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## Dissipation in a weak-link-limited superconductor as a problem of percolation theory

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Broadening of the resistive transition of sintered  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  superconductor has been studied varying the measuring current over the range of five orders of magnitude. The observed well-defined branching points in the resistive transition coincide with the quasi-Ohmic saturation of  $V$ - $I$  curves and scale with the average grain size. The results suggest the validity of a simple percolation model for dissipation in systems whose global superconductivity is realized by Josephson coupling of the superconducting grains. A common origin of structured resistive transition, observed in many superconducting systems, has been also discussed.

Several different mechanisms may cause the broadening of the resistive transition of a superconductor and may be grouped into three classes of effects: thermodynamic fluctuations,<sup>1</sup> thermally activated flux creep<sup>2</sup> or phase slip,<sup>3</sup> and quantum tunneling.<sup>4</sup> There are numerous reports dealing with the role of the first two classes in the broadening of the resistive transition of classical and high-temperature superconductors. The observation of the last effect, quantum tunneling, is highly improbable at high temperature (around  $T_c$ ), but seems to be important at low temperatures.<sup>5,6</sup>

In this paper we report on the resistive broadening induced by effects that may be classified, because of historical reasons,<sup>7</sup> as "sample inhomogeneity type." In particular, we show for superconductors whose behavior is dominated by weak links, such as the bulk (sintered) high-temperature superconductors, that resistive broadening can be understood as a problem of percolation theory.

Sintered (ceramic) high-temperature superconductors (SHTS's) offer a unique opportunity for studying the consequences of the intrinsically small coherence length on the global superconductivity of the sample. This is at variance with the classical granular superconductors<sup>8</sup> whose effective (Pippard) coherence length is reduced by the presence of the grain boundaries. The superconducting state in SHTS's is depressed at the boundary, and the phase coherence of the neighboring grains is established by the Josephson tunneling across weak links formed at the grain boundaries. High sensitivity of the resistive transition ("foot" structure) to small magnetic fields<sup>9</sup> can be ascribed to effects of a magnetic field on Josephson tunneling. However, the interpretation is not straightforward, as many dissipative processes depend on magnetic field and may lead to similar behavior. The study of the resistive broadening as a function of the applied current (instead of the applied magnetic field) is potentially simpler for interpretation of the underlying dissipation.

We present here a study of the resistive transition region of the sintered  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  samples as a function of the measuring current, varied in the range of five orders of magnitude ( $50 \mu\text{A}$ – $5 \text{ A}$ ). The samples were synthesized following a standard procedure for the solid-state reaction of the corresponding powders.<sup>10</sup> The measurements consisted of the recording simultaneously the current and voltage (up to the chosen current level) during a short (1 ms) single-shot, half-sinusoidal current pulse while the temperature was slowly (1 K/h) increasing. Owing to the short pulse duration and the low-resistance contacts, we were able to avoid problems connected to the sample self-heating. A typical result is displayed in Fig. 1. For the resistance data, we have used

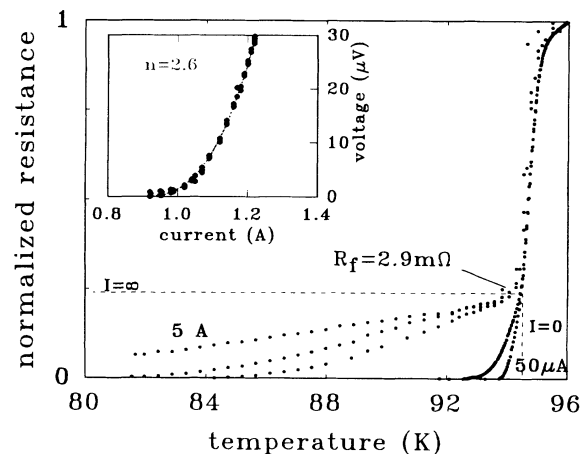


FIG. 1. Resistive transitions of a fine-grained  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  sample, by the use of measuring current in the interval  $50 \mu\text{A}$ – $5 \text{ A}$ . Branching point ( $R_f$ ) coincides with quasi-Ohmic saturation of  $dV/dI$  characteristics. The dashed lines define the region of nonlinear  $V$ - $I$  characteristics. Inset: initial dissipation and fit (dashed line) to power law. Exponent  $n$  does not depend much on temperature, magnetic field, or microstructure.

the conventional “ $V$  divided by  $I$ ” values extracted from  $V$ - $I$  curves. It can be clearly seen that the position of the point that divides the Ohmic (upper part, no broadening) and the non-Ohmic regions (lower part, foot) of the transition is very sharply defined. However, it is important to clarify the meaning of the points in the non-Ohmic part of Fig. 1. Each of these points is associated with a point in the nonlinear  $V$ - $I$  characteristic at the same temperature. More specifically, the ordinate of a resistance point is equal to the slope of the line connecting the associated point in the  $V$ - $I$  diagram with its origin. This slope has obviously no physical significance, and the term “resistance” introduced above is an ill-defined quantity. So the points in the foot region of Fig. 1 merely define the non-linear dissipative region of  $R$ - $T$  diagram and we (unlike the related study of resistive broadening in  $n$ -type granular superconductors<sup>11</sup>) are not going to use the concept of resistance in order to understand the temperature dependence of a position of the points in  $R$ - $T$  diagrams. Rather, we stress the geometrical features of  $R$ - $T$  diagrams (Figs. 1 and 2) as their main physical ingredients. These are the well-defined branching point and boundary lines of the non-Ohmic region. The lines correspond to the obvious saturation of the experimental curves in the limiting cases of measuring current ( $I=0$  and  $I=\infty$ , respectively). The other equivalent representation of the non-Ohmic region are the  $V$ - $I$  diagrams at fixed temperatures, which explicitly show that in this region non-Ohmicity consists of both  $V$ - $I$  nonlinearity and the presence of a critical current. As is well known<sup>12</sup> for SHTS's, the level of the quasi-Ohmic saturation ( $R_f$ ) in the high-current limit of  $dV/dI$  curves, interpreted as an effective Ohmic resistance of the weak-link network, does not depend much on temperature or magnetic field (if it is smaller than a few tesla). The experiments we report on in this paper clearly showed that the value of  $R_f$  is always equal to the resistance of the  $R$ - $T$  branching point. The leveling-off tendency of  $R$ - $T$  curves due to the increasing current (to the temperature-independent  $R$ - $T$  characteristic, i.e., boundary line  $I=\infty$ ) can be now simply understood as a consequence of the existence of a unique, well-defined, and weakly-temperature-dependent Ohmic resistance of the sample,  $R_f$ . (A weak temperature dependence corresponds to the observed temperature-independent resistance of a single grain boundary.<sup>13</sup>) In the other words, the boundary conditions for  $R(T)$  curves are given by the two expressions

$$R(T)|_{I=\text{const}} < \left. \frac{dV}{dI} \right|_{I=\text{const}} \quad \text{and} \quad \lim_{I \rightarrow \infty} R(T) = R_f,$$

while their specific shapes are determined by the temperature dependence of the critical current.<sup>14</sup> Thus the role of  $R_f$  is similar to the geometrical constraints in many types of finite-size effects in transport properties of condensed matter. In our case, however, besides the trivial dependence on the normal-state resistance,  $R_f$  is determined by the microstructural features of the sample such as the average grain size  $\langle a \rangle$ . Namely, as the volume fraction of the grain boundaries (with respect to the total volume occupied by grains themselves) depends sensitive-

ly<sup>10,15</sup> on  $\langle a \rangle$ , the number of nodes of the weak-link network is expected to be determined by  $\langle a \rangle$  as well. This is exactly what has been observed in the series of measurements on  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  differing in  $\langle a \rangle$ , as shown in Fig. 2. The obvious increase of  $\langle a \rangle$ , achieved by the variation of sintering times from 5 to 500 h and keeping all other parameters of the synthesis strictly fixed, is accompanied by the gradual decrease of the transition branching point, thus the decrease of  $R_f$ .

These observations challenge our understanding of the effective dissipative processes involved in the dissipative

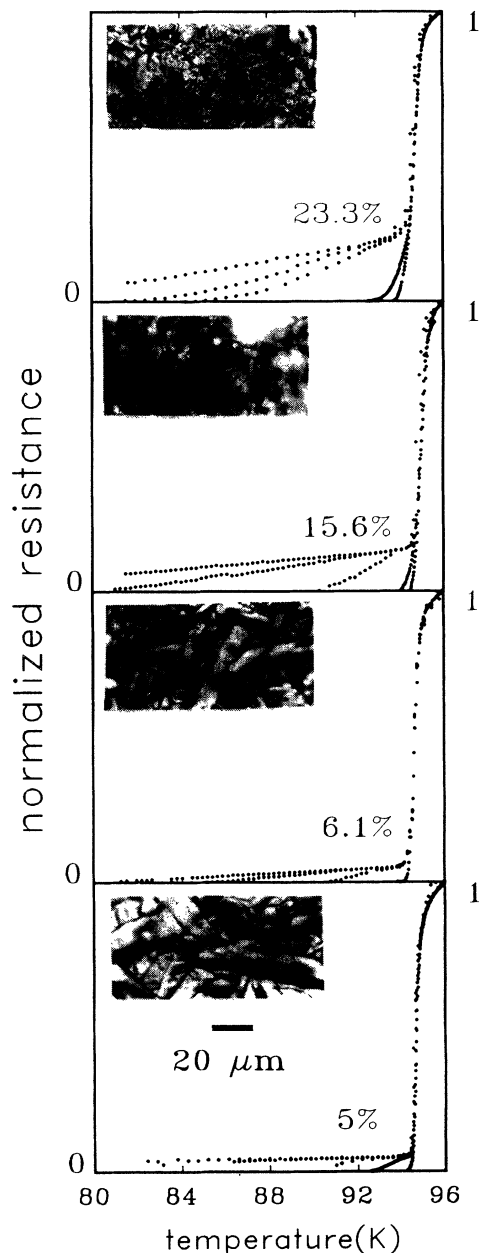


FIG. 2. Resistive transitions of four  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-x}$  samples differing in average grain sizes. The numbers near the branching points designate their positions with respect to the resistance at 96 K. The corresponding microstructures are shown in the insets, with the common length scale in the bottom one.

range of  $V$ - $I$  characteristics of SHTS's (limited from below by the onset of dissipation at the critical current  $I_c$  and from above by the Ohmicity at  $R_f$ ). The usual attempt to introduce for this purpose the flux-pinning scenario of type-II superconductors contradicts many experimental facts. For example, the low-field hysteresis in magnetization studies, attributed to the supracurrent loops of the weak-link network, displays virtually no time (or frequency) effects.<sup>16</sup> If the flux pinning were underlain, the absence of time effects would correspond to unreasonably high pinning potential. Instead of invoking pinning in the mixed state, the penetration of the magnetic flux can be more appropriately described as the formation of the Meissner (i.e., time independent) state of Josephson network,<sup>17</sup> with its large and magnetic-field-dependent penetration depth. Accordingly, we understand the results of  $V$ - $I$  studies [the absence of flux-creep ( $V \propto e^I$ ) behavior or flux-flow ( $R_f \propto H/H_{c2}$ ) behavior for SHTS's], as the experimental arguments for basically flux-pinning-free dissipation in the network of weak links.

We claim that our observations support the latter scenario for dissipation. We assume that both the global zero-resistance and the dissipative state of SHTS's are governed by the excitations in the weak-link network. The basic element of this network, a single-grain-boundary Josephson junction, can be either nondissipative (*open*) or dissipative (*closed*), depending on the position of the actual working point in the  $V$ - $I$  diagram of this particular junction. The binary status of the junction is controlled by the local current, i.e., by the ratio of the local current density and the magnetic-field-dependent Josephson critical current. We note also that the magnetic flux possibly trapped inside a grain controls the status of the neighboring junction on a long-time scale, which may lead, as shown recently,<sup>18</sup> to the logarithmic time decay of the measured dissipation. Thus experimental  $V$ - $I$  curves of SHTS's monitor, in our view, a continuous reduction of the fraction of the *open* elements.

We propose in this paper that the global organization of dissipation is governed by percolation. A well-known problem of percolation theory is the connectivity in two-phase systems, such as superconductor-normal-metal or superconductor-insulator composites.<sup>19</sup> In these composites the conductivity diverges<sup>19</sup> following a characteristic power law at the percolation threshold concentration of the superconducting component. This approach has been already used to study penetration depth divergence near  $T_c$ ,<sup>20</sup> connectivity problem of SHTS-silver composites,<sup>21</sup> and thin films.<sup>22</sup> For the purpose of the interpretation of our results, we extend the concept of percolation from the classical "compositional" problems to the current-controlled properties of the weak-link network in  $V$ - $I$  measurements of nominally well-connected SHTS's. As the externally controlled current determines the fraction of the *open* elements in the network, the critical current corresponds to the percolation threshold concentration of these elements. In this sense our approach is related to the random fuse model<sup>23</sup> or the dynamic random resistor network model,<sup>24</sup> developed for the studies of  $V$ - $I$  nonlinearities of metal-insulator composites.

There are two experimental arguments supporting the

percolation model introduced above. The first one is a well-defined, microstructurally dependent value of Ohmicity  $R_f$ : An increase of the total current above  $I_c$  is accompanied by a further reduction of the number of the *open* links, until the natural limit (given by the total number of links) is achieved and one measures  $R_f$  as a sample differential resistance. A similar saturation characterizes  $V$ - $I$  curves of numerical nonlinear networks.<sup>24</sup> The second argument concerns the functional form of the very onset of the dissipation. In this range of  $V$ - $I$  curves, the best fit can be achieved by the power-law form<sup>25-27</sup>  $V \propto (I - I_c)^n$ . We found that the value of the exponent  $n$  of a large number of samples<sup>25</sup> is in the range of 2-3, similar to the result for polycrystalline Bi-Sr-Ca-Cu-O films.<sup>26</sup> A fit to the power law of the actual sample is shown in the inset to Fig. 1. The constancy of  $n$  values suggests that it could have some universal background.<sup>27</sup> We claim that the associated differential resistance ( $dV/dI$ ) exponent (i.e.,  $n - 1$ ) could be compared with the conductivity exponent  $t$  or the superconducting (or dielectric) breakdown<sup>19</sup> exponent  $s$ . It should be noted that these exponents are originally related to the compositional percolation problems. In the case of  $V$ - $I$  curves, the variable of the problem is the current instead of the concentration of one component. These two variables are not linearly related in our obviously nonlinear problem, but in a small range of current the linearity certainly holds. The power-law exponent of a large number of  $dV/dI$  measurements is, at average, 1.5, not far from the range of the expected values for the conductivity exponent  $t$  in three dimensions<sup>28</sup> (1.6-2). It can be argued that the comparison with the breakdown exponent  $s$  would be a more relevant one. In two dimensions the exponents  $t$  and  $s$  are, however, equal, following the duality argument.<sup>29</sup> In higher dimensions the duality argument does not work and  $s$  is typically 0.8, far from our experimental value. On the other side, the choice of the relevant exponent depends on the way of approaching the critical point. It seems to us that the conditions of  $V$ - $I$  measurement on SHTS's (considering a continuous path through a critical point to the dissipative region) correspond better to the conductivity route, with  $t$  as an effective exponent and in accordance with our experimental exponent value.

The specific structure of the resistive "foot" in SHTS's, is, as described above, related to percolation. However, the very existence of the foot feature is a consequence of the crossover of the resistance-measuring point in the  $V$ - $I$  diagram (with  $T$  as a parameter) from the linear to the nonlinear  $V$ - $I$  characteristics in the course of the cooling run. We believe that the same type of crossover is also the common origin of the foot structures observed in numerous examples of resistive transition studies of superconductors.<sup>6,7,30,31</sup> These structures were attributed to the sample inhomogeneity,<sup>7</sup> contact effects,<sup>30</sup> or quantum tunneling.<sup>31</sup> In our opinion a more relevant fact is that  $V$ - $I$  nonlinearity may set in during the cooling cycle, leading to a new feature in the  $R(T)$  dependence. An example of  $V$ - $I$  nonlinearity onset is the appearance of the critical current (as imposed by the voltage resolution of the experiment) in  $V$ - $I$  characteristic at lower tempera-

ture. A more subtle type of nonlinearity onset may come from the specific  $V$ - $I$  characteristic composed of Ohmic and non-Ohmic regions. In case of phase-slip-related experiments,<sup>30</sup> the latter regions are separated by the temperature-dependent characteristic current  $I_0$  and the nonlinearity onset is expected during the temperature decrease. We note that the studies of the resistive transition of granular metallic films<sup>6,7</sup> should be more related to our study of SHTS's.  $V$ - $I$  nonlinearity may appear in these measurements due to flux-creep (or quantum tunneling)

effects. However, in the light of results of this work, we do not exclude an onset of the nonlinear, weak-link-related dissipation organized as a global percolation. To this extent the important (if not decisive) argument would be the presence of the branching point in  $R(T)$  dependence or, even better, full knowledge of  $V$ - $I$  characteristics at different temperatures.

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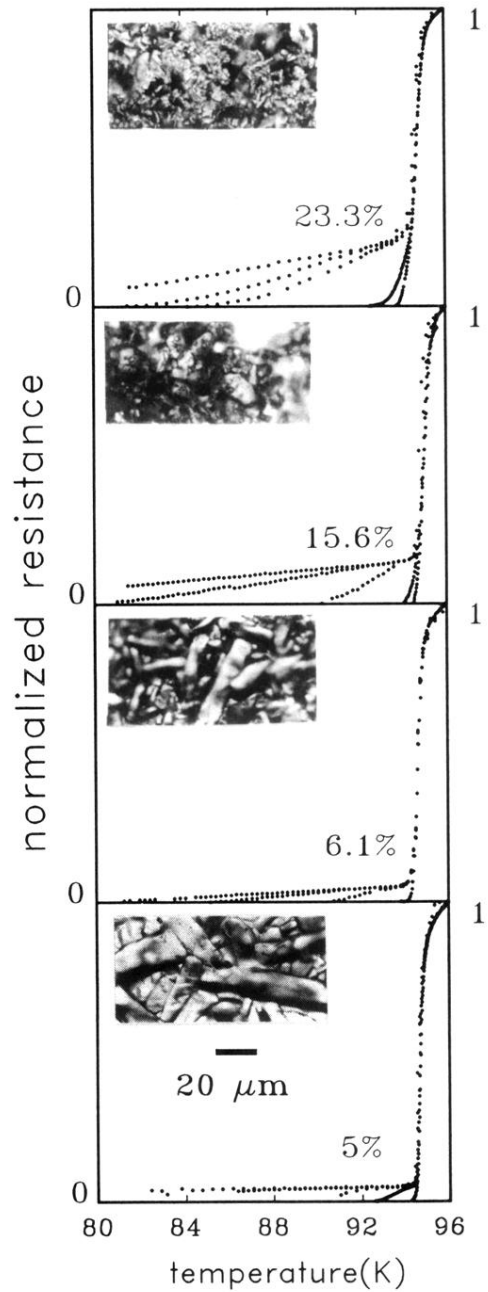


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