First-order vortex lattice melting in twinned $NdBa_2Cu_3O_{7-y}$ single crystals

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We report on the observation of a first-order melting transition of the flux-line lattice in a twinned $NdBa_2Cu_3O_{7-y}$ single crystal by transport (for both $H \| c$ and $H \| ab$ directions) and magnetization measurements. The position of kink and hysteresis in resistance and magnetization jump coincides in the B-T plane. The observed melting transition is a strong function of O_2 partial pressure during growth. The first-order transition becomes continuous and the peak in critical current density first emerges with oxygen partial pressure $\geqslant$0.07%. Our results demonstrate the existence of a first-order melting transition in a twinned RE-123 system containing rare-earth magnetic ions. [S0163-1829(99)06717-X]

I. INTRODUCTION

First-order *melting* transition from an Abrikosov vortex lattice state to a vortex liquid state has been recently observed in clean and untwinned $YBa_2Cu_3O_{7-y}$ (YBCO) single crystals[1–4] in a variety of experiments probing characteristic singularities in specific heat, magnetization, vortex transport, and neutron diffraction. The first-order melting line is characterized by a sharp ''kink,'' hysteresis in resistance near the kink region, and, more importantly, a sharp jump in magnetization[3] revealing the actual thermodynamic signature of the first-order transition. The first-order transition in YBCO single crystals is suppressed by disorders as demonstrated in twinned[5] and electron-irradiated untwinned single crystals.[6] Similar signatures in favor of a first-order transition have been observed in optimally and overdoped $Bi_2Sr_2CaCu_2O_x$ single crystals.[7]

It remains, however, unsuccessful so far to obtain high quality clean other rare-earth-based single crystals to exhibit the first-order transition. The recent investigation of a magnetic rare-earth-based superconductor, $NdBa_2Cu_3O_{7-y}$ (NdBCO) is very exciting due to its high-T_c (~95 K) and high flux pinning resulting in high critical current density, J_c.[8–10] The efficient flux pinning by magnetic rare-earth ions (Nd) on Ba sites in NdBCO is thought to be a natural consequence.[11] The magnetic rare-earth ions on Ba sites are thought to be one of the best flux pinners and pair breakers due to magnetic scattering. For this reason, NdBCO single crystals are believed to retain some disorders both due to Nd on Ba site substitution and oxygen vacancies as well. Apart from these defects, NdBCO crystal contains twin boundaries as found in YBCO crystal. Unlike YBCO, there is no report so far available on a pure NdBCO crystal having 123 stoichiometry. Probably, it has been thought that NdBCO is an intrinsically nonstoichiometric system having $Nd_{1+x}Ba_{2-x}Cu_3O_{7-y}$ structure. Several magnetization and transport measurements have been reported on this crystal in favor of a continuous or vortex glass transition.[12,13]

In this paper, we present transport and magnetization measurements on a very high quality twinned NdBCO single crystal and demonstrated the observation of a first-order melting transition in such a system for the first time. The sharper ''kink'' feature in high field compared to the zero-field resistance, hysteresis in the field dependence of the resistance in both field orientations together with jump in magnetization confirm this interesting observation. Precise current-voltage (I-V) characteristic clearly distinguishes the melting of solid into a liquid phase. The most important point in our observation is that the first-order transition in NdBCO single crystal, previously thought to be an intrinsically nonstoichiometric system, is found to be robust even in the presence of twin boundaries. We have also shown how this first-order transition turns out to be continuous with increasing Nd-Ba substitution.

II. EXPERIMENT

The present samples were grown by self-flux method using a special technique under 0.01–0.07 % O_2 partial pressure (PO_2) in Ar atmosphere and the details will be described elsewhere.[14] The crystals showed superconductivity only after annealing at 320 °C for 300 h. The crystals show twin plane in both (110) and (1-10) directions under polarizing light microscope (with average twin spacing of 1 μm). Although we have carried out measurements in our all crystals, we will mainly concentrate on the crystal for PO $=0.03$ % having dimension, $D=1.16\times0.54\times0.36$ mm^3, and $T_{c0}=95.65$ K and ΔT_{c0} (10–90%)$\leqslant$300 mK. Resistance was measured by the standard four-probe method using an ac technique with a frequency of 17 Hz and at a current density of 0.06 A/cm^2. The current was sent parallel to the ab plane. We used a low-noise ratio transformer to increase the sensitivity of the measurement. The contact pads were gold-sputtered and gold wires were attached with silver epoxy and sintered at 320 °C for 2 h in flowing oxygen. The contact resistance was comparable to the sample resistance. The current contacts were placed on both polished sides of the crystal for uniform current distribution whereas voltage contacts were made onto the surface. I-V measurements were performed using dc current and the current direction was reversed at each value to cancel thermal voltages. Magnetization measurements were performed in a superconducting quantum interference device magnetometer (MPMS) with a 3 cm scan length to avoid field inhomogeneity.

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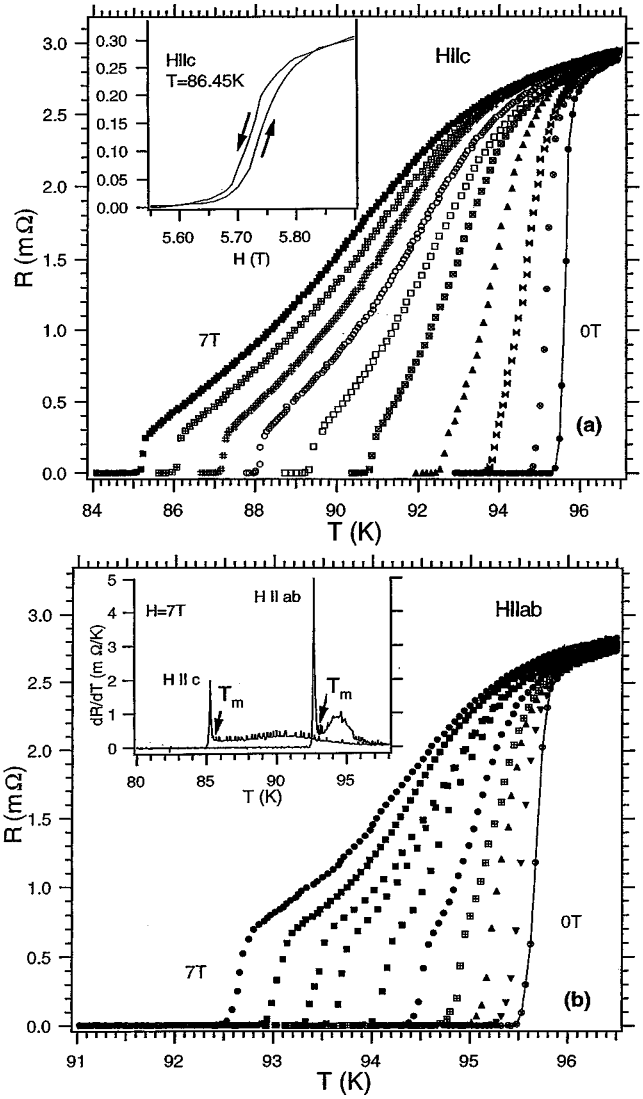

FIG. 1. (a) Resistive transition in magnetic fields of 0, 0.1, 0.5, 1, 2, 3, 4, 5, 6, and 7 T for $H\|c$ in a twinned $NdBa_2Cu_3O_{7-y}$ crystal with $PO_2=0.03\%$. Inset shows the field dependence of the resistance at $T=86.45$ K for $H\|c$ displaying the hysteresis behavior at H_m. (b) Resistive transition in magnetic fields of 0, 0.1, 0.5, 1, 2, 3, 4, 5, 6, and 7 T for $H\|ab$. Inset shows the determination of T_m from inflection peak of dR/dT at $7T$ for $H\|c$ and $H\|ab$.

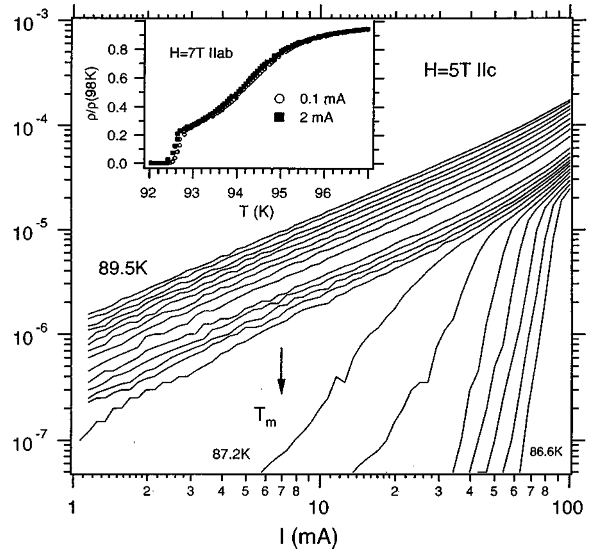

FIG. 2. I-V characteristics in a field of 5 T for $H\|c$ at $T=86.6-0.1$ K interval to 87.7 K and 0.2 K interval to 89.5 K. The corresponding T_m at 87.2 K is shown in the figure. The inset shows the resistive transition in a magnetic field of 7 T for $H\|ab$ for two current densities.

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the resistance for NdBCO single crystal with $PO_2=0.03\%$ in magnetic fields up to 7 T for $H\|c$ and $H\|ab$. A distinct kink in the resistance near $R/R_n \sim 15\%$ for $H\|c$ and $\sim 30\%$ for $H\|ab$ is observed where the resistance drops sharply to zero. This characteristic kink behavior has been attributed to a first-order melting transition of a vortex solid to a vortex liquid. In the inset of Fig. 1(b), we have demonstrated the sharpness of the kink for $H=7$ T for both field orientations by plotting dR/dT as a function of temperature. The kink is extremely sharp with a temperature width ΔT_{kink} (10–90 %)$\leqslant$150 mK at 7 T compared to 300 mK at zero field. The height of the resistance at the kink almost remains constant from 1–7 T, and increases rapidly below 0.5 T and finally disappears. One of the possible reasons of this behavior is the residual sample inhomogeneity that becomes active approaching T_c. We did not observe the shoulder feature in resistance for all measured field values exhibiting any influence of twin boundary pinning as found in YBCO crystal.[4] Though similar kink feature is observed for $H\|ab$ but at higher temperature as shown in Fig. 1(b), the height of resistance at the kink is almost twice than that for $H\|c$. In contrast to twinned YBCO crystal,[4] the height of the resistive kink feature in NdBCO is found to be dependent on field orientation. For $H\|ab$, the kink remains sharp to the highest applied field of $H=7$ T. We carried out the measurements of hysteresis in resistance near the kink region as a function of field at several temperatures as shown in the inset of Fig. 1(a) at one temperature, $T=86.45$ K for $H\|c$. We observed a pronounced hysteresis of about 200 Oe and a sharp drop in the resistance at $H_m=5.73$ T that is exactly consistent with the magnitude of drop in resistance at H_m as shown in Fig. 1(a). While we observed a symmetric hysteresis for $H\|c$, a slightly asymmetric and sharp hysteresis was observed for $H\|ab$. We observed a hysteresis in temperature cycling also at a constant field that rules out the effect due to pinning. The hysteresis and a sharp drop in resistance strongly support that the transition at H_m or at T_m is indeed first-order.

We have shown the influence of current density on the melting transition in the inset of Fig. 2 for two current densities, $I_s=0.06$ A/cm^2 and 1.2 A/cm^2 at $H=7$ T for $H\|ab$ plane. The height of the kink remains constant, however, T_m was found to shift downward only by 0.05 K for higher current density. In order to have more insight into the current dependency on the melting transition, we show I-V curves in Fig. 2 at $H=5$ T for $H\|c$ recorded with a temperature interval of 0.1 K over a wide temperature range near the kink region. The I-V curves display an Ohmic behavior above the kink at 87.2 K. In the narrow transition region of the kink (within 200 mK), the I-V curves show characteristic S-shape curves displaying a drastic change in the vortex state. At low temperature, below 87 K, the I-V curves exhibit concave

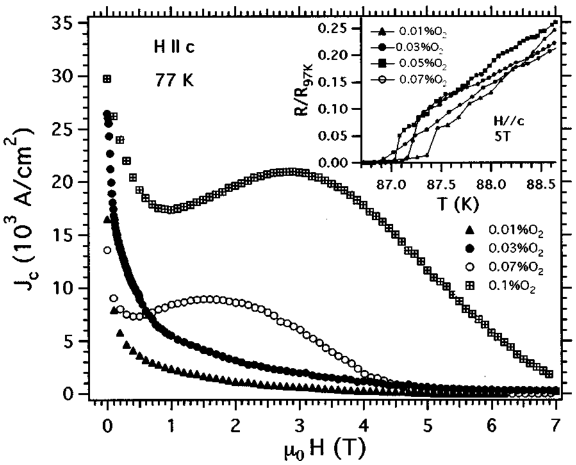

FIG. 3. Field dependent critical current density, J_c at $T=77$ K for crystals grown in various oxygen partial pressure for $H \| c$. Inset shows the resistive transitions for $H \| c$ in these crystals exhibiting a crossover from a first-order melting to a continuous transition at $H=5$ T.

downward curvature arising from the pinned nature of the vortex solid and a finite critical current. This characteristic sharp change in I-V curves strongly support the occurrence of a first-order transition from a liquid to an ordered vortex solid state.

In Fig. 3 we show the field dependence of J_c at 77 K calculated using Bean critical state model for four crystals grown in different PO_2 (0.01–0.1%). J_c curves display a sharp fall with increasing field for $PO_2 \leqslant 0.05\%$ without exhibiting peak effect in the intermediate temperature region as observed in other NdBCO crystals.[8–10,12] However, a well-defined peak effect develops for $PO_2 \geqslant 0.07\%$. It is noted that all these crystals are similarly twinned as $PO_2 = 0.03\%$. The resistive transitions for $H=5$ T applied parallel to the c axis show a smeared first-order transition for $PO_2=0.01$, however a distinct transition for 0.03 and 0.05% as shown in the inset of Fig. 3. The transition becomes continuous for $PO_2 \geqslant 0.07\%$. It may be noted that for $PO_2 = 0.01\%$, the crystal becomes inhomogeneous where as the substitution effect becomes prominent for $PO_2 = 0.07\%$. This suggests that pinning due to either Nd/Ba substitution or oxygen defects are absent or extremely small in the present crystal. The absence of a peak in J_c rules out the pinning due to twin boundary for $PO_2 = 0.03\%$. Therefore the interaction between the generic point defects and twin boundaries plays a major role to suppress the melting transition for $PO_2 \geqslant 0.07\%$.

In order to have a clear comparison, we show the temperature-dependent resistive transitions for $PO_2=0.05$ and 0.07% at various field values in Figs. 4(a) and 4(b), respectively. A distinct kink in resistance is visible for $PO_2 = 0.05\%$ in all measured field values. The inset shows the marked hysteresis in temperature sweep near the melting transition at $H=4$ T. Figure 5 shows the I-V curves over a wide temperature range at $H=5.5$ T displaying a sudden change in the slope as observed for $PO_2 = 0.03\%$. However, the transition for $PO_2 = 0.05\%$ is not as sharp as 0.03%. This is due to the increase of PO_2 that initiates a very small substitution of Nd ions at the Ba sites. The transition becomes

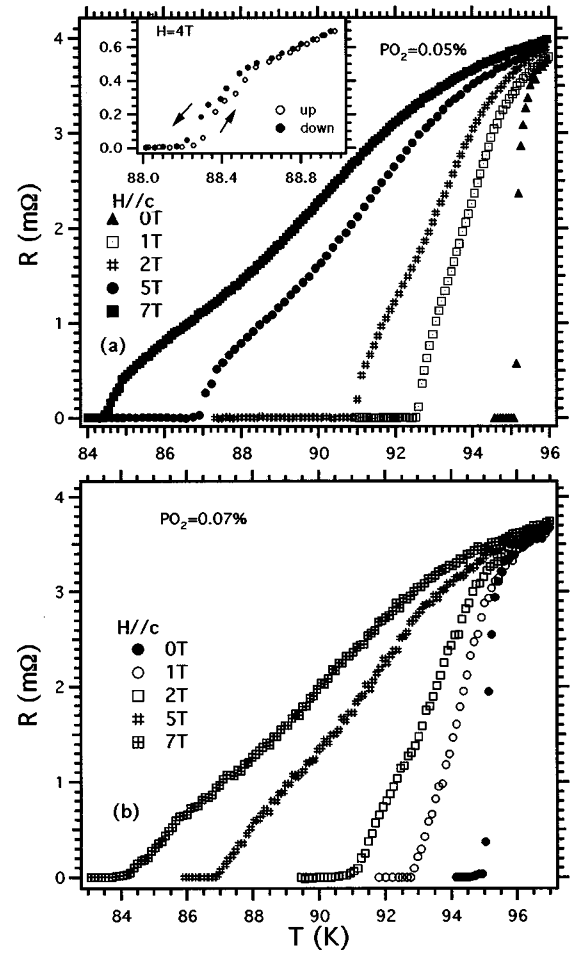

FIG. 4. (a) Resistive transition in magnetic fields for $H \| c$ in a twinned $NdBa_2Cu_3O_{7-y}$ crystal with $PO_2 = 0.05\%$. Inset shows the hysteresis in temperature for $H=4$ T applied parallel to the c axis near the melting transition region. (b) Resistive transition in magnetic fields for $PO_2 = 0.07\%$.

continuous as PO_2 is further increased to 0.07% as shown in Fig. 4(b). This is consistent with the first appearance of peak effect in J_c for $PO_2 = 0.07\%$. The peak effect becomes very pronounced for $PO_2 = 0.1\%$ where major substitution takes

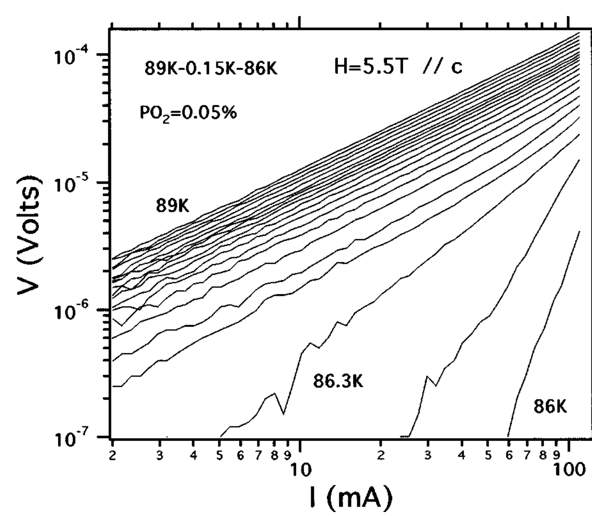

FIG. 5. I-V characteristics in a field of 5.5 T for $H \| c$ for $PO_2 = 0.05\%$. The corresponding T_m is shown in the figure.

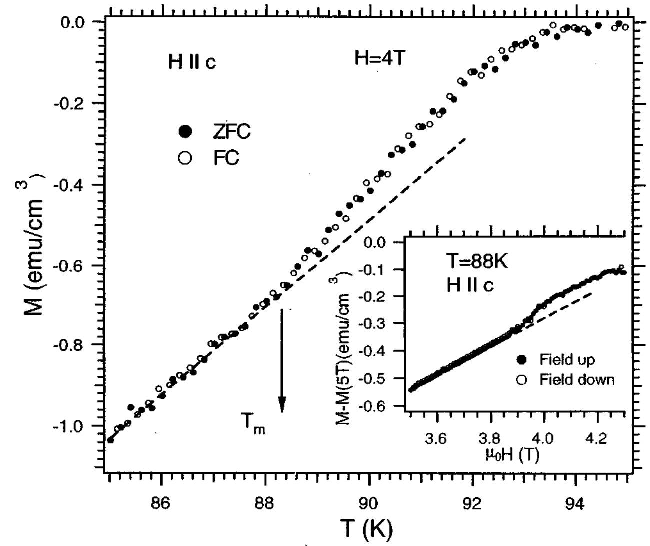

FIG. 6. Temperature dependence of the magnetization at 4 T for $H \| c$ for zero-field-cooled (ZFC) and field-cooled (FC) cases for $PO_2 = 0.03\%$. The dotted line represents a linear extrapolation of the low-temperature variation. The inset shows the M-H curve at 88 K displaying a jump. The dotted line represents a linear extrapolation of the low-field variation.

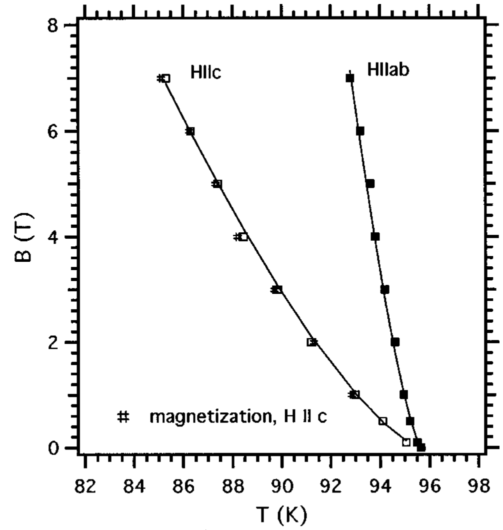

FIG. 7. The magnetic phase diagram of the melting transition for $H \| c$ and $H \| ab$ for $PO_2 = 0.03\%$. The transition from the magnetization jump has also been shown. The solid lines are scaling for melting transition as discussed in the text.

place and acts as pinning centers giving rise to high J_c.

Now we will discuss the thermodynamic evidence of the first-order melting transition in crystal with $PO_2 = 0.03\%$. A distinct change in slope in magnetization is noticed in the temperature dependence of the magnetization as shown in Fig. 6 for a field of 4 T and is consistent with our resistance measurements. In the inset of Fig. 6, we show the M-H curve at $T = 88$ K displaying a pronounced jump in the reversible magnetization, a true thermodynamic behavior of a first-order transition. The jump in magnetization was found for several temperatures and observed consistently above $H \geqslant 1$ T in both field increasing and decreasing cases. The position of the jump is sharp and consistent with the temperature and field at which kink in resistance was observed on the same crystal. This is in agreement with the recent observation of a thermodynamic evidence of a flux-line lattice melting in YBCO crystal.[3]

In Fig. 7 we show the magnetic phase diagram from resistance and magnetization measurements. The melting transitions from resistance curves were determined plotting dR/dT vs T as shown in the inset of Fig. 1(b) and from magnetization jump for magnetization. A very good coincidence was observed establishing a fact that the magnetization jump and resistance kink are the manifestation of the same first-order flux-line lattice transition. The melting line is well described by a power law of the form $H_m(T) = 155(1 - T/T_c)^{1.4}$ and $H_m(T) = 910(1 - T/T_c)^{1.38}$ for $H \| c$ and $H \| ab$, respectively, using T_c value of 95.65 K. The power-law exponent is theoretically predicted as 2 for melting transition in low fields,[15,16] but should be approaching unity for higher fields. Hence the above value is reasonably good for the melting of flux-line lattice. The ratio of the prefactors for above both directions yields a mass anisotropy, $\gamma = (M/m)^{1/2} \sim 5.9$ and this value is comparable or slightly lower than the value for YBCO single crystal.[17]

In order to test the melting transition in view of the Lindemann criterion of vortex lattice melting, we used the following expression[15] for the Lindemann constant c_L which successfully explains the melting transition in YBCO crystal:[1]

$$c_L^4 \approx \{[B_m/H_{c2}(T_m)]/(1 - T_m/T_c)\}(G_i \gamma^2/5.6),$$

where B_m is the melting field, H_{c2} is the upper critical field, and G_i is the Ginzburg parameter $16\pi^2\kappa^4(k_B T_c)^2/\Phi_0^3 H_{c2}(0) \sim 4.6 \times 10^{-5}$ with $\kappa \sim 55$ and $H_{c2}(0) \sim 287$ T (using $dH_{c2}/dT \sim -3$ T/K for $H \| ab$). Using $B_m = 4$ T, $T_m = 88.2$ K, $T_c = 95.65$ K, and $H_{c2} \sim 22$ T, we obtain $c_L \sim 0.16$ which is in excellent agreement with the theoretical prediction. It is noted that the values of $dH/dT \sim -3$ T/K for $H \| ab$ and $dH/dT \sim -1$ T/K for $H \| c$ are quite different than those of YBCO crystal displaying a much stiffer melting line in NdBCO crystal. Considering higher T_c with sharp ΔT_c, absence of flux pinning due to Nd/Ba substitutional defects, a distinct first-order melting transition, and a lower γ value we conclude that this material maintains nearly stoichiometric Nd-123 structure.

The influence of correlated disorder such as twin boundaries on the melting transition has been studied by many authors.[1,5,18] Twin boundaries are known to suppress the melting transition promoting disentanglement, stabilizing a liquid of vortex lines, and establishing the c-axis correlation of vortex lines.[18] The velocity correlation is lost when the sample thickness coincides with the vortex cutting length l_c which is typically less than 100 μm for a twinned crystal. In view of the above, we speculate that (i) the thickness of our crystal is too large (0.36 mm) to promote vortex correlation by twin boundaries to influence on the melting transition and consequently on the kink height and (ii) the twin boundary pinning is either too weak or absent for $H \geqslant 0.2$ T to influence the melting process. We conclude that in contrast to YBCO in which a Bose-glass transition was recently observed by Grigera *et al.*,[19] the presence of twin boundaries alone does not suppress the melting transition in high quality NdBCO single crystal in high-field region. Recently, first-order-like melting transition was observed in twinned YBCO crystal by specific heat[19] and from the resistance[19] measurements in the high-field ($H \geqslant 6$ T) region only for $H \| c$. Another suspect is the misalignment of the angle with respect to $H \| c$ where the influence of twin boundaries is reduced.[4] The accuracy for alignment of angle for magnetization measurements is certainly less than $\sim 0.5°$ for $H \| c$ in which we observed the thermodynamic signature of the first-order transition at the same field at which kink in resistance was observed.

IV. CONCLUSION

In summary, we have shown that the first-order melting transition occurs in twinned NdBCO single crystal, a previously thought intrinsic nonstoichiometric system, displaying a kink in resistance and jump in magnetization. The melting transition is distinctly observed in the crystal with an optimal oxygen partial pressure up to 0.05% during growth and becomes continuous for higher values. We obtain mass anisotropy, $\gamma = 5.9$, and a physically reasonable value of Lindemann constant, $c_L = 0.16$, consistent with the melting criterion. We observed the hysteresis in magnetoresistance near the kink region. The magnetic phase diagram obeys the melting scaling. We obtain a higher melting line in NdBCO crystal in comparison to YBCO. This is an interesting observation of a first-order melting transition in a system other than YBCO with 123 stoichiometry containing magnetic rare-earth ions. Our results clearly demonstrate NdBCO crystal, grown by a special flux method, as a ''clean'' superconductor.

ACKNOWLEDGMENTS

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