Short-coherence-length superconductors

by Guy Deutscher

The new high-T_c oxides present some anomalous electromagnetic properties, such as low critical current densities, a reversible behavior of the magnetization at fields much lower than H_{c2}, and internal Josephson effects, that distinguish them from the conventional low-T_c metals and alloys. These anomalous properties were first observed in bulk-sintered samples and were often ascribed to the poor connectivity of these ceramics. More recently, a qualitatively similar behavior has been observed in single crystals and oriented films. The fundamental role of the short coherence length in determining the behavior of the high-T_c oxides is discussed. We show that the short coherence lengths at the local depressions of the order parameter at crystallographic defects lead to reduced critical currents and cause glassy behavior in the vicinity of T_c.

Introduction

It is now established that the new high-T_c oxides have by and large BCS-like superconducting properties: a clear jump in the heat capacity at T_c whose magnitude is that predicted by BCS [1]; a gap in the density of states at least equal to the BCS value [2]; an upper critical field consistent with type-II behavior [3]; and a critical current density that approaches the de-pairing limit in high-quality thin films at low temperatures [4]. As the sample quality has improved, peculiar features such as a linear term in the heat capacity at low temperatures and a weak cusp in the heat capacity at T_c have given way to more conventional behavior. The known electronic normal-state properties—electrical conductivity, Hall effect, optical reflectivity, heat capacity—may also be explained in terms of a strongly anisotropic Fermi surface and a small Fermi energy [5], although we still lack definitive experiments establishing the existence of a Fermi surface in the high-T_c oxides.

In spite of the above, the new superconductors do present some puzzling properties that are not readily explained by textbook superconductivity. There is growing evidence that the behavior of the heat capacity near T_c is not strictly mean-field, but presents at least the beginning of a critical divergence [6], as had previously been suggested [7]. The critical current density, which is very low in bulk ceramics even in zero fields and at low temperatures, is also quite depressed in single crystals under applied fields well below H_{c2} (particularly at high temperatures) [8]. In a related way, the diamagnetic susceptibility χ is irreversible below a characteristic temperature $T^*(H)$: for $T < T^*(H)$, $|\chi_{FC}| < |\chi_{ZFC}|$, where FC and ZFC stand, respectively, for field-cooled and zero-field-cooled measurements. This is observed both in sintered ceramics [9] and in single crystals [10]. In both cases, $[T_c - T^*(H)] \propto H^{2/3}$, a result that has been interpreted as a de-Almeida–Thouless line indicative of glassy behavior [9], or as reflecting giant flux creep [10]. The temperature dependence of H_{c2} is in general nonlinear near T_c, contrary to the predictions of the Ginzburg–Landau theory; instead, $H_{c2} \propto (T_c - T)^n$, with $n \approx 1.5$ [11]. On the level of local properties, it has been shown that "clean" grain boundaries (i.e., free of second-phase material) act as weak Josephson junctions [12]; and there is strong evidence for the existence of intragrain junctions in a number of magnetization experiments on powders, particularly on powders originating from one single crystal [13].

Although it is not exhaustive, the above list is sufficient to show the breadth of the anomalous properties of the oxides. It has been proposed by Deutscher and Müller [14] that their origin lies in the extremely short coherence length of the new superconductors. These authors showed in particular how planar defects such as stacking faults, off-oxygen stoichiometry regions, twin boundaries (and, of course, *a fortiori* grain boundaries) should behave as Josephson junctions with a depressed order parameter when the coherence length is of the order of interatomic distances. These junctions, in turn, constitute a network of weak links that can result in glassy behavior in the appropriate range of temperatures (near T_c) and applied fields ($H^*_{c1} \ll H \ll H_{c2}$, where H^*_{c1} is an effective first penetration field for the junction). In a related way, the intergrain or intragrain boundaries are also the site of enhanced flux creep, as discussed below.

This paper is organized as follows. In the first section we point out the relationship that exists between the width of the critical region and the maximum available pinning energy; in the following section we review experimental determinations of the coherence length; we then turn to a detailed discussion of the critical current across boundaries and its relation to the coherence length. We conclude with some remarks concerning the relation between the short coherence length and glassy behavior.

Critical behavior and pinning energy

The superconducting transition is the only known second-order phase transition that can be described by mean-field Landau theory, i.e., where critical thermodynamic fluctuations occur over such a narrow range of temperatures around T_c that they are undetectable. The origin of this ideal mean-field behavior lies in the very large number of Cooper pairs that are found within the correlation radius of one pair—any pair sees the mean field of many other pairs. This is contrary to the case of, say, ferromagnets, where a spin is primarily sensitive to the orientation of its nearest neighbors, so that the mean-field approximation is not sufficient.

The width of the critical region can be estimated from the relation

$$\Delta F(\epsilon_c) \cdot \xi^3(\epsilon_c) \simeq k_B T_c, \qquad (1)$$

where ϵ_c is the width of the critical region on the reduced temperature scale $\epsilon = (T_c - T)/T_c$; ΔF is the difference in free energy per unit volume between the disordered and the ordered phases; and ξ is the temperature-dependent coherence length. Equation (1) determines the temperature range $|\epsilon| < \epsilon_c$, where the thermodynamic fluctuations of the order parameter are comparable to its equilibrium value, or larger. The temperature dependences of ΔF and ξ are of course different in the critical ($|\epsilon| < \epsilon_c$) and mean-field ($|\epsilon| > \epsilon_c$) regions. They are universal only in the mean-field region, $\Delta F \propto \epsilon^2$ and $\xi(\epsilon) \propto \epsilon^{-1/2}$, and the condensation energy per coherence volume $U(\epsilon) = \Delta F(\epsilon)\xi^3(\epsilon)$ goes to zero at T_c as $\epsilon^{1/2}$. As we approach T_c, we always reach a temperature where $U(\epsilon) < kT_c$, the fluctuations become large, and the mean-field approximation breaks down [in particular, the temperature dependences $U(\epsilon)$ and $\xi(\epsilon)$ are modified]. Reciprocally, as long as the temperature dependences of these and other measurable quantities remain mean-field, we must be outside the critical region.

In a superconductor, $\Delta F = H_c^2/8\pi$, where H_c is the thermodynamic critical field. According to the above procedure, we can calculate ϵ_c if we know $H_c(0)$ and $\xi(0)$. Unfortunately, these values are not very well known. H_c can in principle be determined by integrating under the magnetization curve, but accurate measurements of $M(H)$ at high fields are difficult and not yet available. The coherence length can be determined from

$$H_{c2} = \frac{\phi_0}{2\pi\xi^2(T)}, \qquad (2)$$

but an accurate measurement of H_{c2} has also proved difficult (see below). Using $H_c(0) = 12\,000$ Oe and $\xi(0) = 12$ Å, one obtains ϵ_c of order unity [7]. But since $(\epsilon_c)^{-1}$ varies as the fourth power of $H_c(0)$ and the sixth power of $\xi(0)$, uncertainties on the values of these quantities bear heavily on the value of ϵ_c. What is clear is that the width of the critical region in the oxides must in any case be much larger than in conventional superconductors, for which we calculate $\epsilon_c \sim 10^{-10}$. This difference comes primarily from the much smaller value of ξ in the oxides.

A further complication in computing the width of the critical region comes from the strongly anisotropic nature of the oxides. Equation (1) assumes basically that they are 3D superconductors—the anisotropy can then simply be taken care of by replacing ξ^3 with $\xi^2_{a,G} \cdot \xi_c$. Below T_c, the oxides are certainly 3D superconductors: The anisotropy of the critical current density does not exceed one order of magnitude. Well above T_c, there is experimental evidence that the excess conductivity due to the fluctuations of the order parameter follows

$$\Delta\sigma \propto (T_c - T)^{-1}, \qquad (3)$$

indicating 2D behavior [15]. The validity of Equation (1) requires that near the superconducting transition the behavior becomes critical when the fluctuations are 3D.

Experimentally, it appears that the critical behavior sets in within 1 K of T_c, or less. This is the temperature range within which $\Delta\sigma$ departs from the mean-field behavior, Equation (3) [15]. It is also the temperature range where some groups have observed a non-mean-field behavior of the heat capacity [6]. Below T_c, mean-field behavior of the London penetration depth has also been reported in the entire investigated temperature range, up to about 0.5 K of T_c [16]. All of these experimental findings converge toward $\epsilon_c \lesssim 0.01$. Following Equation (1), we then obtain

$$U(0) \gtrsim 0.1 \text{ eV}. \quad (4)$$

This result is of considerable practical importance, because U is the characteristic energy available for vortex core pinning, and as such is one of the fundamental parameters that set the upper limit to the critical current density in the presence of a magnetic field [17].

We wish to stress here that it is much safer to obtain U from an experimental determination of the width of the critical region than it is to calculate it from poorly known values of $H_c(0)$ and $\xi(0)$. We also note that $U(0) \simeq 10k_BT_c$; this is less favorable than in conventional superconductors [where $U(0) \gtrsim 100k_BT_c$], yet sufficient to allow high critical current densities at $T < 0.8T_c$.

Experimental determination of the coherence length

Early experimental determinations of the coherence length were based on measurements of H_{c2} either by dc resistivity or by ac susceptibility. Typically, the point where half of the normal state is restored was selected as the upper critical field, although it was noted [18] that the width of the transitions was anomalously large compared to that observed in conventional type-II superconductors. Measurements on single crystals revealed the anisotropy of H_{c2}. Using the effective mass approximation, the values $\xi_{a,G} = 34$ Å and $\xi_c = 7$ Å were obtained [8].

However, Müller et al. [9] had early on shown in bulk ceramics a reversible magnetic behavior at temperatures $T < T^*(H)$, as mentioned in the Introduction. No magnetic flux is trapped in the reversible regime, which at a given temperature covers fields much smaller than H_{c2}. In this range, the Bean critical current must therefore be negligibly small. This observation immediately explains the anomalously broad resistive transitions, although the link between the two sets of experimental observations was not immediately noticed. The same remark applies to single crystals, in which a reversible regime and broad transitions are also seen. A determination of H_{c2} as, say, the midpoint of the resistive transition may thus be an underestimate. A better estimate might be the field at which most of the normal-state resistance—say 90%—is restored. One then gets shorter values for the coherence length: $\xi_{a,G} \simeq 12$ Å, $\xi \simeq 1.5$ Å [11, 19].

Could ξ possibly be even shorter than these values? Magnetization as well as heat capacity measurements under applied fields indicate anomalous free-energy surfaces [20]. The reported magnetization curves $M(H)$ are quite unconventional, M staying practically constant at high fields within experimental accuracy. (dH_{c2}/dT) at T_c would then be of the order of 10 T/K or more, giving $\xi_{a,G}(0) \simeq 7$ Å. An analysis of the resistive transition under applied fields gives good agreement with experiment without assuming any shift of $T_c(H)$ [21]. These recent results confirm an earlier analysis of critical current measurements concluding that (dH_{c2}/dT) is extremely steep at T_c [22].

Thus, the experimental situation concerning the determination of H_{c2} is quite unsatisfactory, but it certainly indicates a very short coherence length, $\xi_{a,G} \lesssim 10$ Å; ξ_c may be one order of magnitude smaller.

These very small values rule out the retardation effects that play an essential role in reducing the Coulomb repulsion in the conventional electron–phonon-mediated superconductivity. Roughly speaking, $\xi_{a,G}$ is of the order of the distance between holes in the CuO planes. Any proposed mechanism for high-temperature superconductivity in the oxides must be consistent with this observation.

Critical current across boundaries

Because of the very short coherence length, the value of the superconducting order parameter Δ is much more sensitive to crystallographic defects in the oxides than it is in conventional long-coherence-length superconductors. In principle, local values of Δ can be either enhanced or depressed, but the highest-T_c oxides appear to be definite compounds, with any substitution leading to a depression of T_c. Thus, in general we expect Δ to be depressed at crystallographic defects—point defects, dislocations, planar defects. For the case of point defects and dislocations, this depression provides a mechanism for vortex pinning. In the case of planar defects, however, pinning is only effective for vortex lines in the plane of the defect and current parallel to it. For the general case where the current has a component perpendicular to the planar defect, we argue that the depressed order parameter results in a lower critical current density. This is because the de-pairing current across the defect is reduced, and also because the locally reduced condensation energy U lowers the pinning energy available for vortices moving in the plane of the defect:

$$U = (1/2)N_i(0)\Delta_i^2\xi^3, \quad (5)$$

where Δ_i is the value of the order parameter at the site of the defect, which we consider as a boundary between two well-crystallized regions or "grains."

Equation (5) rests upon the assumption that the spatial extension of the crystallographic defect is of the order of ξ. This is the main difference between the oxides and the conventional superconductors, for which ξ is much larger than interatomic distances.

In general, the calculation of U_i requires a detailed knowledge of the local electronic structure and a self-consistent solution for the order parameter. This is a difficult problem, which has only been touched upon recently [23], and for which a general solution is not yet available.

For the case of a superconductor–insulator interface, Δ_i can be calculated in a proximity-effect formulation [14, 18]. It is a function of the parameter $\delta = b/[2\xi(T)]$, where the

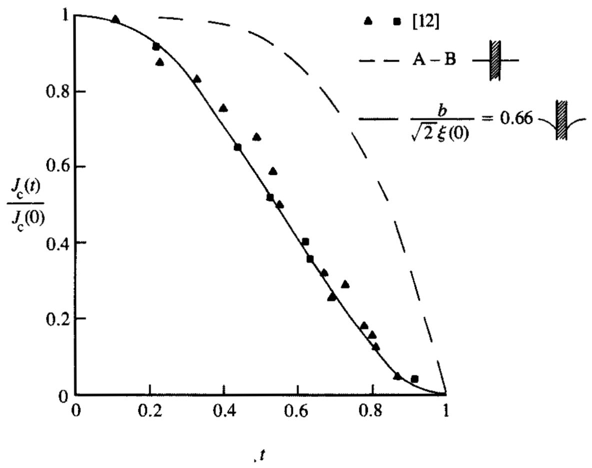

Figure 1

Normalized critical current density as a function of the reduced temperature $t = (T/T_c)$. The discontinuous line represents the Ambegaokar–Baratoff (A–B) theory, with a zero temperature gap value of $2kT_c$. The data points are from Mannhart et al., as presented in [12]. The continuous curve is calculated for a Josephson junction with a depressed order parameter according to the model developed in this paper. It leads to a critical current that is particularly reduced at high temperatures, in agreement with the experiment. The continuous line is calculated for the measured T_c and the same bulk gap as the A–B line.

extrapolation length b is of the order of $[\xi^2(0)/a]$, and a is an interatomic distance.

It appears that this formulation is quite adequate for describing the critical current across a grain boundary, as measured in the experiments reported by P. Chaudhari et al. [24]. Assuming that such a boundary is a Josephson junction characterized by a normal-state resistance R_N and a depressed order parameter Δ_i, we compute the critical current density J_c using the Ambegaokar–Baratoff expression

$$J_c(T) = [\pi\Delta_i(T)/2eR_N]\tanh[\Delta_i(T)/2kT], \tag{6}$$

where $\Delta_i(T)$ is determined from the boundary condition

$$\left|\frac{1}{\Delta}\left(\frac{d\Delta_i}{dx}\right)_i\right| = \frac{1}{b}. \tag{7}$$

Assuming that the bulk order parameter $\Delta(0) = 2kT_c$, we compute $\Delta_i(T)$ from Equation (7) solving the nonlinear Landau–Ginzburg equation, and obtain numerically

$$\frac{J_c(T)}{J_c(0)} = \frac{\Delta_i(T)}{\Delta_i(0)}\tanh\left[\frac{\Delta_i(T)}{\Delta(0)}\frac{T_c}{T}\right] \tag{8}$$

for different values of $\delta(0)$. The temperature dependence of $\Delta(T)$ is that tabulated by Mühlschlegel [25], and the temperature dependence of $\xi(T)$ is that for a clean superconductor [26].

For $\delta(0) = 2/3$, corresponding to $\Delta_i(0)/\Delta(0) = 0.5$, a good fit is obtained over the entire temperature range with the data of Chaudhari et al. [24], as shown in **Figure 1**. The fit is noticeably better than the ones they propose, either with the A–B expression with $\Delta(0) = 5$ meV ($\simeq 0.6kT_c$), or with the Likharev S-N-S model with T_c (junction) = $0.9T_c$ (grain). Our fit uses both the BCS gap and the T_c of the measured grain, and neatly reproduces the high-temperature tail. As noted previously [14], this tail reflects the fact that, near T_c, $J_c \propto \Delta_i^2 \propto [b/\xi(T)]^2 \propto (T_c - T)$, or, in qualitative terms, that the local depression of Δ is strongest near T_c. This is because b is temperature-independent, while ξ diverges at T_c.

The quality of the fit is similar for 5° and 15° boundaries, for which the depression parameter is thus the same. We learn from the analysis of this experiment that a minor crystallographic perturbation such as a 5° boundary parallel to the c axis is sufficient to depress the order parameter and J_c by a factor of 2 at low temperatures. The further decrease in the critical current density at increased boundary angle must be due to a reduced transmission coefficient. For large boundary angles, the total reduction in J_c (about a factor of 50) is dominated by the transmission coefficient at low temperatures; at high temperatures it is dominated by the local depression of Δ, as shown in Figure 1.

The dispersion of the experimental data does not allow a very accurate determination of $\delta(0)$. With the de Gennes expression $b = [\xi^2(0)/a]$, the value of $\xi(0)$ used for the fit corresponds to $\xi(0) = a$. This expression for the extrapolation length strictly applies only when $\xi(0) \gg a$, but it is gratifying to see that the junctions $J_c(T)$ correspond to a coherence length that is indeed very short and of the order of a. The study of the critical current of grain boundaries does allow a determination of $\xi(0)$ that, although not very accurate, is free of the problems that plague its determination from H_{c2} measurements, as discussed above.

The study of boundaries nearly perpendicular to the c axis would be very valuable, since it would give us a determination of ξ_c. If indeed $\xi_c/\xi_{a,G} \sim 0.1$, we expect a further reduction of $J_c(0)$ by about a factor of 10. This would put the upper limit of $J_c(0)$ in a polycrystalline sample at about 20 times less than the de-pairing limit, the loss factor being about 1000 at $t = 0.9$.

The depression of Δ at boundaries also reduces the height of the pinning barriers for vortex motion along the boundary (see **Figure 2**). With $[\Delta_i(0)/\Delta(0)] = 0.5$ we get, from Equations (4) and (5), $U_i \simeq 0.025$ eV. This is an upper limit, since $N(0)$ is certainly also reduced at and near the boundary. This pinning energy is of the order of that measured by Yeshurun et al. in single crystals of YBCO [27].

Conclusions

Studies of the upper critical field and of grain-boundary junction behavior in the high-T_c oxides point to a very short coherence length, which is not much larger than the lattice spacing in the (a, b) plane and smaller along the c axis. The critical current density across grain boundaries can be analyzed in terms of a locally depressed order parameter. For boundaries parallel to the c axis, the reduction factor appears to be about a factor of 2 at low temperatures. There, the Josephson critical current is mostly limited by the normal-state resistance of the junction, but at high temperatures the depression effect dominates. In the presence of a magnetic field parallel to the boundary, and of a current perpendicular to it, it reduces the available vortex pinning energy. We propose that the giant flux creep observed in the high-T_c oxides is due to vortex motion along boundaries: grain boundaries in polycrystalline samples, and internal boundaries (planar defects) in single crystals.

This interpretation is different from that of other authors who have attributed the large flux creep effects to an intrinsically very low bulk pinning energy [27]. According to the experimental width of the critical region $\epsilon_i < 0.01$, we have argued that this bulk pinning energy cannot be smaller than 0.1 eV, while the experimentally determined pinning energy is 0.01 eV in YBaCuO and 0.001 eV in BiSrCaCuO single crystals [27]. Our point of view is that these values are extrinsic rather than intrinsic, and closely related to the planar defect structure of these single crystals.

Generally speaking, the short ξ naturally produces a very broad range of pinning energies corresponding to the many different kinds of defects to be found in the oxides, with varying degrees of depression of the order parameter. It is basically this broad range of pinning energies that is responsible for glassy behavior in ceramics as well as in single crystals, as reported by C. Rossel et al. [28] and I. Morgenstern [29].

A very short coherence length is the distinctive feature of the high-T_c oxides, and probably of any high-T_c superconductor. A self-consistent solution for the order parameter in the oxides is an important and open problem. Understanding and control of their defect structure are of great importance for their practical applications.

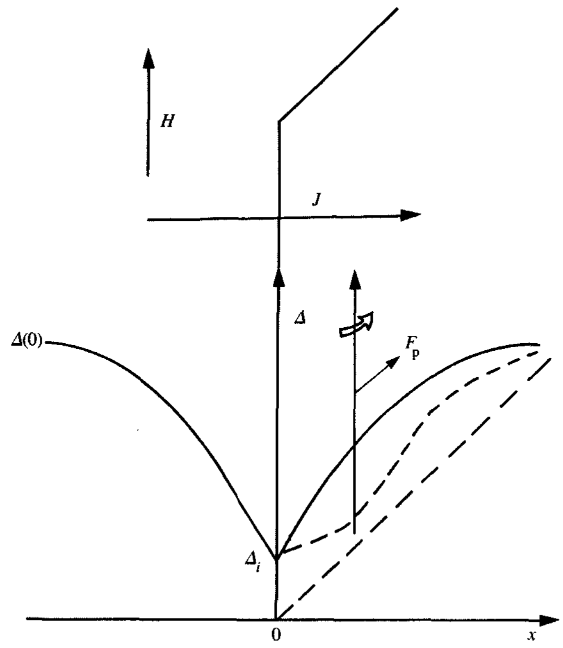

Figure 2

Schematic representation of a vortex line in the plane of a junction with a depressed order parameter, with a current J crossing the junction. The locally reduced condensation energy lowers the pinning barriers against motion of the vortex in the plane of the junction.

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