}/H_c$. This gives a fractional contribution to the surface impedance $\sim (\delta/a)^{2/3} \sim 2\%$ for the present measurements.

[7]It has recently come to the author's attention that the measurements by Aomine [T. Aomine, J. Phys. Soc. Japan 25, 1585 (1968)] of the surface reactance at 0.1

MHz of a pure tin cylinder of 3-mm diameter using a method similar to that described here appear to show the same effect. These results are also included in Fig. 2 and, although somewhat scattered, are in good agreement with the data on aluminum.

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Flux-Flow and Fluctuation Effects in Granular Superconducting Films[*]

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Results are reported on the flux-flow state in granular aluminum films. Anomalously small values of depinning current are reported and attributed to the fact that the temperature-dependent coherence length is appreciably larger than the grain size in these films. Near T_c, the energy dissipated in vortex flow D ($D \propto d\rho_f/dH$, where ρ_f is the flow resistivity) was found to vary exponentially with temperature. This anomalous temperature dependence suggests that the major source of energy dissipation in this regime is the interaction of the vortex current fields with thermodynamic fluctuations. For sufficiently large values of $R_\square^N/\epsilon$ [$R_\square^N$ = normal-state resistance per square and $\epsilon = (T_c - T)/T_c$], the ρ_f-vs-H curves were found to be nonlinear in a manner not reported before in conventional flux-flow experiments. This curvature is attributed to the above-mentioned interaction between the vortices and fluctuations.

I. INTRODUCTION

Resistivity and dissipation in the flux-flow state of superconductors have been the subject of much theoretical and experimental work.[1] In this paper we discuss the low-field flux-flow characteristics of granular Al films and contrast our results with the work by other authors on both bulk samples[1] and thin films.[2-5]

Section I of this paper will serve as an introduction and a brief review of the pertinent work of the past. In Sec. II we discuss current-voltage characteristics including (i) pinning at low current densities and (ii) nonlinear effects at high currents. Section III contains work on the flux-flow resistivity, and Sec. IV discusses the interplay of flux-flow and fluctuation effects close to the superconducting transition temperature T_c.

In the presence of an applied magnetic field H a bulk type-II superconductor or a thin superconductor in a perpendicular magnetic field allows flux to penetrate in the form of quantized Abrikosov vor-

tex lines. Then if a transport current of sufficient magnitude is applied, steady-state vortex flow of velocity v is obtained where v is determined by equating the net force on a vortex $f = f_L - f_P$ (f_L being the Lorentz force $J\phi_0/c$ and f_P the pinning force) to a viscous drag force ηv:

$$\eta v = J\phi_0/c - f_P , \qquad (1)$$

where $\phi_0 = hc/2e$ is the flux quantum. The electric field E_0 arising from motion of the flux lines is given by

$$E_0 = n(v/c)\phi_0 = (v/c)H , \qquad (2)$$

where n is the vortex density such that $n\phi_0 = B \cong H$ (in a situation such as the present one where the macroscopic diamagnetism of the sample can be ignored). Defining the pinning independent-flow resistivity ρ_f as $\partial E_0/\partial J$, we obtain

$$\rho_f = \phi_0 H/\eta c^2 = DH/J^2\phi_0 , \qquad (3)$$

where D is the total power dissipated in vortex flow. Most of the interesting physics lies in un-

TABLE I. Sample parameters.

Sample	$R_{\square}^{N}$ (Ω)	$\rho(\mu\Omega\,cm)^{a}$	$T_{c}(°K)^{b}$	$(\epsilon_{c})_{expt}$
A	100	40	2.41	
B	1300	650	2.09	$1.5\times10^{-5}R_{\square}^{N}$
C	2100	1300	1.99	$0.4\times10^{-5}R_{\square}^{N}$
D	3600	1800	1.89	$0.8\times10^{-5}R_{\square}^{N}$

^{a}See Ref. 9. ^{b}See Ref. 10.

derstanding the sources of dissipation D. In bulk samples at low reduced temperatures,[1] the major contribution to D comes from Joule heating (via the transport current, which is forced to flow through the cores of the moving vortices) in the vortex cores. A local model by Bardeen and Stephen[6] has succeeded in explaining the empirical results of Kim $et\ al.$[1] [i.e., $\rho_{f}/\rho_{N}=H/H_{c2}(0)$, where ρ_{N} is the normal-state resistivity]. The experimental situation at finite temperatures, especially in thin films, is far less satisfactory and is usually plagued by various nonlinear effects. Often in films the current-voltage characteristics are either nonlinear or are linear over only a small range of applied transport current.[2,3,5] This makes defining the flow resistivity difficult[7] and discussing the sources of dissipation nearly impossible. As will be seen in Sec. II, the current-voltage curves for granular Al films are quite linear over large ranges of driving current and applied magnetic field. This allows not only a discussion of the pertinent physics found in D but also a detailed study of the mecha-
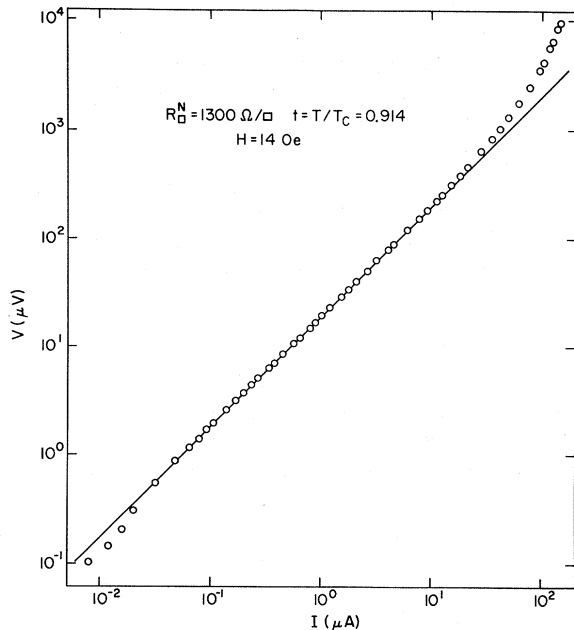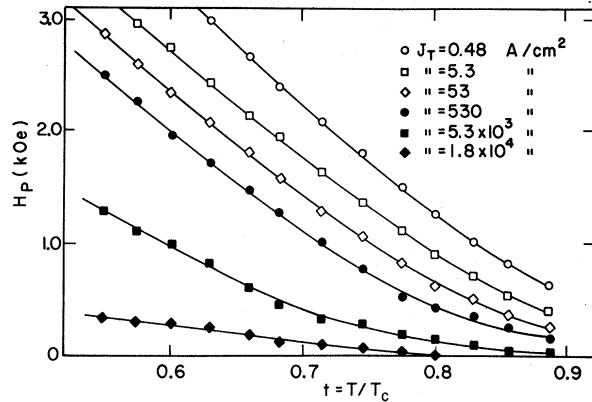

FIG. 2. Depinning field versus temperature for sample A at various transport current densities.

nisms which yield nonlinear effects seen in other samples.

II. CURRENT-VOLTAGE CHARACTERISTICS

The samples used where evaporated Al films (typical thicknesses were about 50 Å) prepared in a manner described elsewhere.[8] Resistivities were measured using a standard dc four-probe technique. The parameters of the samples studied are summarized in Table I. A typical current-voltage curve for the samples is shown in Fig. 1. There are three basic features of this curve which are worth noting: the departure from linearity at both low and high current densities and the large intervening linear region. The departure from linearity at low currents is a pinning phenomenon and marks the current where the pinning force on the vortices f_{P} can no longer be neglected with respect to the Lorentz force $J\phi_{0}/c$. The cause of the departure from

FIG. 1. Current versus voltage for sample B at $H=14$ Oe and $t=T/T_{c}=0.914$.
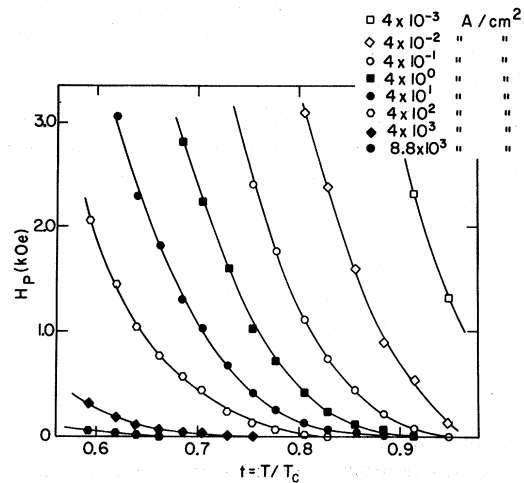

FIG. 3. Depinning field versus temperature for sample B at various transport current densities.
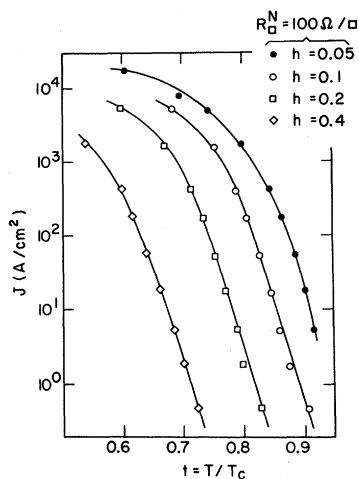
FIG. 4. Depinning current versus temperature for sample A at various magnetic fields.

linearity at large fields is not as well understood and will be discussed later in this section. To get a semiquantitative measure of the strength of pinning, we can measure either the current density $J_p(H, t)$ or the magnetic field $H_p(J, t)$ necessary to obtain some small prespecified nonzero voltage (e. g., 0.1 μV). The results are seen in Figs. 2–5 for samples with 100 and 1300 Ω/□, respectively. Note that the values of pinning current are quite small (results on bulk samples[11] seldom have pinning current densities of less than 1 A/cm²) and decrease as the normal-state resistivity of the sample is increased. This seems to contradict the usual idea that vortex pinning occurs at grain boundaries and at other sample inhomogeneities. However, this contradiction is resolved when one realizes that the temperature-dependent coherence length $\xi(T)$ (i. e., the size of the vortex cores) is larger than the sample grain size[12–14] (which decreases with increasing resistivity). Thus, since the core size is large compared to the scale of the structural disorder, the moving vortex feels a medium which is quite homogeneous. If we examine a sample with a still larger normal-state resistivity, the current-voltage curves take the form seen in Fig. 6. Note that for even the smallest measureable voltages the I-V characteristics are linear and are therefore indicative of vortex flow. Thus, even for the smallest current densities shown in Fig. 6, we are in a region where $f_p \ll J\phi_0/c$. For this sample an experimental upper limit of the pinning current (i. e., that current where the I-V curves depart from linearity) was determined and is plotted in Fig. 7. To our knowledge these pinning current densities are considerably smaller than any previously reported (even in highly annealed bulk samples).

Since the pinning effects in Fig. 4 are manifestly quite small, we can rule out pinning as a source of the nonlinearity at large J values.[15] It has been suggested[2] that nonlinear effects in films occur when the self-field (i. e., the field arising from the transport current) is no longer negligible when compared to the applied field. To check this idea we have determined the current where the I-V curve departs from linearity (determined graphically) and plotted this as a function of both magnetic field and temperature (see Figs. 8 and 9). If the self-field were causing the nonlinearity, we would expect the breakdown current I_B (i. e., that current at which the nonlinearity begins) to be proportional to the applied magnetic field and relatively independent of temperature.[16] A study of Figs. 8 and 9 implies that self-field effects alone are inadequate to describe the nonlinearity. The possibility that the nonlinearity is due to heating of the sample above the helium-bath temperature was dismissed for two reasons: First, a simple calculation using empirical values of the Kapitza resistance between metal surfaces and liquid helium[17] shows that in all cases the sample remains roughly within 10^{-3} K of the bath; second, if the nonlinearity were caused by heating, we would expect the breakdown current to decrease with increasing sample resistivity. From Fig. 8 we see that this is clearly not the case. The actual mechanism which is causing the nonlinearity is not clear and is presently the subject of further study.
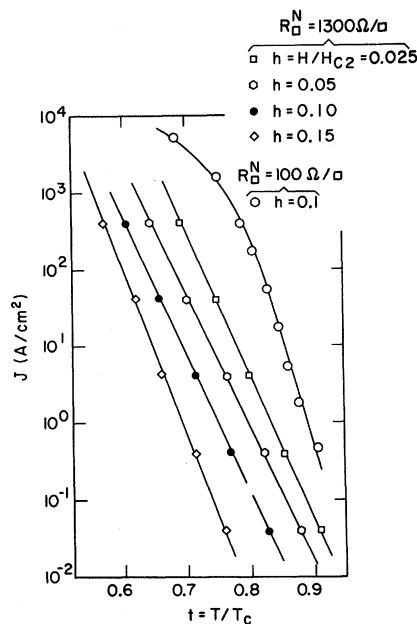

FIG. 5. Depinning current versus temperature for sample B at various magnetic fields. A representative curve from sample A is added for comparison.
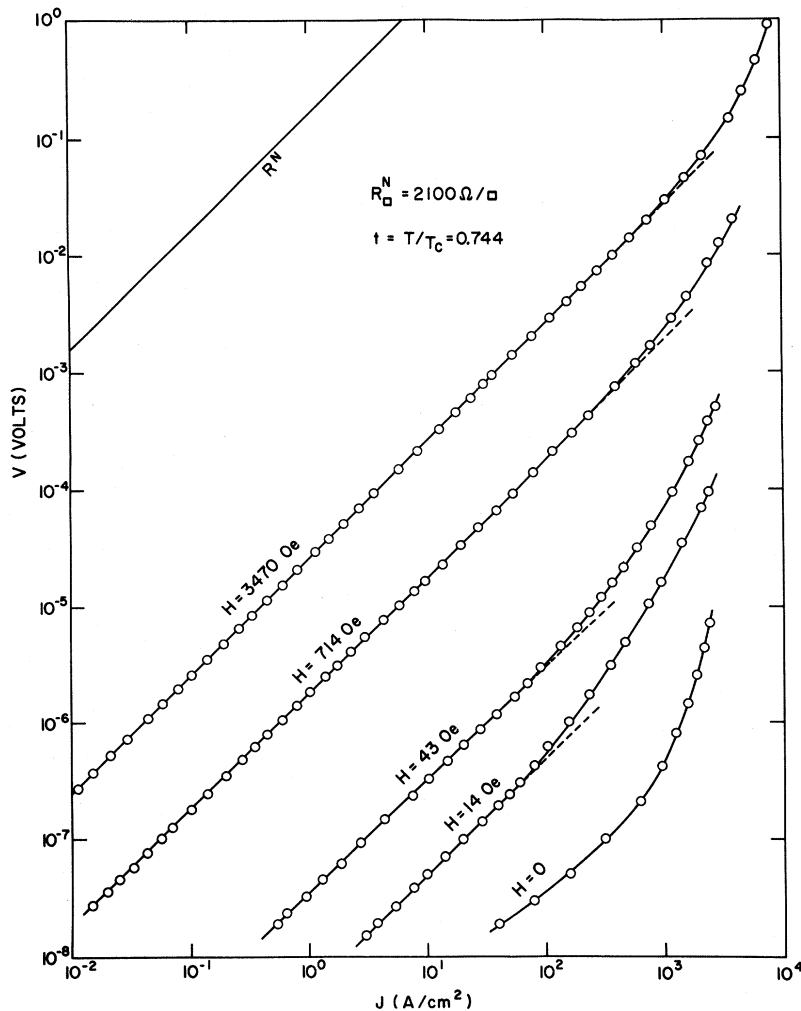
FIG. 6. Current versus voltage for sample C at $t = T/T_c = 0.744$.

III. FLUX-FLOW CHARACTERISTICS

As noted in Sec. II, the smallness of the pinning currents in the granular films allows one to observe large linear regions in the current-voltage curves. From the slopes of these curves in the linear region we can determine ρ_f (recall $\rho_f \equiv \partial E_0 / \partial J$). In Fig. 10 we have plotted ρ_f as a function of magnetic field for various temperatures. Note that near T_c (see Fig. 11) there is a large range of temperatures for which ρ_f is quite linear in applied magnetic field. However, at lower temperatures, the functional dependence of ρ_f on magnetic field deviates from linearity and takes on a power-law appearance [i.e., $\rho_f \propto H^{\beta(T)}$, where $1 \leq \beta(T) \leq 2$]. For example, note that at $t = 0.745$ the resistivity follows an $H^{3/2}$ law over more than two decades in field. These results are qualitatively similar to results reported on both bulk samples[18] and on type-I films[2] except that in bulk samples, ρ_f becomes linear in H at low temperatures instead of high ones as in our case. In the region where ρ_f is linear in H (see
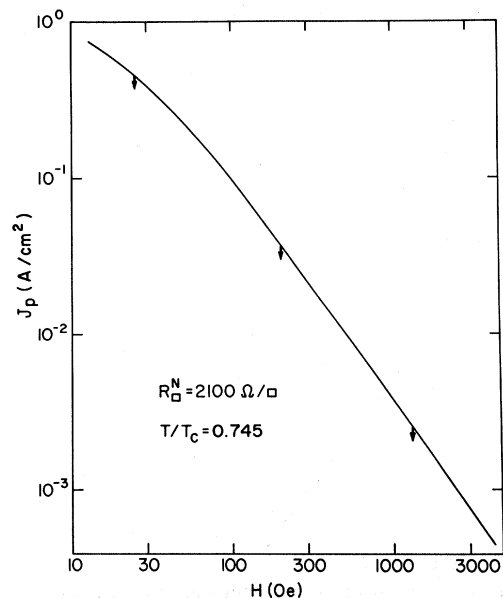

FIG. 7. An experimental upper limit of the depinning current versus temperature for sample C.
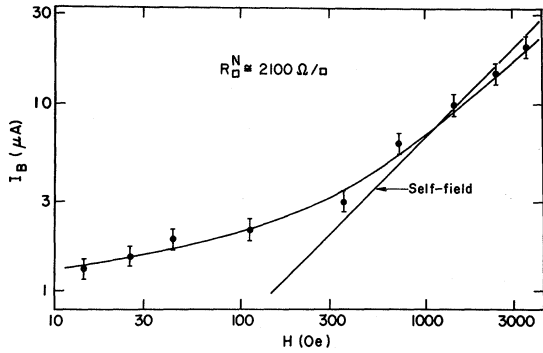
FIG. 8. Current where linearity breaks down, I_B (deduced from Fig. 6), versus magnetic field for sample C at $t = 0.744$.

Fig. 11) there is a notably sharp change in the slope $(d\rho_f/dH)$ as a function of temperature.[19] To exemplify this temperature dependence we have plotted $d\rho_f/dH$ vs $\epsilon T_c/T$ [where $\epsilon = (T_c - T)/T_c$] in Fig. 12. Note that the empirical fit is $d\rho_f/dH$ $\propto \exp[-\epsilon T_c/T(\epsilon_c)_{expt}]$, where $(\epsilon_c)_{expt}$ is a constant [values of $(\epsilon_c)_{expt}$ for the samples studied are shown in Table I]. Since [from Eq. (3)]

$$\frac{d\rho_f}{dH} \propto D \ , \tag{4}$$

we see that the dissipation of energy has an exponential character near T_c. This sharp temperature dependence is not consistent with the usual model[6] of Joule heating in the vortex cores. We present in Sec. IV a plausible explanation for this
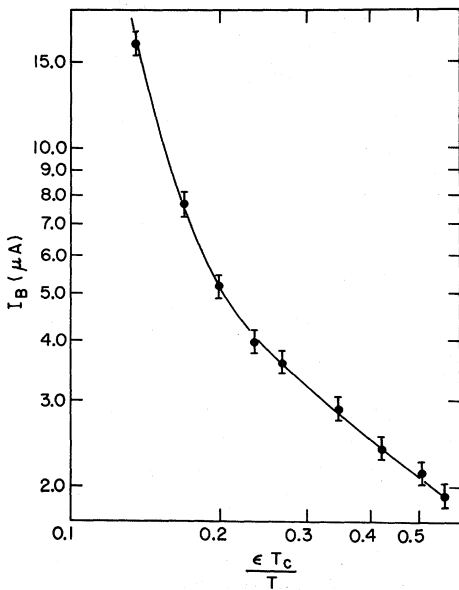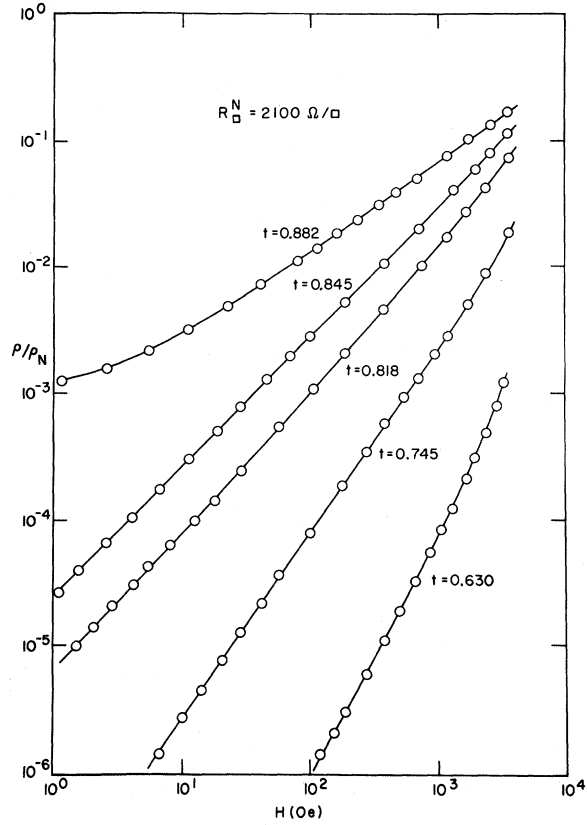

FIG. 10. Log of the flow resistivity ρ_f versus log of the applied magnetic field for sample C.

anomalous temperature dependence.

It is interesting to note that the lower points in Figs. 11 and 12 obey the relation $\rho_f/\rho_N \ll H/H_{c2}(0)$.[20] The work on bulk samples has shown $\rho_f/\rho_N \gtrsim H/H_{c2}(0)$ for all temperatures studied. The discrepancy between our results and those seen in bulk samples is consistent with other experiments on films[4,21] which suggest that the Lorentz force on the vortices may be less than $J\phi_0/c$.

IV. FLUCTUATION EFFECTS

Below but close to the superconducting transition temperature there will occur fluctuations of the superconducting order parameter. Simplistically, one may view such fluctuations as ephemeral bubbles of normal electrons. Such fluctuations may not be visible in ordinary resistivity measurements[8] since presumably the transport current can skirt the normal bubbles. Using the time-independent Ginzburg-Landau (GL) mean-field theory, it is possible to calculate the probability P of such a normal fluctuation:

$$P \propto \exp\left(-\int \Delta f \, dV/kT\right) \ , \tag{5}$$

where Δf is the free-energy difference between the

FIG. 9. Current where linearity breaks down versus $\epsilon T_c/T$ [$\epsilon = (T_c - T)/T_c$] at $H = 357$ Oe for sample C.
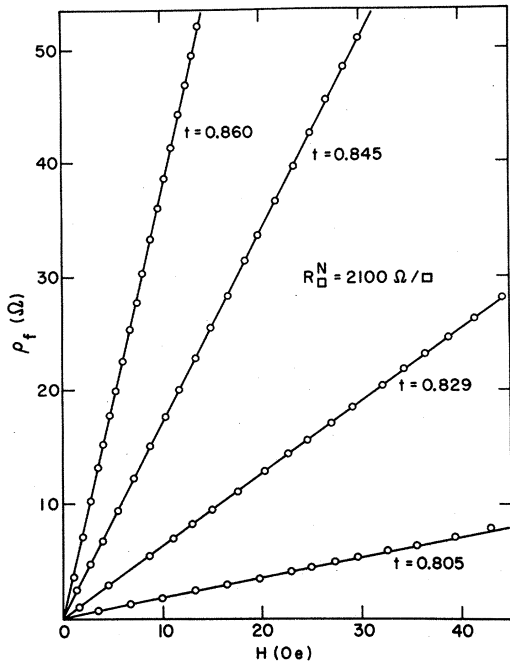
FIG. 11. Linear plot of the flow resistivity versus magnetic field for sample C at various temperatures near T_c.

normal and superconducting states and V is the mean volume of the fluctuation. The form of the GL free-energy functional implies that fluctuations of volume of order $[2\xi(T)]^3$ in three dimensions or $d[2\xi(T)]^2$ in two-dimensional films are the most prevalent fluctuations. In order to obtain a zero-order quantification of the problem, we will consider the effect of a particular fluctuation configuration, viz., a Gaussian fluctuation with mean volume $2\pi\xi^2(T)d$ which brings the order parameter
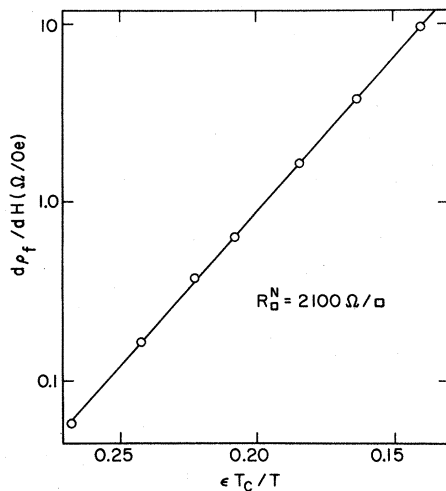

FIG. 12. $d\rho_f/dH$ deduced from Figs. 10 and 11 versus $\epsilon T_c/T$ for sample C.
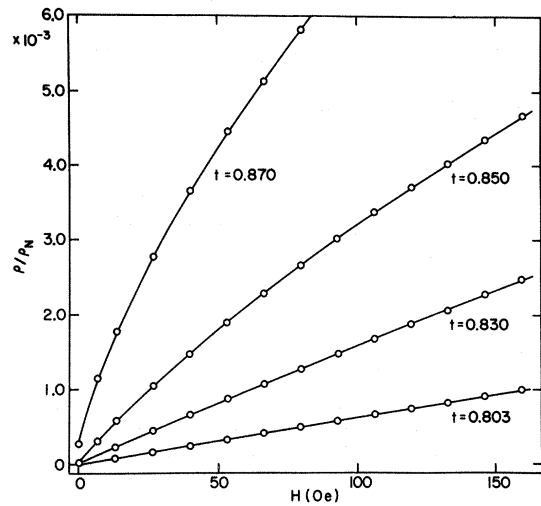
FIG. 13. Linear plot of ρ_f versus magnetic field for sample D at various temperatures near T_c.

$|\psi(r)|^2$ to zero at some point, i.e., $|\psi|^2 = |\psi|^2_{eq}$ $\times[1 - e^{-r^2/2\xi^2(T)}].^{22}$ The energy of this particular fluctuation is given (from the GL theory) by

$$\int \Delta f \, dV = \left(\frac{a^2 d}{2b}\right) 2\pi\xi^2(T) + d\int_0^\infty \left(\frac{\hbar^2}{2m^*}\right)|\nabla\psi|^2 2\pi r \, dr \ ,$$
(6)

where a and b are the standard GL parameters. Evaluating Eq. (6) yields

$$\int \Delta f \, dV = \frac{2\pi a d}{b} \left(\frac{\hbar^2}{2m^*}\right) \ .$$
(7)

Thus, the probability of a normal fluctuation, and hence the fraction of material in the normal state, is given by

$$P \propto \exp\left[-\frac{2\pi a d}{b}\left(\frac{\hbar^2}{2m^*}\right)\frac{1}{kT}\right] \ .$$
(8)

Evaluation of the parameters a and b from the microscopic theory yields

$$P \propto \exp(-\epsilon T_c/\epsilon_c T) \ ,$$
(9)

where

$$\epsilon_c = 0.52\times10^{-5} R_\square^N \ .$$
(10)

The similarity between Eqs. (9) and (10) and the empirical results shown in Fig. 12 strongly suggests that the energy dissipation is resulting from the presence of the normal fluctuations. The agreement between ϵ_c [Eq. (10)] and the experimental values in Table I is surprisingly good considering the crudeness of the above theoretical model. Improvement of this model must await a more detailed understanding of the interaction between the vortices and the normal fluctuations.

As pointed out earlier in this section, normal

fluctuations need not cause resistivity in the absence of vortex flow; however, as viewed from a reference frame tied to a moving vortex, the fluctuations move across the vortex current lines. This is analogous to the situation which leads to dissipation in the vortex cores (i.e., the vortices move across the transport current lines).

For extremely large values of $R_{\square}^{N}$ and at high reduced temperatures (viz., for $R_{\square}^{N}/\epsilon \gtrsim 3600/0.075$), the flux-flow resistivity takes the form seen in Fig. 13. This type of nonlinearity (to our knowledge, ρ_f has never been seen to decrease more slowly than linearly with H) can be explained possibly on the basis of the dissipation mechanism discussed above. If we note that the current fields of vortices in films have a long-range decay[23, 24] (of order λ^2/d, $\lambda = $ penetration depth), then as the magnetic field is increased and hence the density of vortices, the current-density profile of the overlapping current distributions changes. This can lead to a decrease in dissipation since as the vortices become closer together the probability that a fluctuation will occur at a point of decreased current density is increased. This effect is enhanced if the vortices form a symmetric lattice. Since $\lambda^2 \propto \lambda^2(0)/\epsilon \propto R_{\square}^{N}/\epsilon$, we expect the effect to occur at similar values of $R_{\square}^{N}/\epsilon$ in other samples. This

effect was indeed observed in all the samples studied at approximately the appropriate values of ϵ.

In conclusion, we have noted the following effects.

(i) Granular Al films have anomalously small values of depinning current. This is consistent with the notion that $\xi(T)$ is much larger than the grain size.

(ii) The nonlinearity in the I-V curves observed for large currents is not easily explained using a self-field argument.

(iii) The flux-flow resistivity was found to be linear in magnetic field near T_c but became increasingly nonlinear as the temperature was lowered. The existence of power laws other than $\rho_f \propto H$ for the flow resistivity seems to be a rather common phenomenon (such effects having been seen in bulk type-II[1] and type-I[25] superconductors and in type-I films[2]). These power laws are not easily understood in the framework of any of the ordinary accepted[1] sources of dissipation D and might suggest a more fundamental fault in the concept of flux flow.[26]

(iv) The anomalous temperature dependence of the energy dissipated near T_c suggests that the dissipation is occurring via an interaction between the vortex current fields and normal fluctuations.

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*Work supported by the Army Research Office (Durham).

[1]A comprehensive review is given in Y. B. Kim and M. J. Stephen, in *Superconductivity*, edited by R. D. Parks (M. Dekker, New York, 1969).

[2]Paul Thoulfsen and Hans Meissner, Phys. Rev. **185**, 653 (1969).

[3]R. Deltour and M. Tinkham, Phys. Letters **23**, 183 (1966).

[4]John S. Escher and D. M. Ginsburg, Phys. Rev. B **3**, 735 (1971).

[5]T. Ogushi, T. Takayama, and Y. Shebuya, in *Proceedings of the Twelfth International Conference on Low Temperature Physics, Kyoto, 1970*, edited by E. Kanda (Academic Press of Japan, Tokyo, Japan, 1971).

[6]J. Bardeen and M. J. Stephen, Phys. Rev. **140**, A1197 (1965).

[7]Recall that the flow resistivity ρ_f is defined as $\partial E_0/\partial J$. Thus if E_0 is not linear in J, then there does not exist a well-defined (i.e., current-independent) resistivity ρ_f.

[8]W. E. Masker, S. Marčelja, and R. D. Parks, Phys. Rev. **188**, 745 (1969).

[9]Relative uncertainties in the resistivities are about 20% resulting from changes in the film thickness due to surface oxidation.

[10]T_c was determined by a study of paraconductivity above the superconducting transition (and is the mean field T_c) as in W. E. Masker and R. D. Parks, Phys. Rev. B **1**, 2164 (1970).

[11]Some of the smallest reported pinning currents were given in R. A. French, J. Lowell, and K. Mendelssohn, Cryogenics **7**, 83 (1967).

[12]B. Abeles, R. W. Cohen, and G. W. Cullen, Phys.

Rev. Letters **17**, 632 (1966).

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[14]R. W. Cohen and B. Abeles, Phys. Rev. **168**, 444 (1968).

[15]It has been suggested by T. Ogushi et al. (see Ref. 5) that most nonlinear effects in films are due to a distribution of pinning potentials. This may indeed be the correct explanation for the results reported in Ref. 5 but is clearly not the case for granular Al films.

[16]If we assume that the breakdown of linearity occurs when the vortex density created by the driving current (due to the self-field) is some significant fraction f of the density created by the applied field, then the condition for breakdown is $H_{self}/H = f$. Noting that $H_{self} \propto I$ yields $I_B \propto fH$. Furthermore, since the vortex density is temperature independent (i.e., $n = H/\phi_0$), we would expect f to be temperature independent.

[17]G. L. Pollack, Rev. Mod. Phys. **41**, 48 (1969).

[18]A. R. Strnad, G. F. Hempstead, and Y. B. Kim, Phys. Rev. Letters **13**, 794 (1964); see also Ref. 1.

[19]This was previously seen and reported as a fluctuation effect in P. M. Horn, W. F. Masker, and R. D. Parks, Phys. Rev. Letters **26**, 177 (1971).

[20]Typical values of $H_{c2}(0)$ of about 10^4 Oe were obtained on similar films by Abeles et al. (Ref. 13).

[21]R. P. Huebener and A. Scher, Phys. Rev. **181**, 710 (1969).

[22]Note that we have chosen a typical fluctuation rather than statistically weighing fluctuations of different energies. Note, for example, that by our choice of $|\psi|^2$ we are only considering fluctuations which completely depress the order parameter. Clearly, other fluctuations

may be important when determining the flux-flow resistivity near T_c, but their inclusion must await a more exact understanding of the interaction between the vortices and the fluctuations.

[23]See, e.g., A. L. Fetter and P. C. Hohenberg, Phys. Rev. **159**, 330 (1967).

[24]J. Pearl, Appl. Phys. Letters **5**, 65 (1964).

[25]G. M. Foster and R. D. Parks, Bull. Am. Phys. Soc. **16**, 295 (1971); and private communication.

[26]The concept of flow of isolated vortices must be questioned especially where the interaction between the vortices is strong enough to create a symmetric vortex lattice. It should be noted that the concept of flux bundles (i. e., a group of vortices that are so strongly interacting that it becomes more meaningful to talk of the group as a whole instead of its individual constituents) is needed to describe the pinning characteristics [see P. W. Anderson, Phys. Rev. Letters **9**, 309 (1962)] seen in bulk samples (see Ref. 1). Possibly the concept of the flow of flux bundles instead of individual vortices will explain the power laws seen in Fig. 10.

Sharpening of the Resistive Transition of a Superconductor with the Addition of Paramagnetic Impurities*

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We present evidence demonstrating that the addition of magnetic moments to a superconductor suppresses the Maki-Thompson contribution to the fluctuation conductivity. Measurements of the resistive transition of aluminum films with erbium impurities exhibit a sharpening of the transition with increasing impurity concentration which can be explained quantitatively using Thompson's scheme for regularizing the Maki-Thompson conductivity diagrams.

INTRODUCTION

The first systematic study of the resistive transition of moderately clean thin-film superconductors by Masker and Parks[1] revealed clear disagreement with predictions based on the mean-field theory using either a diagrammatic approach[2] or a simplified time-dependent form of the Ginzburg-Landau equations.[3-5]

Maki[6] reconsidered the Green's-function approach and pointed out for the case of bulk superconductors the importance of terms in the fluctuation conductivity which were ignored in the previous microscopic calculations. These terms correspond to an enhanced conductivity of the normal electrons due to their interaction with the ephemeral Cooper pairs. Thompson[7,8] discovered that these terms were divergent in the case of thin films and one-dimensional samples in the absence of pair-breaking perturbations; he proposed a heuristic procedure for regularizing the terms in the presence of pair-breaking perturbations (which are always present in real samples). Crow and co-workers[9] tested Thompson's model by reexamining the fluctuation conductivity in clean aluminum films in the presence of an applied pair breaker, viz., a parallel magnetic field. Later, Thomas and Parks[10] carried out similar experiments on one-dimensional microstrips, and both groups found strong evidence for both the existence of the Maki-Thompson contributions and their suppression in the presence of pair breaking. In the present paper we extend the investigation to a different source of pair breaking, namely, paramagnetic impurities, and find that with increasing concentration of localized moments the anomalous excess conductivity is suppressed and the resistive transition sharpens in the manner predicted by Thompson.

THEORY

Aslamazov and Larkin[2] (AL) considered the contribution to the excess conductivity from fluctuating Cooper pairs. They found that in a two-dimensional sample, where the thickness d is much less than the temperature-dependent coherence length $\xi(T) = \xi(0)\tau^{-1/2}$, the excess conductivity above that of the normal state, i.e.,

$$\sigma' = \sigma(T) - \sigma_N ,$$

is inversely proportional to the reduced temperature $\tau = (T - T_{c0})/T_{c0}$:

$$\sigma'_{AL} = \left(\frac{e^2}{16\hbar}\right)\Big/\tau d . \tag{1}$$

This can be rewritten as

$$\frac{\sigma'_{AL}}{\sigma_N} = \left(\frac{e^2}{16\hbar}\right)\frac{R_\square^N}{\tau} \equiv \frac{\tau_0}{\tau} , \tag{2}$$