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Logarithmic-to-nonlogarithmic flux-creep transition and magnetic-flux hardening in Bi-Sr-Ca-Cu-O superconducting ceramics

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A sharp temperature- and field-dependent transition from a logarithmic to a nonlogarithmic flux-creep behavior has been found in Bi-Sr-Ca-Cu-O superconductors. The transition is not followed by any noticeable change in the magnetization. However, vibrating-reed measurements show that it coincides with a strong hardening of the magnetic structure. The transition occurs at fields and temperatures well below the reversibility line. The possible relation to the recently proposed flux lattice melting is discussed.

The bismuth-based Bi-Sr-Ca-Cu-O high-T_c superconductors show distinctive superconducting properties when compared to Y-Ba-Cu-O or La-Sr-Cu-O materials. The critical current of ceramic samples at 9 kOe approaches[1] zero at 40 K, well below the critical temperature T_c which lies near 100 K. In those experiments the critical current was deduced from magnetization measurements and accordingly, associated to the regions where the superconducting order parameter is strongest. In Ref. 1 it was pointed out that the flux-creep rate is even faster than in the Y-Ba-Cu-O system.

Another interesting and surprising result is the strong dissipation shown[2] by mechanical oscillator experiments in single crystals of Bi-Sr-Ca-Cu-O. The dissipation is made evident by a sharp peak almost independent of the orientation of the crystal, at temperatures of the order of 30 K and fields higher than 1 T. The dissipation occurs together with a change of the resonance frequency, indicating a softening of the flux lattice, when increasing temperature. This result has been interpreted[2] as a melting transition of the vortex lattice. The extremely low critical currents found at those temperatures has been suggested[2] to be a consequence of the melting of the vortex lattice.

In this Rapid Communication we focus our attention on the flux-creep behavior in the low-field region, showing a transition from a logarithmic to a simple power-law time dependence. The transition takes place in a narrow temperature range of about 3 K and is found to be field dependent. The time-dependent magnetization results are compared to vibrating-reed measurements. The transition in the flux-creep regime occurs at the same fields and temperatures where a change in the resonance frequency of the vibrating reed is detected. The reversible magnetization region is found at higher fields and temperatures. As a consequence, if the melting transition were to induce[2] a zero critical current the changes in the vibrating frequency and in the flux-creep regime detected in our experiments should not be associated to that transition.

One of the properties that distinguish the high-T_c oxides from classical superconductors is the $H^{2/3}$ line in the H-T phase diagram, separating reversible from nonreversible magnetic behavior. The interpretation of the reversibility line is still under discussion, being related to the properties of a grain-bond model or glasslike behavior by some authors[3] and to flux-creep effects by others.[4] The basis for a unified point of view has been provided recently,[5] where the grain-bond model and the flux-creep model are considered limits of a unique picture, in which the anomalously small and not well determined coherence length of these materials plays a fundamental role.[6] From our point of view the main difference in the applicability of the models depends on the length scale in which the order parameter is modulated. If it is modulated in lengths of the order of the coherence length the flux-creep model should be the proper limit while the granular picture might be appropriate when the characteristic modulation length is longer than the penetration depth.

The suggestion that melting and zero critical current are the consequence of the same phenomenon[2] implies that the reversibility region should be determined by the flux-melting process rather than by any of the mechanisms suggested in the previously mentioned models.

Due to experimental difficulties[2] the melting line has been determined at fields much larger than H_{c1}. We consider it important to investigate the low-field limit to verify if the reversible region is associated to a melting transition or to the existence of different flux phases, as recently suggested.[7]

To determine the dissipation induced by the flux structure at low fields we have used the vibrating reed method, which has been proved[8] to be a useful technique to study the properties of the vortex lattice. Ceramic samples have been used since the available[9] single crystals are too small for our experimental setup. As it is found[2] that melting is isotropic, the use of ceramic materials should not be a handicap in the detection of the melting transition, provided this phenomenon is the dominant process inducing dissipation. Superconducting quantum interference device (SQUID) magnetization measurements were used to detect the field dependence of the reversibility line and

flux creep at low fields.

The material was prepared by mixing reaction grade copper and bismuth oxides with calcium and strontium carbonates in quantities to satisfy the formula $Bi_4Ca_3Sr_3Cu_4O_{16+y}$. The components were ground and pressed to 1000 kg/cm^2 reacted in air at 800 °C for 18 h and then reground, sintered at 2500 kg/cm^2 and further baked in air for 18 h at 875 °C. X-ray diffractograms were fitted assuming a tetragonal cell with lattice parameters $a=3.82(1)$ Å and $c=30.68(1)$ Å. Energy-dispersive x-ray analysis gives $Bi_4Ca_2Sr_4Cu_4O_{16+y}$ for the formula of the resulting compound. The onset of T_c was at 85 K and resistivity became zero at 65 K with a midpoint at 81 K while the temperature dependence of the resistivity was "metallic" in character from room temperature downwards.

SQUID and vibrating-reed measurements were performed on samples obtained from the same ingot with approximately the same geometry (slabs of $1.3\times0.2\times0.02$ cm^3). Details on the experimental technique used in both types of experiments have been provided elsewhere.[10,11]

Our results indicate strong flux-creep effects in the magnetization, in agreement with Ref. 1. The temperature activated origin of the phenomenon is made evident in the zero-field-cooling experiments (ZFC). At the lowest temperatures, below 10 K, the irreversible zero-field-cooling magnetization is essentially determined by the temperature dependence of the critical current. When the temperature is raised the absolute value of the magnetization is determined by a competition between the temperature dependence of the critical current and the flux creep. Thus, it is evident that the definition of the reversibility line depends on the temperature sweep rate used in the experiment. Nevertheless, from a practical point of view the reversibility line is well defined within the typical time scale used to sweep temperature ($\simeq$1 K/min).

Figure 1 shows the reversibility line found from ZFC and FC magnetization measurements. Results obtained from single crystals[9] are plotted in the same graph. It is interesting to notice the agreement between the ceramic and single-crystal results. To define whether the coincidence represents an intrinsic behavior of the material or if it is a fortuitous result requires further investigation. Nevertheless, it is already an interesting result indicating that the modulation of the order parameter might induce the same magnetic behavior in single crystals and ceramics, in agreement with the discussion made in the introduction.

Figure 2 shows the time dependence of the magnetic flux penetrating into the sample, after applying a field of 100 Oe in a ZFC experiment. It is seen that the flux evolution is logarithmic at 48 K and nonlogarithmic at 53 K. The same transition is observed at around 39 K for a field of 400 Oe. It is interesting to mention that the nonlogarithmic dependence is perfectly fitted by a simple power law (full line). To make more evident the sharp transition in the flux-creep regime we plot in Fig. 3 the exponent β of the time dependence of $d\phi/dt \simeq t^{\beta}$ as a function of temperature for two different fields. The logarithmic dependence is given by $\beta=-1$. The temperatures and fields at which the transition takes place are plotted in Fig. 1. As expected, the change in regime occurs in a region of the phase diagram where nonequilibrium behavior is dominant. It is important to remark that the change in the flux-creep regime is not related to any abrupt change in the ZFC or FC magnetization, indicating that it should not be related to a superconducting transition of a phase with a lower-critical temperature.[12] The transition in the flux-creep regime is produced without discontinuous changes, either in the absolute value of the trapped flux or in the order of magnitude of the flux-creep rate; i.e., it is characterized by a qualitative change in the flux-creep behavior rather

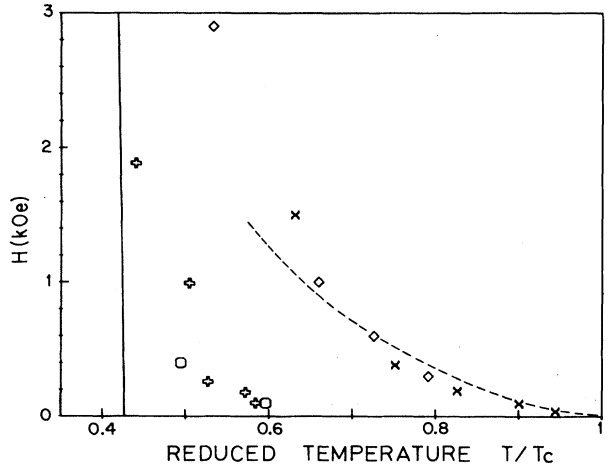

FIG. 1. Reversibility region determined from ZFC and FC measurements; × ceramic, ◇ single crystals. The dashed line represents an $H^{2/3}$ dependence. The □ represents the transition from logarithmic to nonlogarithmic flux-creep dependence in ceramic samples. The ✣ are the vibrating reed results indicating the transition from a weak flux structure to a hard one, as indicated in Fig. 4. The solid line is the extrapolation to low fields from Ref. 2.

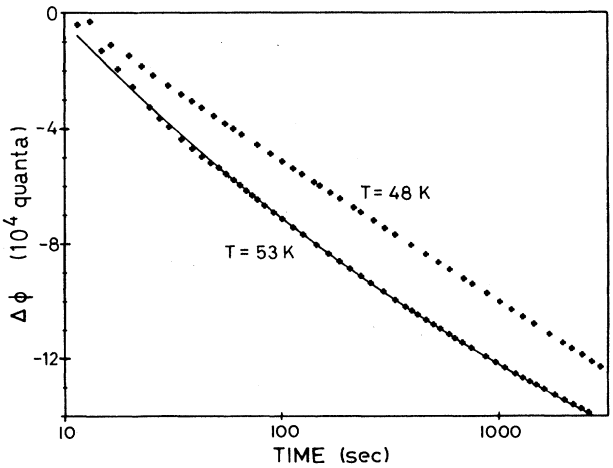

FIG. 2. Magnetic flux creep as a function of time for an applied field of 100 Oe in a ZFC experiment. The data show the logarithmic dependence at 48 K and clear deviations from this dependence at 53 K. The solid line corresponds to a fit with $\beta=1.15$ (see text). The full vertical scale represents about 15% of the ZFC flux exclusion.

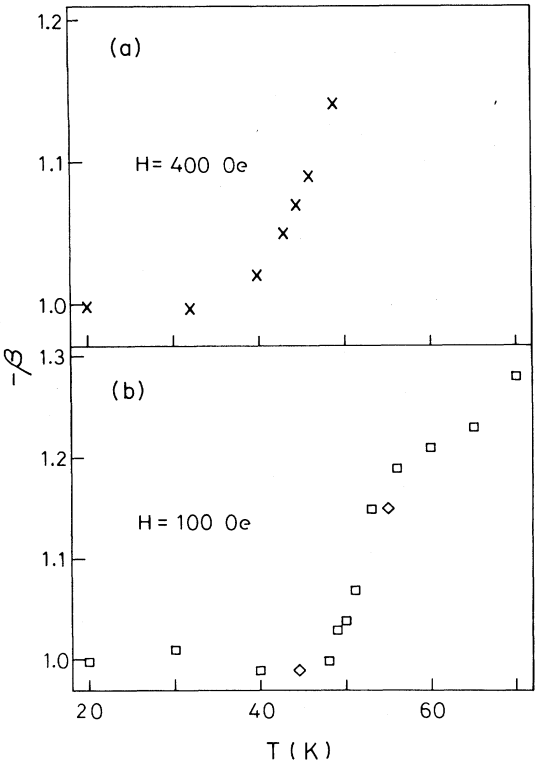

FIG. 3. Temperature dependence of the exponent β, as defined in the text. The exponent $\beta = -1$ corresponds to the logarithmic time dependence. (a) Results for 400 Oe after ZFC. (b) Results for 100 Oe after ZFC (□) and results for 100 Oe after FC in 400 Oe (◇).

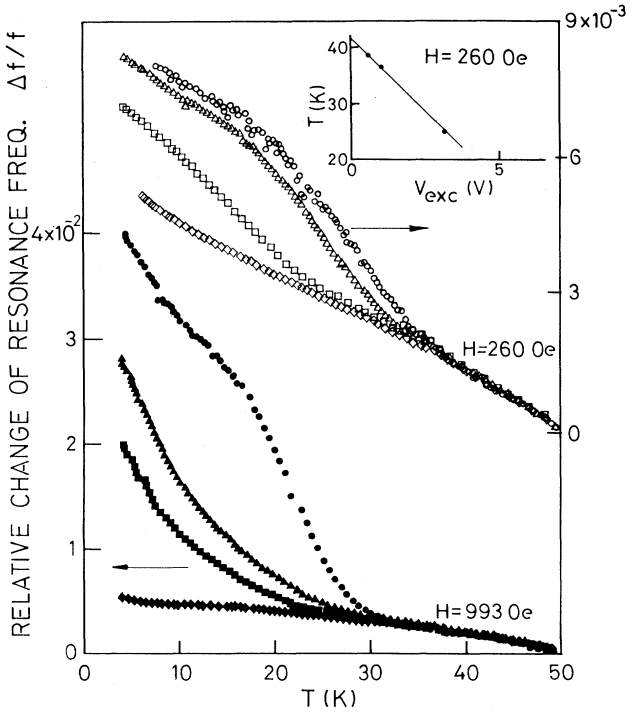

FIG. 4. Change of the resonance frequency of the vibrating reed as a function of temperature, at 260 Oe (open symbols) and 993 Oe (closed symbols) for different excitation voltages (○:V_{exc}=0.60 V; △:1.05 V; □:3.12 V; ◇:7.25 V; ●:5.88 V; ▲: 8.04 V; ■:12.94 V; ◆: 19.4 V). Inset: extrapolation to zero amplitude of the stiffening temperature at H=260 Oe.

than by a quantitative one. Another relevant result is that the temperature and field where the transition takes place is independent of the history of the sample. In fact, the exponent β at 100 Oe is the same whether the flux creep is induced by cooling the sample in zero field and then raising the field up to 100 Oe, or by cooling the sample in a field of 400 Oe and then decreasing it to 100 Oe (see Fig. 3). This shows that the change in the flux-creep regime is well characterized by field and temperature and is independent of the absolute values of the flux trapped inside the sample.

The vibrating reed results are quite interesting by themselves as well as when compared to the magnetization behavior. The results in Fig. 4 show the change of the resonance frequency as a function of temperature for different amplitudes of the driving force and two different applied fields. The results clearly indicate a well-defined temperature where the magnetic flux becomes stiffer, when decreasing temperature. As shown, the change in frequency is amplitude dependent and eventually might disappear at large enough amplitudes. Results for other fields as well as the behavior of the dissipation associated with the change in frequency will be reported elsewhere. For the purpose of this Rapid Communication it is sufficient to plot the fields and temperatures where the flux structure becomes stiffer, i.e., the point at which the frequency starts to increase above the value for large amplitudes. The values were obtained by extrapolating the experimental results to zero amplitude as shown in the inset. It is seen in Fig. 1 that the hardening of the response of the magnetic flux occurs well below the reversibility line and very close to the points indicating the change in the flux-creep regime. We believe these two results are correlated and are indicative of an abrupt change in the magnetic flux properties of the material. The full line in the same figure indicates the extrapolation of the melting line determined in Ref. 2. We find suggestive the coincidence among all these data, indicating that something drastic is happening in the flux lattice well below the reversibility line. Nevertheless, the magnetization measurements fail to indicate any significant change at those temperatures and fields. It is interesting to remark that the coincidence of the "melting curve" with the cancellation of the critical currents at high fields[2] seems to be a result of a coincidence of the melting and reversibility lines at those temperatures and fields rather than arising from the same phenomenon. Our results show that the change in the flux behavior does not imply the cancellation of the critical current.

In conclusion, a novel and sharp transition in the flux-creep regime has been detected at fields and temperatures where the vibrating reed experiments show an abrupt change in the magnetic response of the material. The

transition occurs at fields and temperatures well below those corresponding to the reversibility line. The critical current obtained from magnetization measurements does not show any anomaly corresponding to the transition. The possible relation of the change in flux-creep regime to the melting line reported in Ref. 2 is under investigation.

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