Origin of Superconductive Glassy State and Extrinsic Critical Currents in High-T_c Oxides

G. Deutscher
Department of Physics and Astronomy, Tel Aviv University, Ramat Aviv, 69978 Tel Aviv, Israel

and

K. A. Müller
IBM Research, Zurich Research Laboratory, CH-8803 Rüschlikon, Switzerland

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The short coherence length of high-T_c oxides is shown to induce considerable weakening of the pair potential at surfaces and interfaces. It is argued that this effect is responsible for the existence of internal Josephson junctions at twin boundaries, which are at the origin of the superconductive glassy state, as well as for gapless tunneling characteristics.

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Recent susceptibility and magnetization measurements[1] as well as microwave absorption experiments[2] have been interpreted as resulting from a superconducting glass state. This state consists of superconducting loops with areas typically smaller than the grain size, and thus it cannot be explained by weak links at grain boundaries. The existence of Josephson junctions *inside* the grains had therefore to be assumed.

Analysis of high-T_c superconductors[3,4] indicate that they have extremely short coherence lengths, of the order of the size of the unit cell. Bardeen[3] quotes for the zero-temperature coherence lengths $\xi(0)$ a value of 12 Å in $Y_1Ba_2Cu_3O_7$, while values of 7 and 34 Å along the c and (a,b) axis, respectively, have been inferred from H_{c2} measurements on single crystals.[4] These are to be compared with the values of the lattice parameters $a=3.83$ Å, $b=3.89$ Å, and $c=11.71$ Å.[4] We show here that the existence of a very short coherence length is responsible for a lowering of the pair potential at surfaces and interfaces, this effect being particularly strong near T_c. It also leads to the appearance of internal Josephson junctions responsible for the glassy behavior observed in ceramic samples[1,2,5] as well as in single crystals,[4] to the microwave response of point contacts,[6] and to extrinsic (low) critical currents in single crystals[7] that are otherwise unexplained. We believe that it is also responsible for the gaplesslike characteristics obtained by scanning tunneling spectroscopy.[8]

The boundary condition for the pair potential Δ at a boundary can be written as

$$(\Delta^{-1} d\Delta/dx)_{x=0} = b^{-1}, \tag{1}$$

where the "extrapolation length" b (Fig. 1) is given by[9]

$$\frac{1}{b} = \frac{2}{L}\int_{-\infty}^{+\infty} dx\, \frac{\Delta(x)}{\Delta_0}\left[1 - \frac{N(x)}{N(0)}\right], \tag{2}$$

where $L \simeq \xi^2_{(0)}$, Δ_0 and $N(0)$ are the bulk pair potential and normal-state density of states, respectively. At a superconductor-insulator boundary, the integrand in Eq. (2) is significantly different from zero only for x of the order of the lattice spacing a [for $x \gg a$, $\Delta(x)=0$; for $x \ll -a$, $N(x)=N(0)$]. Then, $b \approx \xi^2(0)/a$.

Inside the superconductor, the pair potential is given by the solution of the nonlinear Landau-Ginzburg equation

$$\Delta(x) = \Delta_0 \tanh[(-x+x_0)/\sqrt{2}\xi(T)], \tag{3}$$

where the value of x_0 is to be determined by the boundary condition (1) and the value of b. This then fixes the value of the pair potential at the boundary, $\Delta(0)$. There are two limiting cases: (i) if $b/\xi(T) \gg 1$, $\Delta(0)/\Delta_0 \simeq 1$ and (ii) if $b/\xi(T) \ll 1$, $\Delta(0)/\Delta_0 \simeq b/\xi(T)$.

Because of the divergence of $\xi(T)$, case (ii) is, in principle, always realized close enough to T_c. However, in usual superconductors $\xi(0)/a \gg 1$ [$\xi(0)/a \approx 10^3$], and that range of temperature is exceedingly small $\{(T_c - T)/T_c \approx [a/\xi(0)]^2 \approx 10^{-6}\}$. In practice, (i) applies at all temperatures and we can use the boundary condition $d\Delta/dx = 0$, $\Delta(0) = \Delta_0$.

Such, however, is not the case for the new high-T_c oxides, for which, as discussed above, $\xi(0) \approx a$ and there-

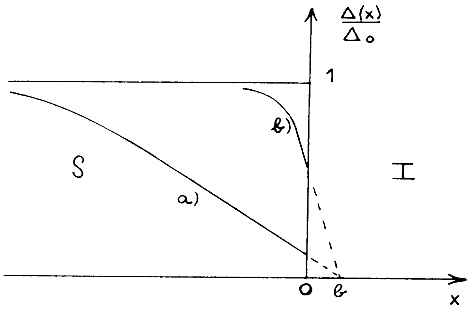

FIG. 1. Profile of the pair potential in a short-coherence-length superconductor near a superconductor-insulator boundary: (curve a) $T \lesssim T_c$; (curve b) $T \ll T_c$.

fore $b \simeq \xi(0)$. Case (ii) is then realized over a broad range of temperatures. This is particularly marked when the boundary is perpendicular to the c axis, since along that direction $\xi(0)$ appears to be even smaller than the lattice parameter. In that case, we actually expect $\Delta(0)/\Delta_0 \ll 1$ over much of the temperature range.

This result has important implications both for quasiparticle tunneling experiments [superconductor–insulator–normal-metal (SIN) junctions] and Josephson effects [superconductor-insulator-superconductor (SIS) junctions]. In tunneling experiments, the strongly reduced value of $\Delta(0)$ will result in a gapless tunneling characteristic, as observed by several authors (see Ref. 8). In Josephson junctions, the critical current will have near T_c an anomalous temperature dependence $J_c \propto (T_c - T)^2$. This is because the critical current is proportional to the product of the pair potential on both sides of the junction, $J_c \propto \Delta_+(0)\Delta_-(0) \propto \Delta_0^2[b/\xi(T)]^2$, instead of the usual result $J_c \propto \Delta_0^2 \propto (T_c - T)$ (near T_c). However, at low temperatures $b/\xi(T) \simeq b/\xi_0 \simeq 1$ and much of the strength of the pair potential at the interface is restored.

In the high-T_c oxides, Josephson junctions may well occur at defects such as twin boundaries inside the grains (as well as at grain boundaries). Observation of a high density of twin boundaries has been reported by several authors.[10] These include twins along the (110) and (001) planes. Intertwin distances of the order of 1000 Å have been observed. This high density is thought to occur as a result of the tetragonal-to-orthorhombic transformation. During this transformation, a given tetragonal grain does not usually seem to transform into a single orthorhombic domain. Rather, the orthorhombic phase may nucleate into different parts of the grain, with the two possible (**a**,**b**) orientations, resulting eventually in the observed twin network. Twin boundaries are essentially regions of the tetragonal phase, about one unit cell thick, that have not been transformed into the orthorhombic phase. Brokman[11] proposes, for instance, that at a (110) twin boundary, the four oxygen atoms around a Cu atom in the Cu-O plane form a square (instead of a rectangle in the orthorhombic phase), with the Cu atom slightly off center. Although the exact structure of the different twin boundaries remains to be studied, we propose that they should be considered essentially as nonorthorhombic and/or nonsuperconducting regions. Furthermore, defect planes due to oxygen overstochiometry and understochiometry have been observed recently.[12] We note that amongst the many types of planar defects observed, some may be metallic and some insulating. In a usual superconductor with a large $\xi(0)$, such thin nonsuperconducting regions would not noticeably affect the superconducting state. However, in the case of the oxides under consideration, the situation is quite different because of the very small value of $\xi(0)$. Insulating regions will result in SIS junctions with *a depressed order parameter at the interface*, hence $j_c \propto (T_c - T)^2$ near T_c as discussed above. [In principle, normal quasiparticle tunneling (modified by the depressed order parameter) may also take place in thick (> 20 Å) insulating regions, but it remains to be seen whether such regions exist.] Normal-metallic regions will result in *proximity-effect* SNS junctions, in which j_c is known[9] to have that same temperature dependence. Basically, the similarity in behavior between SIS and SNS junctions in high-T_c oxides is due to the fact that *at high temperatures* the decay lengths of the superconducting wave function in insulating and in normal-metallic regions are both of the order of $\xi(0)$. The junctions should be particularly weak for (001) twins, in view of the extremely small value of $\xi(0)$ along the c axis. We note that looking down the (110) planes, we have a two-dimensional network of (110) and (001) twins separating the two sorts of domains, which should be of roughly equal total cross sections. We actually do have then a percolation situation, at the (two-dimensional) percolation threshold. A current flowing in any direction in the described planes must then necessarily cross twin boundaries and the inferred Josephson junctions surrounding the domains, or "cluster" to use the percolation description.

A model for such a superconducting glass consisting of many weakly coupled clusters has been investigated with the Hamiltonian[1,13]

$$\beta\mathcal{H} = -\sum J_{ij}\cos(\phi_i - \phi_j - A_{ij}). \tag{4}$$

Here J_{ij} is the Josephson coupling constant between clusters. The phase factors $A_{ij} = \kappa_{ij}H$ introduce randomness and frustration in presence of a magnetic field, H, because the system has many competing ground states of almost the same energy. κ_{ij} is a random geometric factor. After cooling in zero magnetic field, $H=0$, the system is in a Meissner or $-xy$ state. On application of a magnetic field, the system becomes weakly random past a *geometric* critical field,

$$H_{c1}^* = \phi_0/2S,$$

where ϕ_0 is the flux quantum and S the projected area of the superconducting loops with uniform phase in the glass state.

The area S of uniform phase has been estimated originally from the reduction of magnetization M as a function of applied H to be of the order of 0.03 μm^2 *smaller* than the average grain size of 10 μm^2 in the $La_{2-x}Ba_xO_{4-y}$ *ceramics.* The size S could recently also be determined from the maximum in the derivation of microwave absorption $\partial\chi''/\partial H$ by Blazey *et al.*[2] S ranges from 0.5 to 5 μm^2 in either $La_{2-x}Sr_xO_{4-y}$ or $Y_1Ba_2Cu_3O_7$ ceramics. Comparing these sizes with that of the grains obtained from electron transmission microscopy, one arrives again at the conclusion that often the coupling is *inside* the grains. This conclusion is confirmed by the observation of *Josephson I-V characteristics* with normal-conducting Au, Cu, Al metal contacts observed by Estève *et al.*[6] These authors were able to measure $S = 0.1\ \mu\text{m}^2$ from the microwave-induced $I_n(V)$ steps as a function of B-field dependence of Josephson junctions critical current. S obtained from this experiment is in remarkable agreement with the values deduced from the other two experiments.

Additional evidence for the occurrence of intragrain Josephson junctions comes from the recently measured time decay of the magnetization, both in ceramic materials[1,5,14] and in single crystals.[7] In *both* cases, a logarithmic time dependence is observed, indicating glassy behavior.

Finally, we believe that recent single-crystal critical-current measurements[7] also support the internal-junction model. The values of J_c are obtained from magnetic hysteresis loops taken with the field perpendicular $(H_\perp)$ or parallel $(H_\parallel)$ to the Cu-O plane. For the $H_\perp$ case, screening currents flow in the Cu-O planes, and hence do not have to cross (001) twin boundaries. But in the $H_\parallel$ orientation, screening currents must cross both (001) and (110) twin boundaries. In addition, the penetration of vortices in the (001) twins further reduces the strength of these junctions. This difference between the two orientations is directly reflected *in the high-temperature data* shown in Fig. 3 of Ref. 7, which displays a very strong anisotropy of J_c *even at moderate fields*. At 40 K, for instance, the anisotropy is of the order of 300 in a field of 10 kG. It becomes even stronger closer to T_c. We submit that this high-temperature anisotropy is *extrinsic,* i.e., due to the existence of twin boundaries and the associated Josephson junctions, rather than to crystalline anisotropy. At low temperature, as discussed above, the pair potential recovers a substantial fraction of its bulk value at the interface of the twin and the measured anisotropy is most intrinsic (and therefore field independent, as observed).

In conclusion, we have argued that twin boundaries induce *intragrain* Josephson junctions in high-T_c oxides, as a result of their very short superconducting coherence length. These junctions form a network that divides grains into weakly coupled superconducting domains.

We feel that this argument provides a natural explanation for many previously not understood experimental observations, such as the occurrence of a glassy state on a scale smaller than the grain size, microwave-induced steps at NS point contacts, and anomalously large critical-current anisotropy in single crystals at high temperatures and moderate fields. These intragrain junctions—probably mainly those associated with (001) twins—may be of great practical importance, as they considerably reduce the average current density in randomly oriented ceramics. Elimination of these twin boundaries may be a valuable goal towards practical application of the new superconductors.

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