Sequential vortex states upon lattice melting in untwinned single-crystal $YBa_2Cu_3O_7$

Takekazu Ishida and Kiichi Okuda
Department of Physics and Electronics, Osaka Prefecture University, Sakai, Osaka 593, Japan

Hidehito Asaoka
Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki 319-11, Japan

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Simultaneous measurements of the ac susceptibility and the dc magnetization of an untwinned single-crystal $YBa_2Cu_3O_7$ have been carried out to clarify the nature of vortex lattice melting. The first-order melting transition appears as a jump in the dc magnetization. Evidence for effectual pinning at temperatures above the melting transition is obtained as a sharp transition in χ' and a sharp peak in χ'' of width 0.2 K in 10 kG. In addition, a precursor lattice softening phenomenon can be seen in χ' and χ'' at temperatures below the melting temperature. This sequential change is more visible when the ac susceptibility is given in the $\chi'-\chi''$ representation. It has been confirmed that there exists a melting transition of $YBa_2Cu_3O_7$ even below 10 kG. [S0163-1829(97)06133-X]

An exotic phase boundary in the mixed state of high-T_c superconductors has attracted considerable interest in recent years. The vortex lattice melting of the first-order nature was predicted in the early stage of high-T_c researches.[1] Recently, the local-Hall-probe measurements detected the abrupt jump in the magnetization in $Bi_2Sr_2Ca_2Cu_2O_8$ in fields lower than 0.038 T, where the melting transition terminated at a critical point.[2] The entropy change ΔS per vortex per layer increased from zero (at the critical point) to $6k_B$ as the temperature increased to the critical temperature T_c.[3] A characteristic feature of the melting transition is somewhat different between $Bi_2Sr_2Ca_1Cu_2O_8$ and $YBa_2Cu_3O_7$. An untwinned crystal $YBa_2Cu_3O_7$ is supposed to be one of the cleanest systems among various high-T_c cuprates. The resistivity drop,[4] the magnetization jump,[5,6] and the calorimetric measurement[7,8] have been reported by using $YBa_2Cu_3O_7$ crystals. A phase boundary of $H=997(1-T/T_c)^{1.36}$ in kG units was not consistent with the first-order transition but consistent with the critical exponent $2\nu=1.33$ of a three-dimensional XY model. Contrary to $Bi_2Sr_2Ca_1Cu_2O_8$, the first-order melting transition of the latent heat $\Delta S\simeq 0.7k_B$ was evident in fields higher than 10 kG. The disappearance of the latent heat was accompanied by the broadening of the resistive transition. Welp *et al.*[6] explained it as the dominance of single vortex pinning by the residual pinning centers below 10 kG because the number of vortices decreases proportional to the field. In addition, a critical point has not been observed for the melting transition of $YBa_2Cu_3O_7$. It is a mysterious open question why evidence for the first-order melting transition was missing in so-called clean $YBa_2Cu_3O_7$ single crystals in fields lower than 10 kG.

The alternative of doing thermodynamic measurements in a dynamic probe for sensing a sequential change in a vortex state. Since the melting is defined as the varnishing of the shear modulus c_{66}, it is expected to occur a lattice softening as a precursor to the melting phase transition. Pippard[9] discussed the softening effect for a classic superconductor. Compared to the dc measurements, the ac response is sensitive to a subtle change in the sample magnetization whenever a pinning force is active even if weak. Kwok *et al.*[10] argued that the peak effect of the critical current density J_c can be interpreted as a precursor to vortex lattice melting in a twinned single-crystal $YBa_2Cu_3O_7$. They described that the peak effect was due to enhanced vortex pinning by the twin boundaries and disappeared when the field was declined by 4° from the c axis. Giapintzakis *et al.*[11] attributed the onset of the ac susceptibility peak of untwinned $YBa_2Cu_3O_7$ to a vortex lattice melting. They obtained $H=5830(1-T/T_c)^{1.85}$ for a melting line defined as the onset of the ac susceptibility in the field range below 10 kG. It is not *a priori* to answer a question of whether the peak effect appeared in the ac susceptibility is assigned to a melting transition or to a preceding lattice softening. One implicitly assumes that the vortex pinning does not work in a vortex liquid state. It might be useful to compare the ac susceptibility with the dc magnetization for clarifying the mutual relationship upon melting. In this paper, we describe the melting phase transition of an untwinned single-crystal $YBa_2Cu_3O_7$ in the dc field range of 0–50 kG by means of the ac susceptibility and the dc magnetization.

A high-purity single crystal of $YBa_2Cu_3O_7$ was prepared by a flux method in a Y_2O_3 crucible.[12] Annealing was carried out at 550 °C for 7 days in 5-atm oxygen pressure under a unidirectional force to remove the twin boundaries. We cut the twinned parts of the crystal by using a wire saw. The polarization-microscope observation indicated that the crystal was completely free from twin boundaries. The lattice parameters were $a=0.3819$ nm, $b=0.3890$ nm, and $c=1.1680$ nm. The crystal used for the magnetic measurements was 1.0×0.5×0.18 mm in size and 0.6 mg in weight. The complete detwinning was also confirmed by a reciprocal-lattice image of an x-ray precession camera.

The ac susceptibility was measured using a homemade susceptometer. In the ac susceptibility $(\chi'-i\chi'')$ measurements the amplitude and the frequency of the ac field were 1 G and 390 Hz, respectively. Both dc and ac fields were applied parallel to the c axis. We also carried out the ac susceptibility measurements when the c axis is declined by 5°

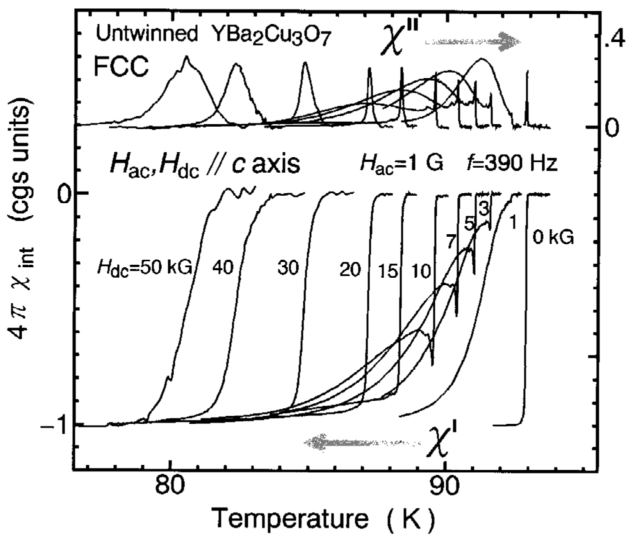

FIG. 1. Internal complex susceptibility χ', χ'' of the untwinned $YBa_2Cu_3O_7$ as a function of T under various dc fields H_{dc} in field-cooled-upon-cooling (FCC) measurements. The dc and ac fields are applied in parallel to the c axis.

from the ac and dc fields. The ac susceptometer was calibrated using a Nb sphere at 4.2 K. We corrected the ac susceptibility for the demagnetizing factor $N=0.678$ of our untwinned sample[13] by using the converting formulas between internal and external complex susceptibilities.[14] The dc magnetization was measured as a function of temperature by using a SQUID (superconducting quantum interference device) magnetometer (Quantum Design MPMS-XL). The field was applied parallel to the c axis. For direct comparisons between the dc magnetization and the ac susceptibility, the SQUID magnetometer was also used to measure the ac susceptibility.

We carried out the ac susceptibility measurements up to 50 kG to investigate a phase diagram. The transition curves in 5 and 7 kG of the ac susceptibility were different among the field-cooled-upon-cooling (FCC), field-cooled-upon-warming (FCW), and zero-field-cooled-upon-warming (ZFC) measurements. In any case, however, we found a peak effect near the onset of the ac susceptibility. In what follows, we concentrate on the FCC measurements for the mutual comparisons of the ac susceptibility. In Fig. 1, we show complex susceptibility χ', χ'' of the untwinned $YBa_2Cu_3O_7$ as a function of T under various dc fields in FCC measurements. The dc and ac fields are applied in parallel to the c axis. The susceptibility is corrected for the demagnetizing factor.

In Fig. 1, there are the three different classes of the χ'' peaks in the 5, 7, and 10 kG fields. In the 10 kG field, for example, one finds (i) a broad χ'' peak at 85 K, (ii) a tiny χ'' peak at 89.4 K, and (iii) a sharp χ'' peak at 89.6 K. We interpret these three χ'' peaks as follows. (1) The broad χ'' peak appearing at lower temperatures arises from weak bulk pinnings when vortices form a lattice. This class of the χ'' peak is only seen in the dc fields below 10 kG. (2) The tiny peak of χ'' seen in the 5, 7, and 10 kG measurements may result from enhanced pinnings due to a vortex lattice softening. Correspondingly, there is a temperature regime where the diamagnetism is weakened as T decreases in the 3, 5, 7, and 10 kG fields. (3) The sharp χ' transition and the corresponding χ'' peak appear in the narrow temperature range near the susceptibility onset. The magnitude of the diamagnetic transition gradually increases to -1 and the transition breadth becomes wider as H_{dc} increases. Contrarily to Giapintzakis *et al.*,[11] we assign this transition to a melting transition rather than a softening peak (see below).

A typical change in the ac susceptibility during a superconducting transition appears as a diamagnetic transition in χ' and a loss peak in χ'' under either zero or finite dc field. A superconducting transition of a homogeneous superconductor often forms a simple domed trajectory in the $\chi'-\chi''$ chart. Either Bean critical-state model, Kim critical-state model, an average-conductivity model, a weakly-connected-loop model, or a diffusive model qualitatively predicts this.[14] The tracing of the trajectory is governed by a single or a few parameter(s), i.e., a critical density J_c in the case of Bean model. In Fig. 2, we show the $\chi'-\chi''$ chart of the untwinned $YBa_2Cu_3O_7$ under various different H_{dc} (FCC). The plot is dome shaped for data taken under 0, 20, 30, 40, 50 kG while it has a three-step structure for 1, 3, 5, 7, 10, 15 kG. (1) At lower temperatures, the behavior is specified by a bulk pinning and more or less similar to that expected for Bean critical state model[15] or a diffusive model.[16] In other words, a sample state is well described by a single parameter such as a critical current density J_c. (2) At intermediate temperatures, the data point almost traces back the lower-temperature branch. A gradual deviation from the lower-temperature branch can be seen for the 7 and 10 kG data. This may be due to a lattice softening and a plastic deformation. (3) At higher temperatures, a trajectory deviates appreciably from the lower-temperature branch in a narrow temperature regime and forms a partial dome. The partial dome shows the evolution to a full dome in fields above 20 kG. Giapintzakis *et al.*[11] considered that the sharp peak of χ'' arises from the synchronous trapping and the enhancement of J_c accompanying a lattice softening. It seems to be more plausible to relate this abrupt change to the first-order melting transition of a vortex lattice because of the strong deviation from the lower-temperature branch.

Kwok *et al.*[10] showed that the peak effect of the twinned sample disappeared when the field was declined by 4°. We also carried out the ac susceptibility measurements of the untwinned $YBa_2Cu_3O_7$ in field-cooled-upon-cooling (FCC) measurements when the c axis of the crystal was declined by 5° from the dc and ac fields. The results were very similar to those shown in Figs. 1 and 2, confirming that the peak effect is not specific to vortices aligned parallel to the c axis.

The dc magnetization M_{dc} was measured as a function of temperature under the constant magnetic fields. In Fig. 3, we show the dc magnetization as a function of temperature in 50 kOe (ZFC and FCC). As shown in the inset of Fig. 3, a clear stepwise change of M_{dc} is seen at temperatures in the reversible region. We fit the FCC data at temperatures just below the step by $M_{dc}=-a+bT$. In Fig. 4, we show $\Delta M_{dc}=M_{dc}-(-a+bT)$ as a function of temperature for various H_{dc}. As reported by Welp *et al.*,[6] the magnetization shows a jump and a change in the temperature derivative of the magnetization at the melting transition. The change of the magnetization per unit volume is $4\pi\Delta m \sim 1.6$ G under 50 kG. The application of the Clausius-Clapeyron equation gives a latent heat of $\Delta S \simeq 1.6 k_B$ per vortex per layer. This value is

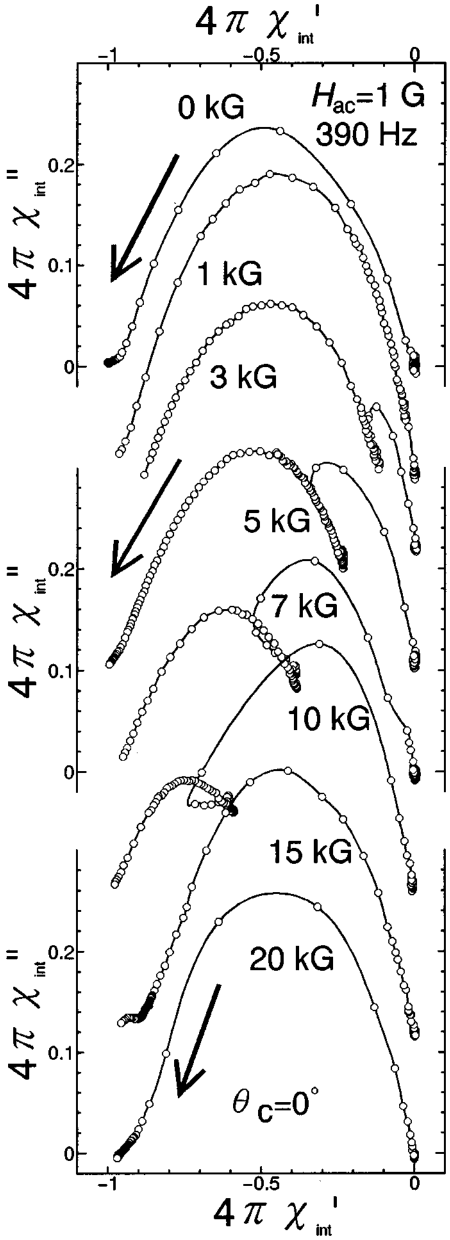

FIG. 2. Internal complex susceptibility of the untwinned $YBa_2Cu_3O_7$ in the χ'' versus χ' representation for FCC measurements. The dc and ac fields are applied in parallel to the c axis ($\theta_c=0°$).

two times larger than that reported in the literature.[6] At lower H_{dc}, a jump in the magnetization is less visible. However, a change in the temperature derivative is clear even in 1 kG. The dotted frames represent the transition region of χ_{ac} under the same fields. The transition region of χ_{ac} is located at temperatures above the magnetization jump.

It is not certain that the χ' dip and the χ'' peak come from the melting transition or from the lattice softening and the resulting synchronous trapping of vortices. To clarify this, we measured the dc magnetization and the ac susceptibility simultaneously. Actually, the measurements are alternatively carried out upon cooling at a constant rate of 0.015 K/min. As shown in Fig. 5, the ac susceptibility $\chi'-i\chi''$ and the dc magnetization M_{dc} were simultaneously measured as a function of temperature under the constant magnetic fields in 10 kOe. In the inset of Fig. 5, we can compare χ', χ'', and ΔM_{dc} without any ambiguity in temperatures. It is clearly seen that the magnetization jump at T_m corresponds to the offset of the sharp diamagnetic transition. We emphasize that the pinning is effective at temperatures (from T_m to T_{HT}) above the melting transition. At temperatures between T_{LT} and T_m, χ' increases as T increases monotonically. This increase is possibly related with the plastic vortex motion reported by Abulafia *et al.*[17] In addition, χ' and χ'' show a tiny dip and a tiny peak at T_s, respectively. We consider that a subtle change at T_s comes from the synchronization effect between vortices and pinning centers. A sequential change of the vortex state is seen remarkably in the ac susceptibility while the dc magnetization only shows a jump and a change in its temperature derivative. One may argue that the transition of χ' and χ'' is not a manifestation of vortex pinning, but may

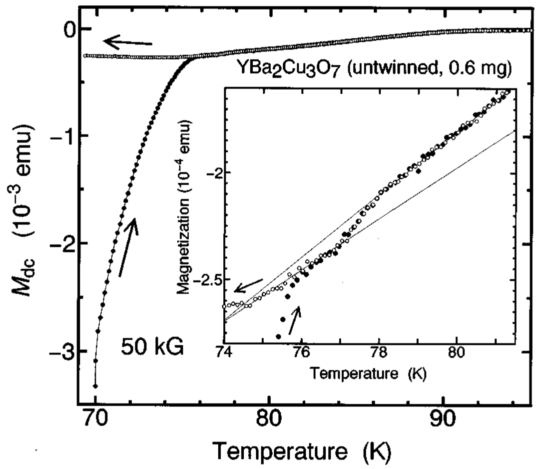

FIG. 3. M_{dc} of the untwinned $YBa_2Cu_3O_7$ as a function of T under a constant field of 50 kG in zero-field-cooled (ZFC) and field-cooled-upon-cooling (FCC) measurements. The dc magnetic field is applied in parallel to the c axis. The inset is an expanded graph at temperatures near the irreversibility onset.

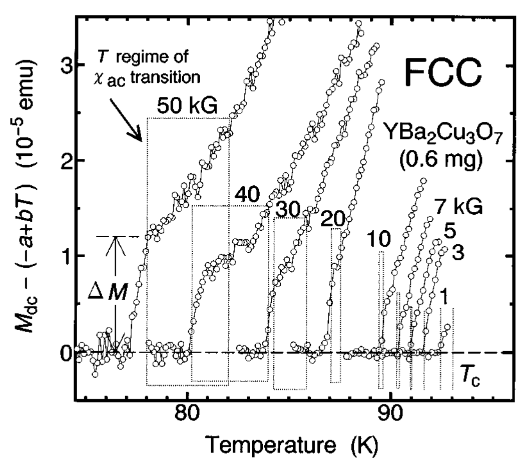

FIG. 4. Relative change in M_{dc} of the untwinned $YBa_2Cu_3O_7$ versus T at temperatures around the magnetization jump. The dc magnetic field is applied in parallel to the c axis. The corresponding temperature regime of the χ_{ac} transition is indicated by a dotted frame.

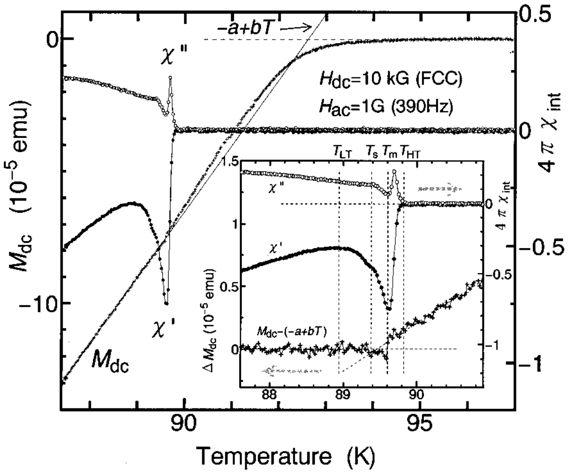

FIG. 5. Simultaneous measurements of M_{dc}, χ', χ'' as functions of temperature in the field of 10 kG. The inset shows a sequential change in the magnetization and the ac susceptibility as a function of temperature.

arise from a rapid change in ohmic resistivity above the melting transition. However, this is not the case because we found a sharp peak in the third-harmonic susceptibility $\chi_3 = \chi_3' - i\chi_3''$ at the corresponding temperatures. Above T_{HT}, both χ' and χ'' are really null within the limit of the SQUID sensitivity, implicating that vortices are liquid and truly free from pinnings. The dc magnetization does not show any anomaly at T_{LT} and T_{HT}, suggesting that these two temperatures do not come from phase transitions but appear as crossover points. A phase boundary represented by the magnetization jump is approximated as $H = 340(1 - T/T_c)^{1.06}$, while the onset of χ' is specified by $H = 740(1 - T/T_c)^{1.27}$ in kG.

In conclusion, vortex lattice melting and softening of an untwinned single-crystal $YBa_2Cu_3O_7$ have been investigated by means of the ac susceptibility and the dc magnetization in the fields of 0–50 kG. The vortex lattice melting is evidenced by a jump in the magnetization and a change in the temperature derivative of the magnetization. From the sharp χ' transition and the χ'' peak in the narrow temperature regime above the melting transition, we discover a crossover change from pinned vortex liquid to free liquid. It is quite interesting to note that the effectual pinning force is active even at temperatures above the melting transition. At temperatures below the melting temperature, there are a tiny χ' dip and a corresponding χ'' peak. Since this regime belongs to a lattice state, we interpret it as a lattice softening and synchronous trapping of vortices as a precursor of lattice melting. The ac susceptibility measurements unveil that a mixed state of $YBa_2Cu_3O_7$ in fields below 10 kG has a clear phase boundary and hence cannot be characterized by the absence of a melting transition.

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