Physica C 153-155 (1988) 50-55
North-Holland, Amsterdam

GRANULAR AND SUPERCONDUCTING-GLASS PROPERTIES OF THE HIGH-TEMPERATURE SUPERCONDUCTORS

John R. CLEM

Ames Laboratory-USDOE* and Department of Physics, Iowa State University, Ames, Iowa 50011 USA

The microstructure of bulk samples of the copper-oxide high-temperature superconductors commonly is describable in terms of anisotropic grains of stoichiometric material separated by layers of nonstoichiometric interface material. The granularity strongly influences the electromagnetic properties, especially the transport critical-current density and the magnetization. In this paper, a simple theoretical model for the granularity is introduced and then used to discuss a number of electrodynamic properties (hysteretic magnetization versus magnetic field, zero-field-cooled and field-cooled magnetization versus temperature, ac susceptibility, and flux creep with logarithmic time dependence). Special attention is drawn to the importance of distinguishing between intragranular and intergranular effects.

1. INTRODUCTION

In their remarkable pioneering paper (1), Bednorz and Müller introduced not only a new class of superconductors but also some of the key concepts by which the electrical and magnetic properties of these materials could be understood. These authors noted that the sintering process used to prepare the Ba-La-Cu-O system yielded granular material. It subsequently has been confirmed by many different kinds of measurements that it is appropriate to describe all bulk copper-oxide-based superconductors as agglomerates of anisotropic grains separated by nonstoichiometric interface material. The granularity strongly affects a number of electrical and magnetic properties of the material. In this paper I introduce a simple theoretical model for this granularity and apply it to calculate several electromagnetic properties of the oxide superconductors.

2. STRONG VERSUS WEAK INTERGRANULAR COUPLING

Many of the previous theoretical treatments of granular superconductors [see, for example, Refs. (2)-(4) and the references therein] have been aimed at the understanding of small-grained (typically 50-100 Å) granular materials, such as granular Al, NbN, and Sn. In Ref. (3), for example, we studied both theoretically and experimentally the critical-current density in films of granular NbN, in which the grain sizes were of order 100 Å. We modeled the material as a cubic array (lattice parameter a_0) of identical grains of volume V_g and assumed nearest-neighbor grains to be coupled by identical Josephson junctions of maximum Josephson current I_0. We then used a Ginzburg-Landau-like free-energy functional that accounted for only the intragranular condensation energy and the intergranular Josephson coupling energy. In such a theory, the important dimensionless parameter is $\varepsilon = E_J/2E_g$, where $E_J = (\hbar/2e)I_0$ measures the Josephson coupling energy and $E_g = (H_{cg}^2/8\pi)V_g$ is the intragranular condensation energy; ε also can be expressed as $\varepsilon = 2\xi_J^2/a_0^2$, where $\xi_J(T)$ is the Ginzburg-Landau temperature-dependent coherence length. Note that ε and ξ_J are monotonically increasing functions of the temperature T, diverging at T_c. [Here we use the subscript g (for grain) to denote intragranular quantities and the subscript J (for Josephson) to denote intergranular quantities.]

When $\varepsilon \ll 1$, any intergranular currents are too small to suppress the order parameter, and the critical-current density is simply $J_c = J_0 = I_0/a_0^2$. According to the Ambegaokar-Baratoff theory (5), $I_0(T) = [\pi\Delta(0)/eR_n]F(T)$, where

$$F(T) = [\Delta(T)/\Delta(0)]\tanh[\Delta(T)/2k_BT], \qquad (1)$$

$\Delta(T)$ is the temperature-dependent gap parameter and R_n is the junction's normal-state tunneling resistance. In this limit the effects of granularity are very pronounced, and the current-carrying capacity and magnetic behavior are both dominated by the Josephson weak links.

When $\varepsilon \gg 1$, current-induced suppression of the order parameter becomes very important, and the theory then reduces exactly to the Ginzburg-Landau theory in the dirty limit (6), with the Josephson coupling energy playing the role of the supercurrent kinetic energy. The main effect of granularity is simply to introduce into the dirty-limit theory (in place of the normal-state resistivity) the effective resistivity $\rho_n = R_na_0$, where R_n is the normal-state intergranular tunneling resistance. The characteristic lengths appearing in the Ginzburg-Landau theory, ξ_J (the length scale for order-parameter variation) and λ_J (the length scale for magnetic-field vari-

* Ames Laboratory is operated for the Department of Energy by Iowa State University under Contract No. W-7405-Eng-82. This work was supported in part by the Director for Energy Research, Office of Basic Energy Sciences and in part by the Electric Power Research Institute.

0921-4534/88/$03.50 © Elsevier Science Publishers B.V.
(North-Holland Physics Publishing Division)

ation), are both much larger than the grain size a_o. These quantities are expressible as $\xi_J = (E_J/E_g)^{1/2}a_o/2$ and $\lambda_J = (E_f/2E_J)^{1/2}a_g/\pi$, where $E_f = \phi_o^2/8\pi a_o$ and $\phi_o = hc/2e$. If $V_g = a_o^3$ and $T_c-T \ll T_c$, these expressions reduce to the dirty-limit results (3, 6, 7)

$$\xi_J = (\pi^2\hbar\Delta^2/4e^2k_BT_c\rho_nH_{cg}^2)^{1/2}, \tag{2}$$

$$\lambda_J = (c^2\hbar k_BT_c\rho_n/2\pi^2\Delta^2)^{1/2}, \tag{3}$$

which diverge as $(T_c-T)^{-1/2}$ at T_c. The thin-film critical-current density in this limit reduces to the Ginzburg-Landau depairing current density (8),

$$J_c = cH_{cg}/3\sqrt{6}\pi\lambda_J, \tag{4}$$

such that J_c is proportional to $(T_c-T)^{3/2}$ near T_c.

The depairing current density for $\varepsilon \gg 1$ [Eq. (4)] is the limiting critical-current density only in thin films at low enough applied fields that no vortices are present in the film. For thicker specimens, however, in which self-fields become important, or at higher applied fields, vortices are produced before the depairing current density is reached. In Ginzburg-Landau theory, the mixed state is characterized by the critical fields (9)

$$H_{c1J} \simeq (\phi_o/4\pi\lambda_J^2)[\ln(\lambda_J/\xi_J) + 0.50], \tag{5}$$

$$H_{c2J} = \phi_o/2\pi\xi_J^2, \tag{6}$$

where ξ_J and λ_J are given in Eqs. (2) and (3). The vortex self-energy $(\phi_oH_{c1J}a_o/4\pi)$ corresponding to a segment of length a_o is of order E_J. The critical-current density J_c at which a voltage appears along the specimen corresponds to the threshold for depinning of the vortex array (10). Here J_c depends upon microstructure that locally changes the vortex energy; J_c is maximized when the length scale of the inhomogeneities is well matched to ξ_J, and J_c is very small when the inhomogeneity length scale is either much larger or much smaller (11) than ξ_J.

If $\varepsilon < 1$ at low temperatures, as it is for NbN, the theoretical expression (3) for the thin-film low-field critical-current density J_c versus T exhibits a crossover from Ambegaokar-Baratoff (5) behavior at low temperatures to Ginzburg-Landau (8) behavior close to T_c, where J_c is proportional to $(T_c-T)^{3/2}$. If $\varepsilon \ll 1$ at low temperatures, the crossover does not occur until T is extremely close to T_c. To obtain a rough estimate of ε for the high-temperature superconductors, let us assume $H_{cg} = 10^4$ Oe, a_o (and grain size) = 1 μm, and I_o = 100 μA, such that $J_o = 10^4$ A/cm^2. We then obtain $\varepsilon \simeq 4 \times 10^{-8}$, which indicates that the effects of granularity are very pronounced at low temperatures and that a weakly-coupled-grain model should hold over a wide temperature range, from T = 0 up to very close to T_c.

Implicit in any mean-field theory, such as that of Ref. 3, is the assumption that thermal fluctuations do not destroy phase coherence. This assumption is valid only when $E_J \ll k_BT$. When $E_J \gg k_BT$, a condition that always occurs close to T_c [because $E_J \propto (T_c-T)$], thermally activated phase slippage (12) readily occurs, a time-averaged voltage appears across any junction that carries current, and the sample as a whole becomes resistive, even if all individual grains are still strongly superconducting. The crossover between phase-locked and phase-fluctuation-dominated behavior (4) occurs at a Josephson phase-locking temperature T_{cJ} given roughly by the equation $E_J(T_{cJ}) \simeq k_BT_{cJ}$. More accurate calculations of T_{cJ} have been given by numerous previous authors [see (13) and references therein]. Since the Ambegaokar-Baratoff theory (5) yields

$$E_J/k_B = (6.35 \times 10^7 \text{ K/A})I_o(0)(1-T/T_c) \tag{7}$$

near T_c, we obtain

$$T_c-T_{cJ} \simeq (1.57 \times 10^{-8} \text{ A/K})T_c^2/I_o(0), \tag{8}$$

which yields $T_c-T_{cJ} \simeq 1.4$ K for T_c = 95 K and $I_o(0) \simeq 10^2$ μA.

3. INTERGRANULAR AND INTRAGRANULAR EFFECTS

To compute the magnetic and current-carrying properties of the high-temperature superconductors, we model the granular material as an array of weakly Josephson-coupled, strongly superconducting anisotropic grains. If, however, the intergranular weak links are not insulating barriers but instead proximity junctions, coupled via semiconducting, normal-conducting, or poorly superconducting material, the temperature dependence of the intergranular critical-current density I_o would need to be modified accordingly (13).

At temperatures $T \ll T_{cJ}$, when the grains are phase-locked, application of a small magnetic field to a bulk sample of a high-temperature superconductor will induce screening currents that flow around the outer surface of the sample. The depth to which the applied field penetrates into a grain is λ_g, which for the present we treat as a scalar. Intragranular crystalline anisotropy can be accounted for later by using an anisotropic effective-mass-tensor formulation of the Ginzburg-Landau theory (see Refs. 14-17 and references therein), in which orthorhombic material is characterized by dimensionless effective masses m_a, m_b, and m_c (normalized such that $m_am_bm_c = 1$) along the three principal directions, such that screening currents along these directions decay exponentially with the corresponding penetration depths $\lambda_i = \lambda\sqrt{m_i}$ (i = a, b, c). Because of the weakness of the intergranular coupling, the applied magnetic field penetrates more deeply between grains, to a depth λ_J, along the grain boundaries. We consider in this paper only the case of sufficiently weak coupling that $\lambda_J \gg \lambda_g$. Moreover, to derive a continuum theory, we further assume that $\lambda_J \gg a_o$.

To calculate λ_J, we first need to know how deeply the local magnetic field penetrates into the grains. Let R_g denote a typical grain radius. Note that typically $R_g \simeq a_o/2$ of the 3D Josephson array discussed earlier. When $R_g < \lambda_g$, λ_J is given to good approximation by (3, 7)

$$\lambda_J = (c\phi_o/8\pi^2 a_o J_o)^{1/2}, \tag{9}$$

which reduces to Eq. (3) near T_c.

When $R_g > \lambda_g$, however, the expression for λ_J depends upon the precise shape of the grain and how the intergranular field wraps around the grain. To bring out the essential physics of how to calculate λ_J, we introduce the following simple model. To avoid the complication of demagnetizing factors, we approximate the granular structure of the bulk high-temperature superconductors by representing a semi-infinite sample of the the material ($x > 0$) as a periodic square array (lattice parameter a_o along $\hat{x}$ and $\hat{y}$) of superconducting cylindrical grains of radius R_g aligned along $\hat{z}$, parallel to the applied or self-generated magnetic field H_a. We assume that the intergranular material is nonmagnetic. The intergranular field H varies approximately as $H_a \exp(-x/\lambda_J)$, where x is the distance from the specimen's surface. The magnetic flux density B (the average over a_o^2, the area per grain, of the local flux density b) can be expressed as

$$B = \mu_{eff} H. \tag{10}$$

Here

$$\mu_{eff}(T) = f_n + f_s[1 - P_{cyl}(R_g/\lambda_g)] \tag{11}$$

is the effective temperature-dependent permeability of the granular material (the temperature dependence arising from that of λ_g), where f_n and f_s are the normal (intergranular) and superconducting (intragranular) volume fractions; P_{cyl} is the factor by which magnetic flux penetration suppresses a cylindrical grain's magnetization below that expected for complete Meissner-state flux exclusion (18, 19),

$$P_{cyl}(x) = 1 - 2I_1(x)/xI_0(x), \tag{12}$$

where $I_n(x)$ is the modified Bessel function. Limiting expressions are $P_{cyl} \simeq 1 - 2/x$ when $x >> 1$, $P_{cyl}(1) = 0.107$, and $P_{cyl} \simeq x^2/8$ when $x << 1$. When $\lambda_g << R_g$, we obtain $\mu_{eff} \simeq f_n$, since in this case a negligible amount of flux penetrates into the grains; the lowest order correction in powers of λ_g/R_g yields $\mu_{eff} \simeq f_n + 2f_s\lambda_g/R_g$. When $\lambda_g >> R_g$, we obtain $\mu_{eff} \simeq 1$, since in this case the grains are nearly fully penetrated by magnetic flux; the lowest order correction in powers of R_g/λ_g gives $\mu_{eff} \simeq 1 - (f_s/8)(R_g/\lambda_g)^2$.

Proceeding with the calculation of λ_J, we follow a procedure similar to that used to derive the critical current of a SQUID (two-Josephson-junction interferometer) and integrate the vector potential counterclockwise around a square contour C, whose corners are at the centers of four adjacent grains. Within a grain we use $\mathbf{a} = -(\phi_o/2\pi)\nabla\gamma - (4\pi\lambda_g^2/c)\mathbf{j}$, where γ is the phase of the order parameter. We define the gauge-invariant phase difference across a Josephson junction between grains a and b via

$$\Delta\gamma_{ab} = \gamma_a - \gamma_b - (2\pi/\phi_o) \int_a^b \mathbf{a}\cdot d\mathbf{r} \tag{13}$$

(the integration extending directly across the intergranular tunnel barrier), such that the Josephson supercurrent density from a to b is $J = J_o \sin \Delta\gamma_{ab}$. We neglect the contributions from the line integrals of $\mathbf{j}$ inside the grains. (This can be shown to be a valid approximation when $\lambda_J >> \lambda_g$.) Noting that no supercurrent flows along $\hat{x}$ (so that $\Delta\gamma = 0$ for such junctions), we obtain

$$\Delta\gamma(x+a_o/2) - \Delta\gamma(x-a_o/2) = -2\pi B a_o^2/\phi_o, \tag{14}$$

where $\Delta\gamma$ is the gauge-invariant phase for a junction carrying current along $\hat{y}$. When $\lambda_J >> a_o$, such that $\Delta\gamma(x)$ varies slowly on the scale of a_o, we may reexpress Eq. (14) in terms of $d\gamma/dx$. When $J << J_o$, we have $J \simeq J_o\Delta\gamma$, such that the locally averaged current density (along $\hat{y}$) obeys

$$dJ/dx = -(2\pi a_o J_o \mu_{eff}/\phi_o)H. \tag{15}$$

With the help of Ampere's law, $dH/dx = -(4\pi/c)J$, we thus obtain

$$d^2H/dx^2 = H/\lambda_J^2, \tag{16}$$

where

$$\lambda_J = (c\phi_o/8\pi^2 a_o J_o \mu_{eff})^{1/2}. \tag{17}$$

Equation (17) has a much wider range of validity than its derivation would suggest. Other models for the geometry of the grain and grain boundary change only the expression for $\mu_{eff}(T)$. For example, if we model the grain boundaries as a periodic array of parallel insulating barriers, all parallel to the xz plane, with periodicity a_o and thickness d_i in the y direction, Eqs. (10), (16), and (17) again hold, but with

$$\mu_{eff}(T) = f_n + f_s[1 - P_{slab}(d_s/2\lambda_g)], \tag{18}$$

$$P_{slab}(x) = 1 - x^{-1} \tanh x, \tag{19}$$

where $d_s = a_o - d_i$. When $\lambda_g << d_s$, we obtain $\mu_{eff} \simeq f_n + 2f_s\lambda_g/d_s$ and $a_o\mu_{eff} \simeq d_i + 2\lambda_g$, such that

$$\lambda_J = [c\phi_o/8\pi^2 J_o(d_i+2\lambda_g)]^{1/2}, \tag{20}$$

the familiar expression for the Josephson penetration depth of a long Josephson junction. On the other hand, when $\lambda_g >> d_s$, we obtain $\mu_{eff} \simeq 1$, such that Eq. (17) reduces to Eq. (9).

Returning to our earlier model of the square array of cylindrical grains, we see that for small H_a the intergranular field obeys $H = H_a \exp(-x/\lambda_J)$, where $\lambda_J >> \lambda_g$. When the applied field H_a exceeds H_{c1J}, however, it becomes energetically favorable for intergranular vortices to be present. Here H_{c1J} is the Josephson lower critical field, which can be obtained by calculating $\varepsilon_{1J} = \phi_o H_{c1J}/4\pi$, the energy cost per unit length to produce an isolated Josephson vortex. Such a vortex resembles an intragranular vortex, except that its axis threads through the intergranular regions rather than through a grain. It carries one quantum of magnetic flux $\phi_0 = hc/2e$, which, roughly speaking, is confined to a cylinder of cross-sectional area $\pi\lambda_J^2$, such that the magnetic field energy per unit length is approximately $\phi_0^2/8\pi^2\lambda_J^2\mu_{eff}$. An even more important contribution to the energy per unit length is the Josephson coupling energy stored in the junctions that carry the circulating vortex supercurrents. Accounting only for this energy when $\lambda_J >> a_0$, we have, to logarithmic accuracy,

$$H_{c1J} = (\phi_0/4\pi\lambda_J^2\mu_{eff}) \ln (2\lambda_J/a_0). \tag{21}$$

When the dimensionless parameter $\varepsilon >> 1$ (i.e., $\xi_J >> a_0$, as in granular Al), the intergranular vortex has a core of suppressed order parameter. The argument of the logarithmic term in the expression for H_{c1J} [Eq. (5)] then contains ξ_J, essentially because it appears as the lower cutoff to the integral over the supercurrent kinetic energy density. On the other hand, when $\varepsilon << 1$ (i.e., $\xi_J << a_0$, as in the high-temperature superconductors), an intergranular vortex does not have a core of suppressed order parameter. In place of ξ_J in the logarithmic term in the expression for H_{c1J} [Eq. (21)] is $a_0/2$, the lower cutoff to the integral that approximates the discrete sum over junction coupling energies.

From Eqs. (17) and (21) we obtain the estimates $\lambda_J = 3.0$ μm and $H_{c1J} = 1.1$ Oe, if we assume $I_0 = 100$ μA, $a_0 = 1$ μm, $f_n = 0.3$, and $\mu_{eff} = 0.3$ as typical values for high-temperature superconductors. Application of an applied field H_a somewhat larger than H_{c1J} thus should cause an array of intergranular vortices to penetrate into the granular specimen. The net internal magnetic field can be thought of as a linear superposition of individual intergranular vortex contributions (flux ϕ_0 localized within area $\sim\pi\lambda_J^2$); these contributions strongly overlap at even low magnetic fields. For example, when H = 20 Oe and $\mu_{eff} = 0.3$, we have [Eq. (10)] B = 6 G, which corresponds to an intervortex spacing $(\phi_0/B)^{1/2} \simeq$ 1.9 μm. If $\lambda_J \simeq 3.0$ μm, the resulting intergranular field will be nearly uniform.

How deeply the intergranular vortices penetrate into the sample depends upon the extent to which these vortices are pinned. Pinning can arise from either discreteness of the Josephson-junction array or inhomogeneity of the junction coupling strengths. In planar geometry we thus expect the spatial dependence of H to be given by a critical-state equation

$$dH/dx = \pm(4\pi/c)J_{cJ}, \tag{22}$$

where $J_{cJ}(H,T)$ is the intergranular (transport) critical-current density. Although it is clear that the upper limit to J_{cJ} is the maximum Josephson current density J_0, it is possible that J_{cJ} may be much less than J_0. Numerical calculations of the energy versus position of an isolated vortex in a two-dimensional array of identical Josephson junctions (19) suggest that the critical-current density to depin an isolated vortex is given approximately by $J_c(0,T) \simeq 0.1\ J_0$, where $J_0 = I_0/a_0^2$. With increasing field H, J_{cJ} is further suppressed, both because the mutual repulsion of vortices within λ_J of each other makes it difficult for each vortex to take optimum advantage of the pinning potential (10) and because J_0 is suppressed according to the well-known Frauenhofer-like diffraction pattern (20)

$$J_0(H) = J_0(0)|\sin(\pi\Phi/\phi_0)|/(\pi\Phi/\phi_0), \tag{23}$$

where $\Phi = HA_J$ and A_J is the effective field-penetrated junction area. When J_{cJ} is small, the intergranular field H becomes nearly uniform across the sample. For example, a critical-current density of $J_{cJ} = 10$ A/cm^2 in a granular slab of thickness 2 mm can support a field difference ΔH (from the surface to the center) of only 1.3 Oe. With increasing temperature, J_{cJ} is expected to decrease, chiefly because J_0 decreases. In addition, when T approaches T_{cJ}, intergranular vortices no longer can be permanently pinned, because k_BT can quickly excite vortices from one pinning site over the energy barrier into an adjacent pinning site; consequently J_{cJ} effectively vanishes near and above T_{cJ}. For $T< T_{cJ}$, flux creep with a logarithmic time dependence results when the electric field has the thermally activated form $E \propto \exp(J/J_{th})$, where $J_{th} = 4ek_BT/h\pi a_0^2$.

To discuss the magnetization of a granular superconductor in a parallel applied field H_a, consider a cylindrical sample (radius R), modeled as an array of cylindrical grains (radius R_g). If the sample initially contains no trapped (intergranular or intragranular) vortices, the magnetization (magnetic moment per unit volume of the sample, including possible voids) in a field $H_a < H_{c1J}$ is given by

$$-4\pi M = H_a\{1 - \mu_{eff}[1 - P_{cyl}(R/\lambda_J)]\}, \tag{24}$$

where μ_{eff} and P_{cyl} are defined in Eqs. (11) and (12). When $R >> \lambda_J$, we obtain $-4\pi M \simeq H_a(1 - 2\mu_{eff}\lambda_J/R)$; if $\lambda_J \simeq 3.0$ μm and $R \simeq 1$ mm, for example, we have $-4\pi M \simeq H_a$, corresponding to nearly perfect screening.

When $H_{c1J} < H_a < H_{c1g}$, intergranular vortices enter, and the contribution they make to the magnetization can be calculated with the help of the critical-state equation in cylindrical geometry,

$$dH/dr = \pm(4\pi/c)J_{cJ}. \tag{25}$$

When $\lambda_J << R$, the magnetization is given by

$$-4\pi M = H_a[1 - \mu_{eff}x(1 - x/3)],\ x \le 1, \tag{26a}$$

$$= H_a[1 - \mu_{eff}(1 - 1/3x)],\ x \ge 1, \tag{26b}$$

where $x = cH_a/4\pi J_{cJ}R$. As a function of increasing field H_a or temperature T, $J_{cJ}(H_a,T)$ decreases and x increases. Assuming, for example, that $H_a = 10$ Oe and R = 1 mm, if $J_{cJ} = 8 \times 10^3$ A/cm^2, we obtain $x = 0.01$, such that $-4\pi M \approx H_a$ and the critical-state profile penetrates only to a depth of 0.01 R. If $J_{cJ} = 80$ A/cm^2, we obtain $x = 1$, such that $-4\pi M = H_a(1-2\mu_{eff}/3)$ and the critical-state profile penetrates just to the center of the sample. And if $J_{cJ} = 0.8$ A/cm^2, we obtain $x = 100$, such that $-4\pi M \approx H_a(1-\mu_{eff})$ and the intergranular field H is nearly uniform across the cross section. (When $x \geq 1$, the field at the center differs from the applied field by $\Delta H = H_a/x$.)

To examine the effect of intragranular vortices upon the magnetization, let us initially assume that the intergranular field is nearly uniform ($\Delta H << H_a$). If the grains initially contain no trapped vortices, then in the Meissner state ($H_a < H_{c1g}$) the magnetization (defined via $-4\pi M = H_a - \langle B \rangle$, the volume average including the intergranular material and any voids) is [Eqs. (10)-(12) or (26b) with $x \to \infty$]

$$-4\pi M = H_a f_s P_{cyl}(R_g/\lambda_g). \tag{27}$$

If $\lambda_g(0) \approx 0.1$ µm and $R_g \approx 1$ µm, the magnetization suppression factor has the value $P_{cyl} \approx 0.8$ at low temperatures and decreases monotonically with increasing temperature. With the help of the dirty-limit result (21)

$$[\lambda_g(0)/\lambda_g(T)]^2 = F(T), \tag{28}$$

where F(T) is given in Eq. (1), we obtain $\lambda_g(T) \approx \lambda_0'(1-T/T_c)^{-1/2}$ near T_c, where $\lambda_0' = 0.613\ \lambda_g(0)$, such that

$$-4\pi M \approx (H_a/8) f_s (R_g/\lambda_0')^2 (1-T/T_c) \tag{29}$$

near T_c when $R_g/\lambda_g << 1$ and $H_a < H_{c1g}$.

For applied fields in the range $H_{c1g} < H_a < H_{c2g}$ there is no general analytic expression for the magnetization. Denoting by M_0 the magnetization for an infinite superconductor, we expect the magnetization corrected for sample-size effects to be given by

$$-4\pi M = -4\pi M_0 f_s P_{cyl}(R_g/\lambda_{eff}), \tag{30}$$

where λ_{eff} is the field- and temperature-dependent effective penetration depth (22), well approximated for high-κ (κ = Ginzburg-Landau parameter) superconductors by $\lambda_{eff}(B,T) = \lambda_g(T)(1-B/B_{c2g})^{-1/2}$. In the absence of pinning, the maximum value of $-4\pi M$ in equilibrium occurs at $H_a = H_{c1g}$ and is then given by Eq. (27). Near T_c we may use the Abrikosov result (23), $-4\pi M_0 = (H_{c2g}-H_a)/\beta_A(2\kappa_g^2-1)$, where $\beta_A = 1.16$. (We ignore here the effects of normal-state paramagnetism.) Close to H_{c2g}, where $B \approx H_a$ and $R_g/\lambda_{eff} << 1$, we obtain for $\kappa_g >> 1$

$$-4\pi M = f_s R_g^2 (H_{c2g}-H_a)^2/16\beta_A \kappa_g^2 H_{c2g} \lambda_g^2(T). \tag{31}$$

For fixed H_a and varying temperature, it is more useful to express Eq. (31) as

$$-4\pi M = A(T_{c2g}-T)^2, \tag{32}$$

where

$$A = f_s[(-dH_{c2g}/dT)/16\beta_A \kappa_g^2 T_c](R_g/\lambda_0')^2 \tag{33}$$

and $T_{c2g} = T_c + H_a/(dH_{c2g}/dT)$ is the temperature at which $H_a = H_{c2g}$. Different assumptions about the grain shape yield results similar to Eqs. (29)-(33) but with different numerical prefactors (18, 24).

Equations (27)-(33) give expressions for the contributions of the grains to the reversible, equilibrium magnetization of the sample. Flux pinning, which permits excess current density J_{cg} to flow about the axis of a cylindrical grain in a clockwise or a counterclockwise sense, depending upon whether the applied field is increasing (↑) or decreasing (↓), leads to magnetization hysteresis, $\Delta M = M_\uparrow - M_\downarrow$. Evaluation of the magnetic moments (18) yields, for the intragranular contribution to ΔM,

$$-4\pi\Delta M = f_s(8\pi/3c)J_{cg}R_g. \tag{34}$$

A similar derivation, including the effect of intergranular vortices, in which critical-state field profiles are assumed to fully penetrate to the centers of the sample (radius R) and the grains (radius R_g), yields

$$-4\pi\Delta M = (8\pi/3c)J_{cJ}R + f_s(8\pi/3c)J_{cg}R_g. \tag{35}$$

Since magnetization hysteresis can be produced by intragranular flux motion even if $J_{cJ} = 0$, this expression shows why measurements of the magnetization hysteresis yield only an upper limit to the transport critical-current density J_{cJ}.

Measurements of the complex permeability $\tilde{\mu} = \mu' + i\mu''$ (or the complex susceptibility $\tilde{\chi} = \chi' + i\chi''$, where $\tilde{\mu} = 1 + 4\pi\tilde{\chi}$) of a cylindrical sample in a parallel field $H_a(t) = H_0 + h_0 \cos \omega t$ give important information about J_{cJ} and J_{cg}. Experiments (25) have shown that μ'' versus T can have two peaks, one near T_c associated with J_{cg} and one at lower temperature associated with J_{cJ}. The important dimensionless quantities (26) are $x_J = ch_0/4\pi J_{cJ}(H_0,T)R$ and $x_g = ch_0/4\pi J_{cg}(H_0,T)R_g$. If $x_g \sim 1$ and $x_J >> 1$ near T_c, the intergranular field H is nearly uniform, and the losses are dominated by intragranular flux motion. Then μ'' versus T has a maximum at $x_g = 1$, such that $\mu''_{max} = 0.212\ f_s$ if $\lambda_g << R_g$ and $H_0 >> H_{c1g}$. The height of the peak, however, is strongly suppressed, as in Eq. (30), when $\lambda_g \geq R_g$. The value of J_{cg} at the temperature where the peak occurs can be determined from $J_{cg} = ch_0/4\pi R_g$. At low temperatures, J_{cg} increases and x_g becomes very small. [If $h_0 = 1$ Oe, $R_g = 1$ µm, and $J_{cg} = 10^5$ A/cm^2, for example, $x_g = 0.08$.] In this case, losses due to flux penetration into the grains become negligible, $B \approx f_n H$, and a maximum in μ'' versus T occurs at $x_J = 1$, where $\mu''_{max} = 0.212\ f_n$ if $\lambda_J << R$. The value of J_{cJ} at the temperature where the peak occurs can be determined from $J_{cJ} = ch_0/4\pi R$. Complex permeability measurements done at a variety of different h_0, H_0, and T thus can be used to determine the functional dependence of $J_{cg}(H_0,T)$ and $J_{cJ}(H_0,T)$.

Measurements of the zero-field-cooled magnetization consist in first cooling the sample to a very low temperature in zero magnetic field, applying a small measuring field, and then monitoring the magnetization with increasing temperature. Such measurements, when performed with very small H_a (well below 1 Oe), probe the intergranular properties without complications from intragranular flux motion. A given $H_a < H_{c1J}(0)$ intersects the curves of $H_{c1J}(T)$, $H_{c1g}(T)$, and $H_{c2g}(T)$ at the corresponding temperatures T_{c1J}, T_{c1g}, and T_{c2g}. Initially, when $T < T_{c1J}$, no vortices are present in the sample, and the magnetization is given by Eq. (24); when $\lambda_J << R$, we have $-4\pi M \approx H_a$. When $T_{c1J} < T < T_{c1g}$, intergranular vortices (but no intragranular vortices) enter the specimen; when $\lambda_J << R$, the magnetization is given by Eqs. (26). At first, x is very small and $-4\pi M \approx H_a$. As the temperature increases, x increases, and $-4\pi M$ decreases. Finally, when $T \approx T_{cJ}$, as occurs in the experiments of Ref. 27, J_{cJ} vanishes, $x \to \infty$, and H becomes nearly uniform across the cross section, such that $-4\pi M \approx H_a(1-\mu_{eff})$ [see Eqs. (10)-(12)]. [When H_a = 0.1 Oe and H_{c1g} > 100 Oe, the intragranular vortex state ($T_{c1g} < T < T_{c2g}$) is confined to within 0.1 K of T_c.]

Field-cooled magnetization experiments consist in applying a small measuring field H_a above T_c, then monitoring the magnetization with decreasing temperature. The intragranular flux density $\langle b\rangle_{tr}$ trapped as intragranular vortices depends upon the value of the dimensionless quantity $\gamma_{c1g} = 4\pi J_{cg}(H_a,T_{c1g})R_g/cH_a$. When $\gamma_{c1g} << 1$, we have $\langle b\rangle_{tr} << H_a$; i.e., nearly all intragranular vortices are expelled. (This condition is met in the experiments of Ref. 27.) Whenever $\gamma_{c1g} >> 1$, we obtain $\langle b\rangle_{tr} \approx H_a$. Similarly, the intergranular flux density H_{tr} trapped as intergranular vortices depends upon $\gamma_{c1J} = 4\pi J_{cJ}(H_a,T_{c1J})R/cH_a$. When $\gamma_{c1J} >> 1$, we have $H_{tr} \approx H_a$; i.e., nearly all intergranular vortices are trapped. (This condition is met in Ref. 27). Only when $\gamma_{c1J} <<$ 1 would we have $H_{tr} << H_a$. The experiments of Ref. 27 thus yield $-4\pi M = H_a(1-\mu_{eff})$ for T decreasing from T_{c1g} to zero. Ref. 27 describes measurements of the remanent moment versus increasing temperature after the applied field then was turned off at low temperature. The critical-state model can be used to calculate this moment, expressed in a form similar to Eq. (26). As the temperature increases, $x = cH_a/4\pi J_{cJ}R$ increases, and the trapped flux decreases. About half the trapped flux is gone when $x \sim 1$, and all of it disappears when $T = T_{cJ}$, when $J_{cJ} \to 0$, and $x \to \infty$.

4. CONCLUSIONS

Most of the electromagnetic properties of bulk high-temperature superconductors can be understood by modeling the material as an array of weakly coupled, strongly superconducting anisotropic grains.

REFERENCES

(1) J.G. Bednorz and K.A. Müller, Z. Phys. B. 64 (1986) 189.
(2) G. Deutscher, Y. Imry, and L. Gunther, Phys. Rev. B10 (1974) 4598.
(3) J.R. Clem, B. Bumble, S.I. Raider, W.J. Gallagher, and Y.C. Shih, Phys. Rev. B35 (1987).
(4) B.G. Orr, J.R. Clem, H.M. Jaeger, and A.M. Goldman, Phys. Rev. B34 (1986) 3491.
(5) V. Ambegaokar and A. Baratoff, Phys. Rev. Lett. 10 (1963) 486; erratum 11 (1963) 104.
(6) L.P. Gor'kov, Zh. Eksperim. i Teor. Fiz. 36 (1959) 1918; 37 (1959) 1407 [Soviet Phys. JETP 9 (1959) 1364; 10 (1960) 998].
(7) B. Giovannini and L. Weiss, Solid State Commun. 27 (1978) 1005.
(8) M. Tinkham, Introduction to Superconductivity (McGraw-Hill, New York, 1975), p. 117.
(9) C.R. Hu, Phys. Rev. B6 (1972) 1756.
(10) A.M. Campbell and J.E. Evetts, Adv. Phys. 21 (1972) 199.
(11) J. W. Ekin, Phys. Rev. B12 (1975) 2676.
(12) V. Ambegaokar and B.I. Halperin, Phys. Rev. Lett. 22 (1969) 1364.
(13) W.Y. Shih, C. Ebner, and D. Stroud, Phys. Rev. B30 (1984) 134.
(14) R.A. Klemm and J.R. Clem, Phys. Rev. B21 (1980) 1868.
(15) V.G. Kogan, Phys. Rev. B24 (1981) 1572.
(16) V.G. Kogan and J.R. Clem, Phys. Rev. B24 (1981) 2497.
(17) V.G. Kogan and J.R. Clem, Jpn. J. Appl. Phys. 26 (Suppl. 26-3) (1987) 1159.
(18) J.R. Clem and V.G. Kogan, Jpn. J. Appl. Phys. 26 (Suppl. 26-3) (1987) 1161.
(19) C.J. Lobb, D.W. Abraham, and M. Tinkham, Phys. Rev. B27 (1983) 150.
(20) Ref. 8, p. 199.
(21) A.A. Abrikosov, L.P. Gor'kov, and I.E. Dzyaloshinski, Methods of Quantum Field Theory in Statistical Physics (Dover, New York, 1963).
(22) J.R. Clem, Phenomenological Theory of the Local Magnetic Field in Type-II Superconductors, in: Low Temperature Physics--LT14, Vol. 2, eds. M. Krusius and M. Vuorio (North-Holland, Amsterdam, 1975), p. 285-288.
(23) A. A. Abrikosov, Zh. Eksp. Teor. Fiz. 32 (1957) 1442 [Sov. Phys. JETP 5 (1957) 1174].
(24) D.K. Finnemore, R.N. Shelton, J.R. Clem, R.W. McCallum, H.C. Ku, R.E. McCarley, S.C. Chen, P. Klavins, and V.G. Kogan, Phys. Rev. B35 (1987) 5319.
(25) B. Renker, I. Apfelstedt, H. Küpfer, C. Politis, H. Rietschel, W. Schauer, H. Wühl, U. Gottwick, H. Kneissel, U. Rauchschwalbe, H. Spille, and F. Steglich, Z. Phys. B 67 (1987) 1.
(26) J.R. Clem, "AC Losses in Type-II Superconductors," Tech. Report No. IS-M 280, Ames Laboratory, Ames, Iowa.
(27) D.C. Cronemeyer, A.P. Malozemoff, and T.R. McGuire, in: Proceedings of the 1987 Fall Meeting of the Materials Research Society, in press.