Competition between surface barriers and bulk pinning in a $Nd_{2-x}Ce_xCuO_{4-y}$ single crystal down to $T/T_c = 0.02$

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Isothermal dc magnetization measurements with the magnetic field applied parallel to the tetragonal c axis were made on an overdoped $Nd_{2-x}Ce_xCuO_{4-y}$ single crystal with a superconducting critical temperature $T_c \approx 19$ K down to a reduced temperature $T/T_c \approx 0.02$, an order of magnitude lower than values reported to date. The measurements revealed a pronounced ''peak effect'' anomaly in the irreversible part of the magnetization loop in which the magnetic field at which the peak occurs increases linearly with decreasing temperature down to $T/T_c \approx 0.05$. The field where flux initially penetrates the sample increases exponentially as $T \rightarrow 0$ due to surface barriers, obscuring the features of the peak anomaly at low temperatures. The irreversibility field follows a power law down to $T/T_c \approx 0.2$ and deviates from the power law at low temperatures, again indicative of surface barriers. [S0163-1829(98)51602-5]

The vortex ensemble in the mixed state of high-temperature cuprate superconductors has been investigated extensively during the past decade.[1–3] However, due to the enormously high upper critical fields H_{c2} of these materials, the H-T phase diagram in the limit $T \rightarrow 0$ has not been completely explored. It is of fundamental interest to expand the studies of the vortex phases to lower temperatures where pinning induced effects prevail over thermal distortions of the flux line lattice. The irreversibility line and the peak anomaly, or ''peak effect,'' are two striking examples of pinning-related transformations in the vortex ensemble. The peak anomaly, manifested as a region of increased irreversibility in isothermal magnetization loops, has been observed in a number of high-T_c cuprate superconductors including the highly anisotropic $R_{2-x}Ce_xCuO_{4-y}$ (R = Pr, Nd) (Refs. 4 and 5) and $Bi_2Sr_2CaCu_2O_x$ (Bi-2212) compounds[6–9] as well as the less anisotropic $YBa_2Cu_3O_{7-\delta}$ (YBCO) compound.[10–13] Considerable efforts have been made to explain the physical origin of these peaks, and their possible connection with phase transitions in the vortex ensemble. Some low-T_c compounds such as $CeRu_2$ (Ref. 14) and 2H-$NbSe_2$ (Ref. 15) also exhibit peak anomalies which generally occur just below the melting line of the vortex lattice, i.e., near H_{c2}. Generally, the enhanced pinning is thought to occur as a result of a softening of the vortex lattice[16] which allows the vortices to better accommodate to the random array of pinning sites in the sample. Recent theoretical[17] and experimental[18,19] results have shed light on the fascinating dynamics of this ''plastically'' deformed vortex lattice.

The irreversibility line $H_{irr}(T)$ lies below the mean-field second-order phase transition at $H_{c2}(T)$ and separates the region with critical current $j_c \neq 0$ (lower temperatures and fields) from the one with $j_c = 0$ (higher temperatures and fields). There have been numerous studies of $H_{irr}(T)$ near T_c (Refs. 6 and 20–24) but there is a lack of experimental data well below T_c due to the very high fields that are typically required.

Our experiments were performed on single crystals of $Nd_{2-x}Ce_xCuO_{4-y}$. The $Nd_{2-x}Ce_xCuO_{4-y}$ system is an ex-

cellent candidate to study the effects of pinning in the low-temperature–high-field region of the magnetic phase diagram since it is intrinsically disordered, is not twinned, and has a much lower value of T_c than many of the other cuprate superconductors. The lower value of T_c results in a lower value of $H_{c2}(0)$, making the phase diagram easily accessible to experimentally available magnetic fields.

We report results of isothermal magnetic hysteresis loop measurements on an overdoped $Nd_{2-x}Ce_xCuO_{4-y}$ (NCCO) single crystal with a T_c of ~19 K. The crystal was shaped like a rectangular parallelepiped of dimensions $1 \times 0.8 \times 0.005$ mm^3 and had a mass of 0.200 mg. This value of T_c is reduced compared to the maximum value of $T_c \approx 25$ K observed for optimally doped NCCO samples. The NCCO single crystal was grown using a self-flux method and annealed at 1000 °C for 30 h in flowing argon to reduce the oxygen concentration to its near optimum value.[25] Overdoping with cerium was accomplished by growing the crystal from a starting material containing excess cerium ($x = 0.29$).

The magnetization $M(H)$ measurements revealed a pronounced peak effect in the mixed state that persists throughout the entire range of reduced temperatures investigated ($0.02 \leq T/T_c \leq 1$). From these measurements, the temperature dependence of the peak anomaly and the irreversibility line $H_{irr}(T)$ were determined to the lowest reduced temperatures heretofore achieved, $T/T_c \approx 0.02$. The temperature dependence of the onset H_{on} of the peak anomaly in the high-temperature range ($T/T_c > 0.3$) suggests the occurrence of a field-induced transition of the vortex lattice from an ordered to a more disordered state as proposed by Ertas and Nelson[26] and Giamarchi and Le Doussal.[27] At lower temperatures $T/T_c < 0.3$, we find that it becomes difficult to discern H_{on} in the magnetization curves, apparently due to the influence of surface barriers.

Magnetization $M(H)$ measurements for $H \| c$ were made after cooling the sample in zero field to the desired temperature using a commercial superconducting quantum interfer-

ence device (SQUID) magnetometer (Quantum Design) operating in magnetic fields up to 5.5 T and at temperatures in the range $1.8 \leq T \leq 300$ K. The magnetization loops were traced while increasing/decreasing the magnetic field in small steps (≤ 25 Oe). The low-temperature data ($T \leq 2$ K) were obtained using a Faraday magnetometer (FM). Because two different methods were used in the $M(H)$ measurements, the consistency between the two magnetometers was checked by comparing magnetization loops measured with both magnetometers in the region $2 \leq T \leq 4$ K. We concluded that spurious effects[28,29] due to movement of the sample encountered in commercial SQUID magnetometers was not a significant source of error. As our investigation of the magnetic phase diagram does not require absolute values of the magnetization, we did not take into account small corrections arising from the sample holder susceptibility in the measurements with the SQUID magnetometer. For the FM measurements, a large background signal due to the quartz sample holder was subtracted from the measured magnetization.

Shown in Fig. 1 are plots of the irreversible part of the isothermal magnetization. For clarity, $M(H)$ curves for positive applied magnetic field at three selected temperatures are included in the figure. Measurements of the entire hysteresis loop for positive and negative magnetic fields (not shown) for a few temperatures revealed no unusual behavior.

From Fig. 1, we see that the peak anomaly is temperature dependent, occurs at low fields, and is still discernible at the lowest measured temperature as illustrated in the inset of Fig. 1(b) ($T = 0.4$ K). For increasing magnetic field, we identify three main features in the $M(H)$ curves: (1) the initial penetration of flux into the sample at H_{pen} [inset of Fig. 1(a)]; (2) the downturn in the magnetization at the onset H_{on} of the peak anomaly, and (3) the maximum in the difference in magnetization for increasing and decreasing H of the peak anomaly at H_{peak} (open arrows in Fig. 1). It is important to emphasize that H_{pen} does not represent the lower critical field H_{c1} which is located at lower fields [we estimate $H_{c1}(T=0) \approx 25$ Oe, which is suppressed due to the sample's large calculated demagnetizing factor $D \approx 0.9$]. The field H_{on} appears to be associated with the onset of a new pinning regime, while H_{peak} represents the field at which the pinning is a maximum. However, there is not a clear break in slope in $M(H)$ signaling H_{on}, which makes it difficult to track H_{on} precisely. For temperatures below 1.5 K, H_{on} and H_{peak} become indistinguishable [see inset of Fig. 1(b)], although it is evident that there is still an unusual dependence of the magnetization on the field in this region, suggesting that the mechanism producing the peak anomaly is still operative. In Fig. 1(b), the irreversibility field H_{irr} is indicated for $T = 4.3$ K. We define the irreversibility field from the $M(H)$ curves as the point where the difference between the values of the magnetization for increasing and decreasing fields begins to deviate from zero to within the accuracy of the experiment ($\pm 10^{-5}$ emu).

Transitions in the vortex lattice of cuprate superconductors induced by pointlike disorder were recently addressed in two complementary models.[26,27] In these models, it was proposed that a competition between pinning, thermal, and elastic energies in the vortex ensemble can explain the experimentally observed vortex phases in the mixed state. For instance, the irreversibility line is found by equating the pin-
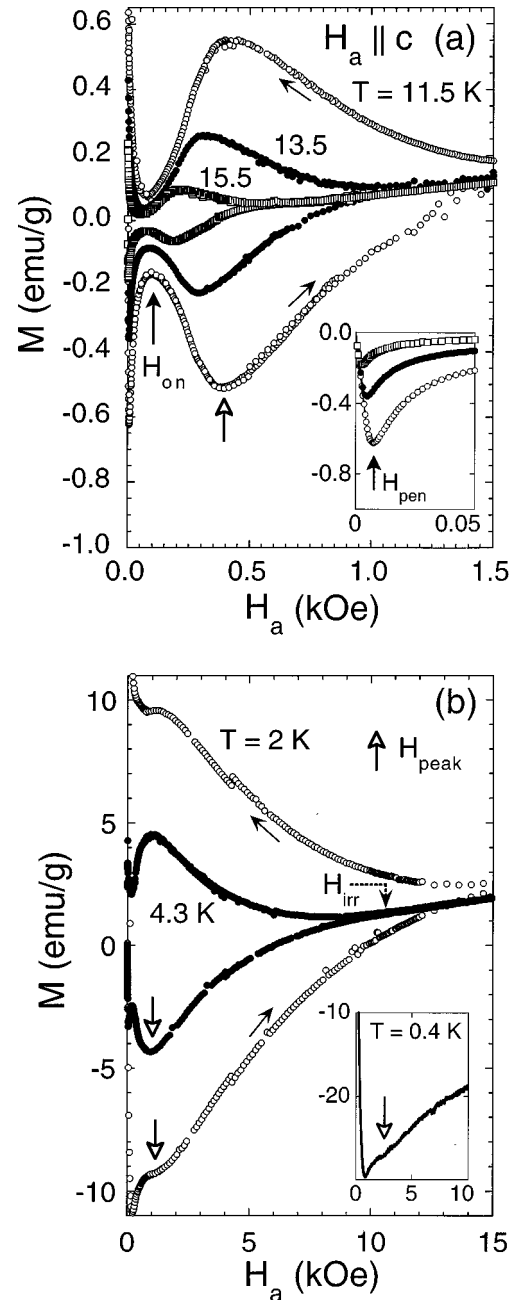

FIG. 1. Isothermal magnetization curves at both high (a) and low (b) temperatures in the superconducting state of a NCCO single crystal, plotted as a function of the externally applied magnetic field H_a. The peak anomaly at H_{peak} is indicated by the open arrows. In panel (a), the onset field of the peak anomaly H_{on} as well as the initial flux penetration at H_{pen} (see inset) are indicated for $T = 11.5$ K. Panel (b) shows the irreversibility field H_{irr} for $T = 4.3$ K, and the peak anomaly at the lowest temperature $T = 0.4$ K (see inset).

ning and thermal energies, and the melting line from equating the thermal and elastic energies. In the case of NCCO, where $T_c \approx 20$ K is lower than for most of the cuprates, it is largely the competition of the pinning and elastic energies which determine the behavior of the vortex ensemble. Specifically, a crossover from a relatively ordered, elastic vortex lattice to an entangled, pinning-dominated vortex solid is predicted as the applied field is increased. Using local mag-
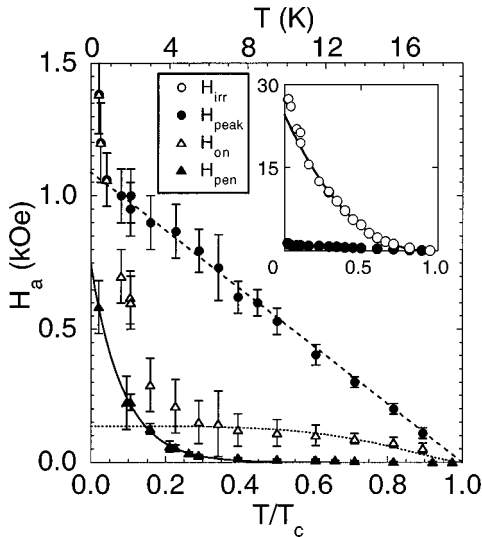
FIG. 2. Superconducting H-T phase diagram for a NCCO single crystal, showing the initial flux penetration field H_{pen}, the fields H_{on} and H_{peak} for the onset and maximum of the peak anomaly, and the irreversibility field H_{irr} (inset). See text for discussion.

netic measurements, Giller *et al.*[30] identified the onset of the peak anomaly at H_{on} in NCCO with this crossover, making the intuitive assumption that the pinning of the entangled vortices is enhanced over that of the ordered vortex lattice. By equating the elastic and pinning energies, and assuming that pinning arises from local fluctuations in T_c, the following behavior was predicted:

$$H_{on}(T) = H_{on}(0)[1 - (T/T_c)^4]^{3/2}. \qquad (1)$$

Giller *et al.* reported measurements on NCCO spanning the temperature range $0.35 < T/T_c < 0.91$, for which Eq. (1) describes the data well. We find that Eq. (1) provides a good description of our data for $H_{on}(T)$ over a similar range $0.3 < T/T_c < 1$. Shown in Fig. 2 is a superconducting H-T phase diagram for the NCCO crystal studied in this work, with H_{on} indicated by open triangles, and the fit to Eq. (1) as a dotted line.

Extending the magnetic measurements to lower temperatures than in previous work, we observe a dramatic enhancement of H_{on} over the behavior expected from the fit of Eq. (1) to the data at higher temperature which appears to be due to barriers to vortex penetration at the surface of the sample. The significance of so-called Bean-Livingston surface barriers[31] has been noted previously[5] in the NCCO system. Due to the extreme anisotropic nature of these materials, it has been proposed[32] that two-dimensional pancake vortices may be thermally activated over the surface barriers, in contrast to a conventional superconductor in which the barrier to a three-dimensional vortex line is infinite if the applied field is less than the field of initial vortex penetration H_{pen}. In a conventional superconductor, H_{pen} is of the order of the thermodynamic critical field H_c which saturates to a constant value at low temperatures. However, in the case of layered superconductors, thermal activation of pancake vortices over the surface barriers dramatically alters the value of $H_{pen}(T)$, which has been shown to take the following form at low temperatures:[33]

$$H_{pen}(T) = H_{pen}(0) e^{-T/T_0}, \qquad (2)$$

where T_0 is a constant.[34] We find that our $H_{pen}(T)$ data (solid triangles in Fig. 2) are described well by Eq. (2) for $T/T_c < 0.3$ (solid line). We notice from Fig. 2 that it is in this same temperature range that H_{on} begins to deviate from the fit to Eq. (1). This is a strong indication that thermally activated vortex creep over surface barriers complicates the determination of the onset of the peak anomaly at low temperatures.

Although H_{on} seems to be related to a transition in the vortex structure, it is not clear that H_{peak} also tracks this transition. There have been different explanations for the peak anomaly in the less anisotropic cuprate superconductor Y-Ba-Cu-O.[11–13,35,36] The correlation between the peak anomaly and a possible phase transition in the vortex lattice of the highly anisotropic Bi-2212 surfaced in light of neutron and μSR experiments[37,38] which revealed a disappearance of the ordered vortex lattice in the vicinity of the peak anomaly. Most of the peaks observed in highly anisotropic materials[4,7,39] have at most a weak temperature dependence, in contrast with the relatively strong T dependence of H_{peak} observed here. We note that a dimensional crossover in the vortex ensemble is ruled out because it predicts a temperature-independent crossover field.[40] A striking result from the present work is that $H_{peak}(T)$ (solid circles in Fig. 2) has a simple linear T dependence $H_{peak}(T) = H_{peak}(0)(1 - T/T_c)$ for essentially all temperatures ($T/T_c > 0.05$). Again, deviations as $T \rightarrow 0$ are thought to be due to surface barriers.

We find that the temperature dependence of the irreversibility field $H_{irr}(T)$ for most of the temperature-field region is consistent with the phenomenological Lindemann melting criterion and can be described as a power law of the form $H_{irr} = A(1 - T/T_c)^m$ with $A = 23$ kOe and $m \approx 2.4$ in the range $T/T_c \geq 0.2$. (See inset of Fig. 2, where the solid line represents the power-law fit.) The interpretation of the irreversibility line as the melting of the flux line lattice[41–43] has been confirmed in certain cases.[24,37,44] We did not observe evidence of a thermodynamic phase transition, such as a jump in the magnetization, as one would naturally expect if indeed the irreversibility and melting lines coincide.[24] However, this could be related to the experimental sensitivity. An interesting observation is that for $T/T_c \geq 0.2$, the irreversibility line has a steeper, perhaps exponential temperature dependence. Recent studies[45,46] have indicated that surface and geometrical barriers[47] may play an important role in determining not only the penetration field $H_{pen}(T)$, as discussed earlier, but also the irreversibility line. A similar exponential dependence $H_{irr} \propto \exp(-T/T^*)$, where T^* is a constant, fits our data well for $T/T_c < 0.5$, implying that thermally activated flux creep might influence the position of the irreversibility line at low temperatures. Further study of the vortex state in this system will be directed at discerning intrinsic bulk pinning properties from surface effects, and in theoretical modeling of the $T \rightarrow 0$ behavior of the irreversibility line.

In summary, we have determined the temperature dependence of the peak anomaly and the irreversibility line in a single-crystal sample of NCCO over a wide range in temperature, shedding light on the $T \rightarrow 0$ limiting behavior. The

field at which the peak anomaly exhibits a maximum has a linear temperature dependence throughout the phase diagram. The onset of the peak anomaly is consistent with the temperature dependence proposed for a field-induced disorder transition in the vortex ensemble. However, a sharp departure from this behavior is observed at low temperatures, which may be related to a competition between surface barriers and bulk pinning. The irreversibility line follows a power law predicted for the melting of the vortex lattice for most of the field-temperature range. However, it is clear that more work is needed in order to understand the role of surface and geometrical barriers versus bulk vortex pinning in determining the phase diagram of this unique cuprate superconductor.

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