Weak links as a subsystem that monitors the intragranular flux creep in high-T_c superconductors

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The time dependence of the dc voltage that remains on a sintered $YBa_2Cu_3O_{7-x}$ sample after the application of a magnetic field has been systematically studied in various magnetic fields and at three different temperatures. The obtained logarithmic time dependence has been related to the intragranular flux creep assuming that the dissipative part of the sample, the subsystem of weak links, is controlled by local magnetic fields. A one-dimensional model for the dissipation has been introduced and solved numerically, showing that the inverse of the relaxation rate approaches the intragranular pinning potential in the limit of high applied fields. The experimental values of appropriately defined pinning potentials increases with temperature.

The time dependence of the magnetization of a superconductor is related to its flux-pinning properties. The unusually fast time dependence in high-temperature superconductors[1] (HTSC) has been a subject of considerable interest.[2–8] As reported by many authors, the observed logarithmic time dependence is a consequence of the thermally assisted flux creep, similar to the Anderson-Kim relaxation in hard type-II superconductors.[9] Alternatively, the presence of disorder of various types in HTSC and the observations of non-logarithmic time dependencies justify the studies of the analogy with spin-glass systems.[1,10]

Complementary to the studies of magnetization, the measurements of relaxation of microwave[11] or usual dc (Refs. 12 and 13) dissipation as well as of magnetic nonlinearities,[14] all on polycrystalline samples, reveal also the logarithmic time dependence. However, these types of measurements challenge not only our insight into the nature of the logarithmic relaxation but also our understanding of the specific dissipative mechanisms in the sinterates. As regards their transport and magnetic properties, the sinterates are composed of two distinct subsystems (grain-boundary weak links and more compact grain interiors) and a measured property can be usually classified either as an inter or an intragrain phenomenon. The most famous example is the double-peak structure of the imaginary part of ac susceptibility.

In this paper, we present our measurements of the time decay of the dissipative dc voltage of the sintered $YBa_2Cu_3O_{7-x}$ (Y-Ba-Cu-O) samples in a simple I-V arrangement, at various temperatures and as a function of the applied steplike magnetic fields. We have adopted the Anderson flux creep as the basic process behind the long-time dependence of the dc voltage and developed a numerical model that facilitates the analysis of the measurements. The apparent pinning potential was found to increase with temperature and saturate in increasing magnetic fields. A clue to the explanation of our experimental data is provided by our understanding of the effective role of the subsystem of weak links in our measurements: A weak link network serves as a sensor for the detection of the intragranular flux creep. The experimental argument for this standpoint comes from the study of nonlinear magnetic ac response (third-harmonic component) by Shaulov *et al.*[14] This nonlinear signal is attributed to weak links subjected to local magnetic fields of the grains in the mixed state.

The rectangular sintered samples (preparative details are described in Ref. 15) were mounted in a conventional four-probe arrangement. The biasing dc current was supplied by a highly stable current source. In the course of the measurements the sample was immersed in different cryogenic baths (nitrogen, argon, or oxygen) in order to ensure virtually constant temperature and eliminate the thermal instabilities due to heating at the contacts. The samples were first zero-field-cooled from a temperature far above T_c to the measuring temperature. A constant current (approximately equal to the critical current) was switched on and subsequently the dc magnetic field was applied to the sample space. The application of the magnetic field was accompanied by an onset of a voltage (dissipation) along the sample and the time evolution of this voltage was carefully monitored upon the removal of the field.

The voltage taken in a typical measurement is shown in Fig. 1 for an Y-Ba-Cu-O sample at 80 K. There are generally three distinct regimes in the voltage response. First, a sharp drop immediately after switching off the field, a long-time dependence ($t > 5$ s) and an intermediate regime (10 ms $< t < 5$ s). The "drop" is actually an exponential with a 2 ms time constant. It could be argued that this decay simply reproduces the inductive smearing of the turning-off edge of the magnetic field. That this is not the case is proved by the sharp edge of the current in the magnetic coil circuit. The time constant of the decay does not depend either on temperature (in the interval 60–87 K) or the applied magnetic field ($H_a < 500$ Oe). The long-time dependence was studied by taking the voltage recorded at $t = 5$ s ($= V_0$) as the first point of the relaxation. The obviously slow time decay (Fig. 1) can be nicely fitted (inset to Fig. 1) to the logarithmic dependence of Anderson's type[9,16]

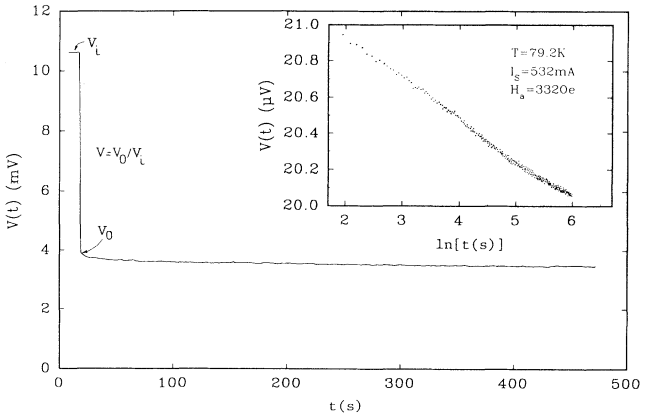

FIG. 1. Typical time dependence of the remnant voltage on Y-Ba-Cu-O sample biased with dc current, after the application of external magnetic field H_a. Inset: Voltage vs ln (time). Voltage recorded at $t=5$ s (V_0) was chosen as the first point of the relaxation.

$$V(t)=V_0[1-A\ln(t)] \tag{1}$$

at all three temperatures where the measurements have been performed (68, 80, and 87 K). We note however that the quality of the fit increases with the magnitude of the applied magnetic field H_a. At lower values of H_a the voltage response has a noisy component possibly connected with a rather erratic fluxoid motion. The results for the intermediate regime, where the time dependence is neither exponential nor logarithmic, will be reported elsewhere. Coming back to the long-time regime, we note that there is a threshold magnetic field H_t for the observation of any remnant voltage: The applied field lower than H_t gives only the exponential decay to zero voltage. The actual value of H_t is the sample-dependent and determined by other factors (sample geometry, biasing current, temperature) but its lower limit is about 100 Oe (i.e., higher than the lower critical field H_{c1} of the compound). In this sense our field conditions are more in the "strong"-field limit (majority of grains in the mixed state) compared to the "weak"-field limit of the experiment in Ref. 17. For this reason we think that the model of flux creep of the Josephson medium proposed in that work:[17] cannot be applied to our experiment.

We have found that there are two experimental quantities that are relevant for the interpretation of the logarithmic voltage dependence: normalized drop voltage v ($v=V_0/V_i$, V_i is the initial voltage before switching of the field, Fig. 2) and the slope A of the logarithmic relaxation (represented in Fig. 3 by its inverse $U^*=kT/A$). For the applied fields above H_t both of these quantities depend strongly on H_a, up to the high-field (500 Oe) saturation (Figs. 2 and 3). The tendency of interpreting the quantity U^* as an intragranular flux-pinning potential,[11–13] although supported by reasonable experimental U^* values, should be analyzed with great care. The reason for this is, besides an unexpectedly strong-field dependence of U^*, that the dissipation that sets in above the critical current I_c is rather unique in sintered samples:[18,19] Even for high values of biasing current ($I \gg I_c$) and/or external magnetic fields, no clear flux-creep or flux-flow dominated dissipative regime can be identified. The simplest experimental argument for the latter is the basic magnetic-field independence[18,20] (if $H_a <$ a few Tesla) on the quasiohmic differential resistance R_f, contrary to the linear dependence expected and observed for the flux-flow type of dissipation.[21] As pointed out by several authors[18,22] the dissipative sites are localized in the Josephson network of weak links (primarily grain boundaries). The status of a particular link will be determined by the local current with respect to the magnetic-field-dependent maximum Josephson current of the junction. If the local current and/or magnetic field overcome the respective thresholds, the working point in the I-V characteristic of a junction will be switched to the tunneling (dissipative) region. Thus in the sintered sample carrying a transport current $I > I_c$ there is a coexistence of the Ohmic dissipation of the "excited" (dissipative) weak links with the grains in the Meissner or mixed state. Consequently, for the sample in the status described by R_f (that is also the initial status of our samples before turning off the magnetic field) all grain-boundary weak links will be excited. Within the range of current and fields of our experiment the intragranular flux-creep contribution of the grains in the mixed state to the measured dissipation (voltage) is too small to be observed. Under these circumstances the dissipative voltage along the sintered sample is a measure of *the number* of the excited weak links. Accordingly, any time-dependent voltage on the sample should reflect the time evolution of this num-

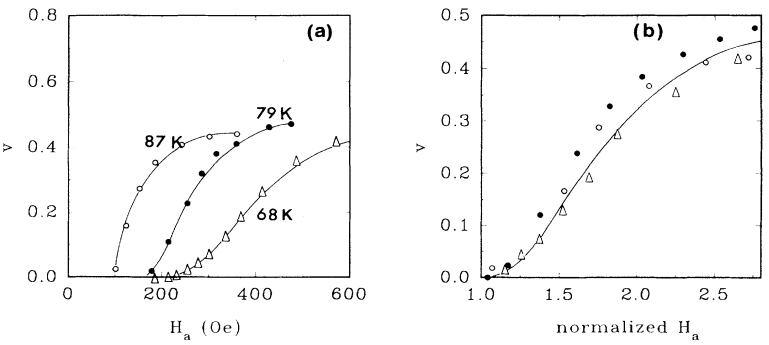

FIG. 2. Sample voltage v recorded 5 s after magnetic-field removal (normalized by the voltage V_{in} when the field was on) vs magnitude of applied magnetic field at three different temperatures is shown in (a). (b) shows the same as (a) with the field scaled by the minimum field sufficient for the onset of remnant voltage.

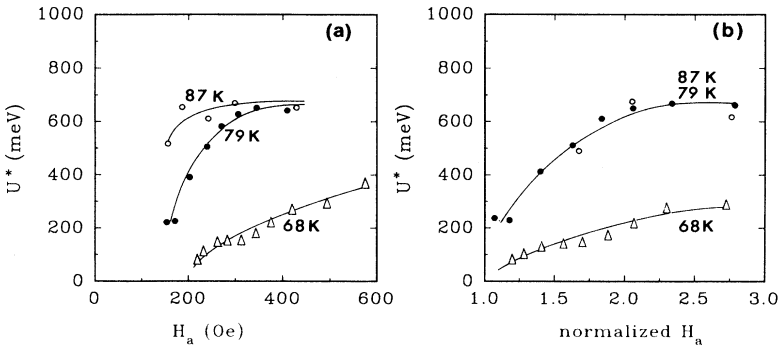

FIG. 3. Inverse of the normalized relaxation rate (multiplied by kT) vs magnitude of applied field at three different temperatures is shown in (a). (b) shows the same as (a) with the field scaled by the minimum field sufficient for the onset of remnant voltage.

ber. In the case of our experiments the applied magnetic field excites all weak links and additionally, if it is higher than the lower critical field, it penetrates into the grains and remains trapped inside. As the local field at the link site is determined by the sum of the applied and the dipole field of the neighboring grains, the weak links will be, after the removal of the external field, naturally divided into two classes. The resolving criterium is the relationship of the local field to the Josephson threshold H_J. The first class comprises those links that flip back to the nondissipative state immediately after the removal of the external magnetic field as the grain-boundary vortices should be very weakly pinned. We believe this process to be responsible for the initial exponential decay of voltage in our measurements. The millisecond time scale contrasts somewhat with the expected "instantaneous" process claimed in Ref. 11. The weak links of the second class remain in an excited state until the intragranular flux-creep-related decrease of the local field (presumably logarithmic in time) allows the reestablishment of the nondissipative current transport through these links.

The distribution of the local parameters of the problem (see below) introduces a time succession of the flip-back events in the long-time tail of our experiments. However, it is not clear why the measured voltage, i.e., the time-dependent *number* of excited links, should be logarithmic in time at all. In order to understand this point we analyzed a simple numerical one-dimensional model. The total resistance of a linear chain of 10 000 resistors connected in series was calculated under the condition that a resistance of an element is degenerate (0 or 1). The actual resistor value is determined by an external variable (dipole magnetic field of a neighboring grain). It was assumed that the external variable $H^k(t)$ (k is the resistor index) depends, below its starting value H_0^k, logarithmically on time:

$$H^k(t)=H_o^k[1-\alpha \ln(t)] , \tag{2}$$

where α is the usual Anderson's relaxation rate constant $\alpha=kT/U_a$. U_a is the apparent intragranular pinning potential, an observable of magnetization studies that will be discussed later. The moment t_c of a reversal event ($1\rightarrow 0$) of a resistor is defined by the validity of equation $H^k(t_c)=H_J^k$, i.e., by the moment when the field variable reaches the Josephson threshold H_J^k.

In order to apply this model to the physical situation a distribution of the local properties must be taken into account. There are many reasons why all parameters of the model (α, H_J^k, H_0^k) should be different at various resistor (i.e., weak-link) sites. The influence of the distribution function of flux-pinning potentials (i.e., of the parameter α) has been studied at length by Hagen and Griessen[23] and that of the strength of Josephson junctions (i.e., of parameter H_J^k) recently by Mee *et al.*[17] Looking for the qualitative behavior we neglect the spatial variation of H_J^k and α, keeping only the distribution of the local magnetic-field parameter H_0^k. The main reason for the nonuniformity of the local fields comes from the presence of the grain-size distribution in sintered samples. Indeed, the calculated dipole magnetic field at the grain boundary,[24] that takes into account also the various stages of flux-grain penetration, is a grain-size-dependent quantity. So the distribution function of the local field implicitly includes the distribution of the grain sizes. For the latter we have used the log-normal distribution function.[25]

The inset to Fig. 4 shows the time dependence of the calculated total resistance $R(t)$ after the external magnetic field had been turned off at $t=0$. It is obvious that the general features of the observed voltage response (Fig. 1) are reproduced by the model. The magnetic-field-dependent resistive drop at $t=0$ (from the initial resistance R_i to R_0) comes from those resistors whose local conditions ($H_0^k<H_J$) allow an immediate $1\rightarrow 0$ transition. After the drop the calculated resistance is time dependent (in Fig. 4, the choice for α was $\alpha=0.1$). It can be seen the this dependence is generally nonlogarithmic (note the ln t time scale). However, for a few initial decades of time an approximately logarithmic dependence can be identified. This allows modeling the resistance time dependence in the familiar "Anderson's" form $R(t)=R_0(1-\alpha_{ef}\ln t)$. Here α_{ef} is an effective relaxation rate, related but of course not identical to α.

In Fig. 5 we show the magnetic-field dependence of the resistive drop r ($r=R_0/R_i$, a quantity analogous to v), α_{ef} and $1/\alpha_{ef}$ (analogous to U^*). The magnetic field is normalized by the field value for which the resistive drop feature first sets in. The saturating behavior of the curves reflects a gradual penetration of the flux towards the center of the grains, when the trapping limit is achieved. The same type of behavior was observed in our voltage measurements (Figs. 2 and 3; the alternative explanation will be discussed later) and also in some magnetization

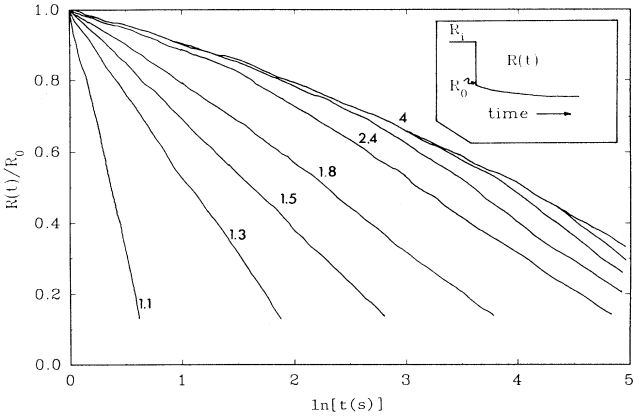

FIG. 4. Time dependence of the calculated total resistance of the chain resistor (see text), normalized by its value R_0 at $t=0^+$ in different magnetic fields. The field unit is the minimum field sufficient for the onset of nonzero resistance at $t=0^+$. Note the ln$t$ scale. Inset: Time dependence of calculated total resistance $R(t)$ shown schematically with the definition of specific resistive values R_i and R_0.

studies.[6,8] Very similar behavior has also been observed in the studies of magnetic nonlinearities[14] where the third-harmonic component saturates above the same field (approx. 400 Oe) at 77 K as in our dc voltage measurements. We note a large difference of α_{ef} and α in small fields, but their practically coincident values in higher fields.

As the main features of the voltage relaxation experiment are well-reproduced by the proposed model, the experimental data have been analyzed in terms of the formalism of this model. The magnetic field is thus normalized by the field that gives the first remnant voltage onset (Figs. 2 and 3). The drop curves (v, Fig. 2) are expected to be independent of temperature changes of the pinning potential as their model counterparts (r) do not depend on the choice of α. This is the reason for merging of normalized v curves at all three temperatures. The more important quantity, the flux-pinning-related U^* is the counterpart of α_{ef}. After the normalization, the two upper-temperature U^* curves (79 and 87 K) overlap and saturate at the factor-of-2 higher level compared to the lower-temperature (68 K) U^* curve. Applying the model result that α_{ef} tends to α in increasing magnetic fields, the level of U^* reached in saturation may be considered as a good estimate for the apparent intragranular pinning potential (U_a) itself. According to our measurements U_a increases with temperature from 68 to 79 K and remains constant up to 87 K. Above 87 K we did not detect any remnant voltage.

The temperature increase of U^* in our measurements is reminiscent of the temperature increase of U_a in magnetization studies.[6,7] This type of temperature dependence has been a subject of some controversy, as is not expected for a "true" pinning barrier U_p. However, it turns out that the main experimental results for U^* obtained by our measurements, i.e., its magnetic field and temperature dependence as well as its magnitude, can be qualitatively understood within the concept of an effective barrier[6,16,26] U_e actually involved in the flux-creep process. U_e is composed of the "true" pinning potential U_p diminished by the Lorentz interaction between the flux bundles and current J associated to the magnetic-field gradient. U_e can be expressed as $U_e=U_p f(J/J_{max})$, where f is generally a nonlinear function of J. J_{max} is a hypothetical critical current that would be effective in the absence of any creep and which gives $U_e(J_{max})=0$. If J is not much smaller than J_{max} the apparent pinning potential U_a is primarily determined by the slope of $U_e(J)$ curve ($U_a \propto \partial U_e/\partial J$). The magnitude of U_a is, consequently, a function of the position of the actual operating point on the $U_e(J)$ curve. Now both of the observed dependencies, on the magnetic field and temperature, may have the same origin—a critical current reduction [shift of the operating point to the region of higher $U_e(J)$ slope] due to an increase of H or T. Also, the magnitude of the saturation values of U^* is close to 1 eV, the latter value being the result of resistivity measurements[27] for the "true" pinning potential U_p. This suggests that the conditions of our experiment are compatible with a low operating intragranular critical current level. This is at variance with magnetization measurements where generally smaller measured U_a values could be attributed to the position of the operating point closer to the critical state. However, the simple "slope model" described above is of a limited value for the current far from J_{max} where a more complex dependence ($U_a \propto J\partial U_e/\partial J$) has to be taken into account.[6,16]

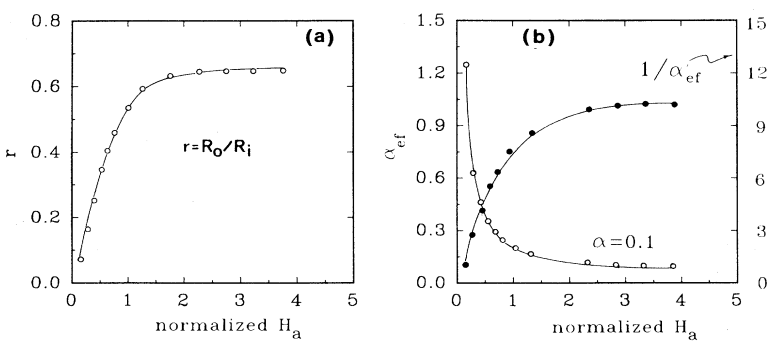

FIG. 5. Total resistance of the chain at $t=0^+$, normalized by the initial resistance R_i, as a function of the applied field is shown in (a). Field unit is the same as in Fig. 4. (b) shows the effective relaxation rates α_{ef} (initial slopes of curves in Fig. 4). Parameter of the calculation α is $\alpha=0.1$. The inverse $1/\alpha_{ef}$ corresponds to U^* of Fig. 3.

As far as the temperature dependence of U_a is considered, we believe that a convincing explanation can be found in the framework of the collective-creep theory.[28,29] In the work of Feigel'man *et al.*[29] it has been shown that U_a is constant at low temperatures but grows linearly with T at higher temperatures. The other explanation has been proposed by the work of Hagen and Griessen,[23] where U_a is related to the distribution function of the pinning energies. In their model the large value of U_a, reported by several authors including our present report, is justified by the high-energy tail in the asymmetric distribution function.

In conclusion, we have shown that the problem of flux creep can be successfully studied, complementary to the investigations of magnetization, by the measurements of (in most cases logarithmic) time decay of the remnant voltage along a sintered HTSC sample. The normalized relaxation rate of the logarithmic decay strongly depends on the applied magnetic field above some threshold value for the onset of remnant voltage and its inverse cannot be simply identified as an intragranular pinning potential. The interpretation of the experimental data takes into account that under conditions of our measurements, the dissipative voltage comes entirely from weak links. However, the time evolution of dissipation in this network is controlled by the grain magnetic field and the introduced numerical model shows that the normalized relaxation rate approaches the inverse of the intragranular pinning potential in the high-field limit. The pinning potential obtained in this way increases with temperature, in agreement with the results of magnetization studies and some recent theoretical concepts.

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