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Two-step transition towards the reversibility region in $Bi_2Sr_2CaCu_2O_{8-\delta}$ single crystals

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We have performed magnetization measurements on $Bi_2Sr_2CaCu_2O_{8-\delta}$ single crystals in the $\hat{c}$ crystallographic direction for fields from 2 Oe up to 700 Oe. The results strongly suggest that the reversible thermodynamic region is achieved after the vortex flux structure shows an abrupt transition at a temperature lower than that determined by the irreversibility line.

Recent ac susceptibility and mechanical oscillator measurements[1] in a wide range of fields and temperatures in $Bi_2Sr_2CaCu_2O_{8-\delta}$ single crystals have shown that the transition towards the reversibility region is made in two steps, characterized by two well-separated dissipation peaks at two different temperatures. The high-temperature peak is produced by the ac shielding current flowing in the Cu-O plains, while that at low temperature is determined by the current flowing through them. The corresponding H-T lines[1] in the phase diagram are found to be determined by the component of the dc field parallel to the $\hat{c}$ crystalline direction. The results become particularly interesting[2] in the low-field region where the shielding capability of the ac current flowing through the Cu-O planes is increased. This is indicated[2] by the shift of the transition towards higher temperatures. The low-temperature peak tends to disappear, and the dissipation takes place mainly at a single temperature, corresponding to that of the high-temperature peak.

In this picture[1,2] the two ac peaks are associated to changes in the intrinsic bulk properties of the vortex lattice. As a consequence, we expect that the dc magnetization of the same well characterized[3] $Bi_2Sr_2CaCu_2O_{8-\delta}$ single crystals should show a corresponding structure.

Due to the characteristics of our experimental setup we have concentrated the magnetization measurements in the low-field region, where even though the experimental data are scarce, the magnetic phase diagram shows[4] interesting but not well-understood behavior.

The zero-field-cooling (ZFC) and field-cooling (FC) magnetization measurements have been used to determine the dc irreversibility line and the thermodynamic behavior of the magnetization in the low-field region.

The dc magnetization results are shown to be quite similar to those obtained by the ac susceptibility, providing strong evidence that the two-step transition toward the reversible state is the consequence of the intrinsic behavior of the bulk properties of the flux structure.

The magnetization measurements were made using a custom-made cryostat and a commercial superconducting quantum interference device. Details on the experimental apparatus and measuring technique have been published elsewhere.[4] For the purpose of this work it is relevant to remark that the magnetic background in the sample holder is less than three flux quanta per Oe in the temperature range between 4 and 100 K. To avoid undesirable spurious magnetic signals induced by the movement of the sample in an inhomogeneous field, the magnetic flux was measured sweeping temperature at constant applied field. The earth remanent field was reduced to 10 mOe by proper shielding.

The sample was a single crystal with composition $Bi_2Sr_2CaCu_2O_{8-\delta}$ of dimensions 1 mm×1 mm×40 μm grown and characterized as reported elsewhere.[3] The quality of these samples has been further checked through low-field high-resolution magnetization[1] and flux lattice decorations.[5]

Figure 1 shows the ZFC flux exclusion, $\Delta\Phi(T)$, as a function of temperature for fields oriented in the $\hat{c}$ direction. Although our measurements are concentrated at relatively low fields, the flux exclusion for fields higher than 100 Oe shows a characteristic behavior of what we will identify as the "high-field" regime.

In the high-field range the decrease of the shielding current when the temperature is increased is characterized by a rapid change of the flux penetration at an almost field-independent temperature around 20 K. At higher temperatures the irreversible shielding current decreases smoothly with temperature until the reversible magnetization region is reached at $T_I(H)$ (see inset in Fig. 1, where two of the high-temperature FC and ZFC measurements are shown).

The magnetic flux exclusion in the low-field region is plotted in Fig. 2(a). In order to show the data in a single figure the flux exclusion is normalized by the applied field, H. The low-field magnetic shielding does not show the rapid decrease at 20 K, instead the shielding current decreases continuously in the whole range of temperatures, up to $T_I(H)$.

The high-field behavior reported here has been

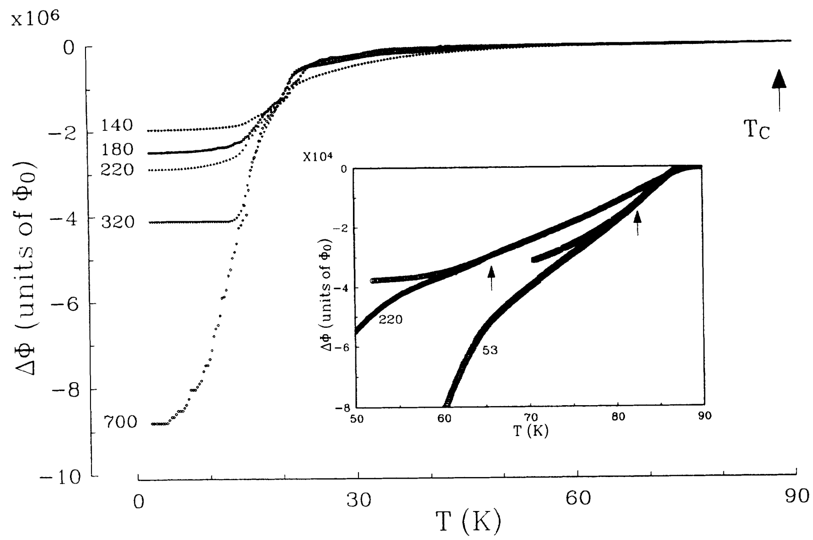

FIG. 1. ZFC flux exclusion defined as $\Delta\Phi(T) = \Phi(T) - \Phi(T > T_c)$ for different applied fields referenced by the labels. The inset shows a detail of the ZFC and FC expulsions for two different applied fields. The arrows indicate the temperature at which irreversibility for each field is first detected.

observed[6] in other experiments, up to much higher fields than those used in our measurements. However, the data are usually incomplete since the interest has been focused on the behavior of the irreversibility line. As a consequence, the low-temperature data are usually omitted in the literature. Similar data in the low-field region have been recently obtained using a small Hall probe technique.[7]

It is useful to analyze the data in terms of the behavior of critical currents. In principle, the flux exclusion in ZFC experiments is determined by two types of currents: the field- and temperature-dependent critical current of the material and the diamagnetic reversible current associated to the thermodynamic equilibrium state.

The total flux exclusion defined as $\Delta\Phi_T = \Phi(T_c) - \Phi(5$ K) is found to be nearly proportional to the applied field up to 320 Oe, as seen in Figs. 1 and 2(a). This indicates that at low temperatures and within the range of fields indicated, the critical state is limited to a small region of the sample. The relatively weak temperature dependence of the irreversible flux exclusion at temperatures below 20 K, shown in Figs. 1 and 2(a), can be interpreted within the critical-state model. In this picture the temperature dependence of the ZFC magnetic field profile is induced by the temperature dependence of the critical current. The experimental results show two well-defined temperature regions: One at $0 < T < 20$ K, where the flux exclusion is nearly proportional to the field and weakly temperature dependent until close to 20 K and the other at higher temperatures where the flux exclusion decreases with increasing field. Both regions are separated by a narrow temperature boundary where the flux penetrates rapidly into the sample. At that temperature, the critical current undergoes an almost discontinuous transition towards a smaller value, inducing the critical state in the whole sample, as indicated by the reduction of the flux exclusion to an esentially independent value at 20 K, well below the reversible

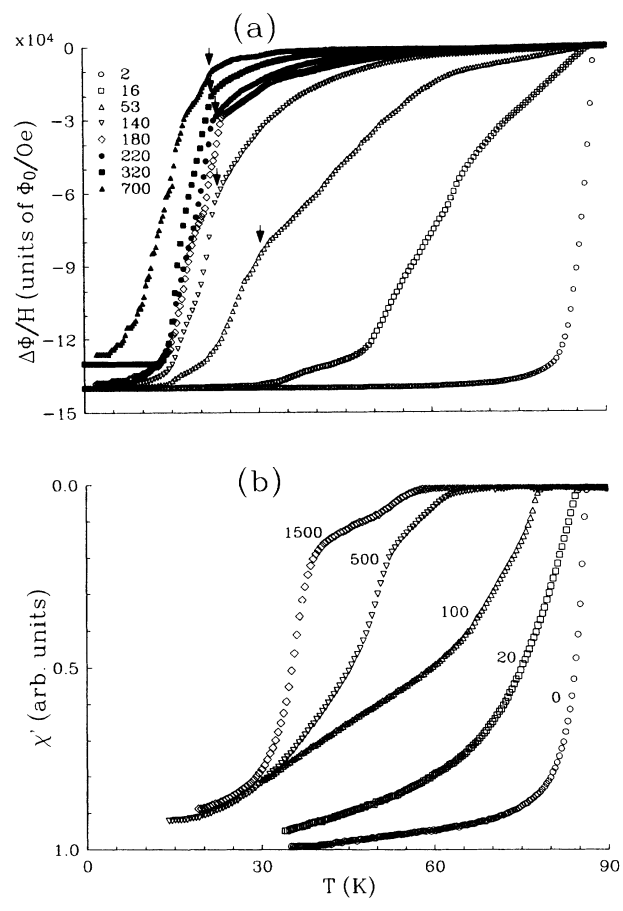

FIG. 2. (a) ZFC flux exclusion normalized by the applied field for different values of the applied field. The numbers indicate the value of the field in Oe. The arrows indicate T_M (see text) for each field. (b) ac susceptibility for different applied fields parallel to the $\hat{c}$ crystallographic direction from Ref. 2.

region. This is a rather unexpected behavior because in a type-II superconductor with pinning, the critical currents decrease continuously with temperature leading to the establishment of the critical state in the whole sample at a field-dependent temperature. It is interesting to contrast the difference between this rapid change of the flux penetration in the low-temperature range with the smooth and continuous approach to the reversible state at $T_I(H)$. This rapid change in the shielding capability indicates a transition from a vortex pinned state to another pinned phase.

The results in Fig. 2(a) show the change in the behavior of the normalized flux exclusion as a function of field. It can be seen that for fields under 100 Oe the relative flux exclusion at 20 K is continuously increased with decreasing field. The data at 140 Oe show already a clear change in the behavior of the flux penetration, when compared to that at higher fields: Although the flux starts to penetrate rapidly below 20 K a well-defined change in the curvature of $\Delta\Phi(T)/H$ indicates the transition towards a state characterized by a larger normalized flux exclusion compared to that of higher fields. The temperature, T_M, where the change in regime takes place is indicated by an arrow in the figure. As the field is lowered T_M shifts towards higher temperatures.

The real component of the ac susceptibility, χ', found in Ref. 2 is shown in Fig. 2(b). The similarity between the ac and dc data is remarkable. This is particularly relevant because the response of pinned vortices to ac fields is known to be qualitatively different from that expected in dc experiments.

To be able to compare the ac and dc results it is convenient to discuss the expected response of a vortex system to the ac field. The ac data shown in Fig. 2(b) were taken[2] in a configuration where the ac field, **h**, induced by the primary of a mutual inductance, is perpendicular to **H**. If the vortices are in thermodynamic equilibrium the ac field modulates the magnetization, **M**, changing its angle with respect to the applied field **H**. In the limit of $|\mathbf{h}| << |\mathbf{H}|$ the voltage induced in a secondary, parallel to **h**, is given by $V \simeq \chi'_\perp = |\mathbf{M}|/|\mathbf{H}|$. On the other hand if the vortex lattice is fully pinned, shielding currents avoid the change of the flux inside the sample and the response to the ac field is the same as that of the Meissner state.

In this discussion we assume an instantaneous response of the magnetization, disregarding the important effects[8] induced by the finite resistance of the superconductor in the equilibrium state. Since our discussion concerns the behavior of vortices in the H-T region of the phase diagram, well below the irreversibility line, the linear response theory[8] cannot be used to discuss our results.

The results of Figs. 2(a) and 2(b) in the low-temperature region show the behavior expected for a pinned vortex system. The large $\Delta\Phi/H$ of Fig. 2(a) is determined by the critical current flowing in the sample as previously discussed. The corresponding ac response indicates a field-independent behavior as expected from a fully pinned system. However, the data for $T > 20$ K show that $\Delta\Phi(T)/H$ is similar to χ'. The imaginary part of the susceptibility shows[2] a peak at the temperature where the slope of χ' is maximum. This dissipation demonstrates that above 20 K there is a variation of flux in the sample associated to the ac field. Since the magnetization of the sample is proportional to $\Delta\Phi$ we see that the data of Fig. 2 strongly suggest that in the high-temperature range 20 K$< T < T_I(H)$, χ' is determined by $\Delta\Phi/H$. The shift of the transition towards higher temperatures as measured by χ' is probably due to the frequency dependence of the transition.[2] The combination of the ac and dc measurements demonstrates that the transition at T_M found in the $Bi_2Sr_2CaCu_2O_{8-\delta}$ crystals is determined by a change in the bulk intrinsic properties. The results disregard the possibility that the ac transition is related to a geometrical matching of a skin depth to some sample dimension.[8] The almost discontinuous variation of the critical current supports the suggestion made in Refs. 1 and 2 that the magnetic flux structure shows a bulk phase transition at T_M, before reaching T_I. It is remarkable that previous measurements[9] in the same compound have shown a change in the time dependence of the flux creep magnetization decay, at temperatures close to T_M. It is important to notice that the combination of the ac and dc results indicates that the flux in the sample in the ac experiments is modulated by the alternating field. On the other hand the dc data show that below T_I there is a finite critical current. The simultaneous presence of flux modulation and pinning points toward the existence of two different types of superconducting currents, as will be discussed later.

Figure 3 shows T_M as a function of field. In the same figure we have plotted the irreversibility line, $T_I(H)$, obtained from the magnetization data in the same experiments. The difference between the two lines is evident and they resemble the two lines reported in Refs. 1 and 2 obtained from ac experiments.

Up to now we have described the experimental results. The data support the existence of a vortex phase transition. The ac data suggest[1,2] that the low-temperature transition could be related to a loss of phase coherence in the $\hat{\mathbf{c}}$ direction while the irreversibility line would represent the depinning of the quasi-two-dimensional vortices. From this point of view the experimental data indicate a

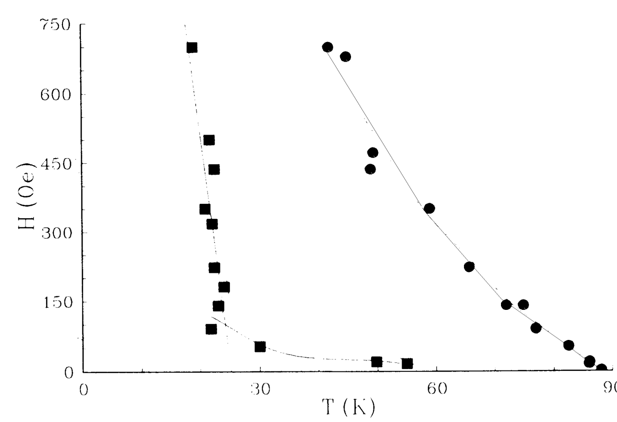

FIG. 3. H-T phase diagram obtained from dc magnetization measurements. (■) T_M, (•) T_I; see text for definitions.

well-defined change in the shielding capability at an almost constant temperature, as expected in a transition from a three-dimensional (3D) behavior at low temperatures and high fields to a 2D vortex system[10] at high temperatures. The low-field data suggest a continuous transition probably towards a 3D behavior in the whole range of temperatures.

An intriguing result is the lack of detection of a phase transition in FC experiments. However, this is not necessarily surprising, since the current induced by the metastable FC state is much lower than that in ZFC experiments and could be sustained by the pinned vortices in any of the two phases. On the other hand if the transition at 20 K is the same as that detected through picovoltmeter experiments[11] and suggested to be of first order, the FC flux expulsion should show a discontinuity[12] at T_M. Careful measurements are in progress to determine that possibility.

Although a melting transition has been discussed theoretically by many authors,[13] the two-step transition toward the reversibility region was theoretically predicted by Glazman and Koshelev.[14] They suggested an ac experiment with a geometry identical to that used in Refs. 1 and 2. Their theoretical description of the high-field regime coincides qualitatively with the experimental results. In recent Monte Carlo simulations Ryu *et al.*[15] have found two melting curves in the phase diagram. In this case the low-temperature melting is associated to the disappearence of the in-plane translational order.

One recent study of the dynamic response of the vortex lattice[16] is particularly interesting. It was found that at a well-defined temperature a transverse phononic mode tends to zero and this temperature is defined as the melting temperature. When these calculations are applied to the $Bi_2Sr_2CaCu_2O_{8-\delta}$ system it is found[16] that the melting line lies at lower temperatures than the experimental irreversibility line, in agreement with our results.

Although the dc magnetization results reported in this work strongly support the existence of two lines in the phase diagram we cannot make a quantitative comparison with theoretical results that have not taken into account the presence of pinning, as experimentally observed in the dc and ac results.

In conclusion, the dc ZFC flux exclusion shows an almost discontinuous change of the shielding capability, at a temperature which is weakly field dependent. The magnetic flux response above and below that temperature is qualitatively and quantitatively different. The experimental results, if interpreted within the usual critical current models, show that for fields higher than 100 Oe the critical state in the whole sample is achieved at a temperature close to 20 K, almost independent of the applied field and well below the irreversibility line. The results strongly suggest that the reversible thermodynamic region of $Bi_2Sr_2CaCu_2O_{8-\delta}$ crystals in the high-field range is achieved after the vortex flux structure has gone through a thermodynamic phase transition, in agreement with previous ac results.

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