First-order vortex phase transition and anisotropic resistivities in the trilayered $Hg(Re)Ba_2Ca_2Cu_3O_y$ superconductor

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We report observation of the first-order phase transition of vortex lattice in $Hg(Re)Ba_2Ca_2Cu_3O_y$ with critical temperature (T_c) over 100 K from in-plane magnetoresistance. The behaviors of H_{pt} for high-T_c superconductors could be universally understood in terms of a thermally induced sublimation model in wide ranges of the anisotropy factor (γ^2) and T_c, and in multilayered systems including triple CuO_2 layers. The estimated value of γ^2 was extremely small ($\sim$500) in spite of possessing large superconducting layer spacing, which is consistent with the resistivity anisotropy (ρ_c/ρ_{ab}) and is comparable with that in nearly optimally doped $(La_{1-x}Sr_x)_2CuO_4$.

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The vortex phases in the mixed state of high-temperature superconductors (HTSC's) vary complicatedly with temperature and magnetic field reflecting their layered crystal structures. Therefore, physics of the vortex states has fascinated many researchers both from theoretical and experimental aspects. In the early stage, after the discovery of HTSC materials, a number of theoretical concepts and ideas have been proposed for dynamical behavior of vortex matters, such as vortex-lattice melting transition,[1] vortex glass transition,[2] and decoupling transition.[3,4] However, experimentally, the first-order vortex transition (FOT) was first suggested from a resistivity measurement in $YBa_2Cu_3O_y$ (Y123),[5] and then experimental evidence was clearly and thermodynamically confirmed from calorimetric[6] and magnetization[7] measurements in Y123 and $Bi_2Sr_2CaCu_2O_y$ (Bi2212), respectively. Consequently, these experimental developments triggered acceleration of more theoretical and experimental activities for universal understanding of the FOT in HTSC.

Recently, for several HTSC materials possessing single or double CuO_2 planes in their unit-cell structures, a universal temperature dependence of the FOT under $H \| c$ was empirically established[9,8] in terms of the electromagnetic anisotropy parameter ($\gamma^2 = m_c^*/m_{ab}^*$) and the spacing between the CuO_2 blocks (s) as the scaling parameters on the basis of the thermally induced vortex-lattice sublimation model. These reports, however, were limited only for HTSC materials having CuO_2 monolayers or bilayers in the unit cell, because high quality and large single crystals suitable for probing the FOT were not obtained for the compounds with three or more CuO_2 layers. Therefore, the vortex behaviors in trilayered or mulitiple layered cuprates with T_c over 100 K have been unclear. These crystal structures consist of two kinds of CuO_2 planes, pyramidal outer plane (OP) and square inner plane (IP). In $Cu(C)Ba_2Ca_3Cu_4O_y$ (Cu1234), it was suggested that IP and OP were in underdoped and heavily overdoped conditions, respectively, which was reported by Tokunaga *et al*. through ^{63}Cu nuclear magnetic resonance studies.[10] For $HgBa_2Ca_2Cu_3O_y$ (Hg1223), similar studies done on a *c*-axis aligned polycrystalline sample revealed that T_c in the IP was higher than that in the OP's.[11] These cuprates having both IP and OP therefore are also attractive from a viewpoint of vortex physics.

Hg1223 exhibits $T_c \sim 135$ K being the highest among all HTSC materials,[12] and possesses triple CuO_2 planes (two OP's and one IP) in the unit cell. However, difficulty of material synthesis and lack of chemical stability in ambient pressure of the Hg-based superconductors including the Hg1223 phase prevented us from reliable studies on their physical properties using single crystals. In particular, transport properties have not been clearly understood yet, while only a single report[13] has been published so far. We have previously reported that material synthesis problems can be circumvented by partial substitution of rhenium (Re) ions for mercury (Hg) sites in 1223 polycrystalline samples.[14] Additional effect by the Re-doping was also found in the enhanced flux-pinning properties. Very recently, we have succeeded in the single-crystal growth of Re doped $HgBa_2Ca_2Cu_3O_y$ [Hg(Re)1223] by flux method in a $BaZrO_3$ crucible which is less reactive with liquid phase containing Ba-Cu-O.[15] The obtained single-crystals were confirmed to be chemically stable in air and of high quality with very few stacking faults of Hg(Re)1212.

Based on the above backgrounds, the in-plane and out-of-plane resistivities and in-plane magnetoresistance of Hg(Re)1223 single crystals were carefully studied in the present paper in order to obtain its basic physical information, such as electromagnetic anisotropy and vortex dynamic states.

Single crystals were grown by a self-flux method using $BaZrO_3$ crucibles in a quartz ampoule. Details of crystal growth have been reported elsewhere.[15] The typical size of the crystals was approximately $0.8 \times 0.4 \times 0.06^{\| c}$ mm^3. Compositional analysis by a wavelength dispersed x-ray microanalysis (WDX) revealed that the chemical formula of the obtained crystals was approximately $Hg_{0.84}Re_{0.16}Ba_2Ca_2Cu_3O_y$. In order to control the oxygen content of the crystals, post-annealing was performed at 270 °C for 100 h under three different oxygen pressures, $P_{O_2} = 0.01, 0.1$, and 1 atm. In-plane ρ_{ab} and out-of-plane ρ_c resistivity measurements were carried out between 100 and

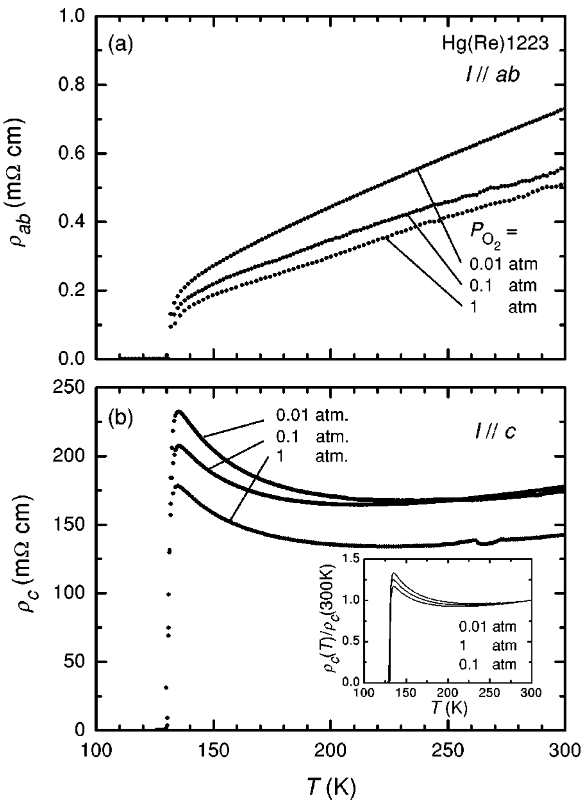

FIG. 1. Temperature dependence of resistivities along the a-b plane (a), ρ_{ab}, and the c axis (b), ρ_c of Hg(Re)1223 crystals that were annealed at three different conditions, in $P_{O_2}=0.01$,0.1, and 1 atm at 270 °C. Inset shows ρ_c normalized by ρ_c (300 K).

300 K using a standard four-probe and a quasi-Montgomery method, respectively, by applying small ac current (1 and 0.1 mA for ρ_{ab} and ρ_c, respectively). The ρ_{ab} measurements were conducted in magnetic fields (H) applied parallel to the c axis up to 90 kOe. Sensitivity limit of the magnetoresistivity measurement was approximately 5×10^{-8} Ω cm.

Figures 1(a,b) show the temperature dependence of ρ_{ab} and ρ_c for postannealed Hg(Re)1223 crystals, respectively. Sharp superconducting transition was observed around 130 K for each crystal, rather independent of the post-annealing conditions. Moreover, as shown in Fig. 1(a), temperature dependence of ρ_{ab} in the normal state showed deviation from a T-linear behavior, which is generic for underdoped HTSC's.[16] Therefore, these three crystals are in slightly underdoped states. The reduction of resistivities in the normal state with an increase of P_{O_2} apparently suggests that carrier-doping is promoted with P_{O_2} and the carrier doping level of the annealed crystal at $P_{O_2}=1$ atm seems to be close to the optimal doping condition.

However, the ρ_c of all the annealed crystals showed metallic temperature dependence down to ~200 K and then changed to semiconducting as shown in Fig. 1(b). This behavior observed in the Hg(Re)1223 is apparently different from that of Re-free Hg1223, which shows semiconducting ρ_c in a whole temperature region below 300 K.[13] The observed ρ_c-T curves were rather insensitive to the annealing conditions and located in a narrow range less than one order of magnitude, while a drastic change of ρ_c from semiconducting to metallic was expected from the systematic decrease of ρ_{ab} due to the difference in the carrier doping level as were in the cases of Y123 (Ref. 17) and $La_{2-x}Sr_xCuO_4$ systems.[18] From these resistivity measurements, anisotropy factor $\gamma^2(=\rho_c/\rho_{ab})$ of Hg(Re)1223 was estimated to be approximately 1000 at 150 K, almost independent of annealing atmosphere. Our previous study[19] revealed that each Re cation, which was partially substituted for the Hg site in Hg(Re)1223, introduced extra oxygen atoms and rigid ReO_6 octahedron was formed. From a thermogravimetric measurement, Serquis *et al.*[20] reported that excess oxygen δ in $Hg_{1-x}Re_xBa_2CuO_{4+\delta}$ (1201) increased drastically with increase of Re content, x, while ranges of δ between two equilibria of annealed conditions, $P_{O_2}=10^{-5}$ and 1 atm at 300 °C, were 0.05, 0.04, and 0.03 for $x=0$,0.10, and 0.15, respectively. Apparently, for the Re-rich samples, a large change of carrier doping levels cannot be expected by the post-annealing at low temperatures below 300 °C. The insensitive behaviors both in T_c and of ρ_c to post-annealing conditions in the present study are probably due to essentially small change of the oxygen content in Hg(Re)1223.

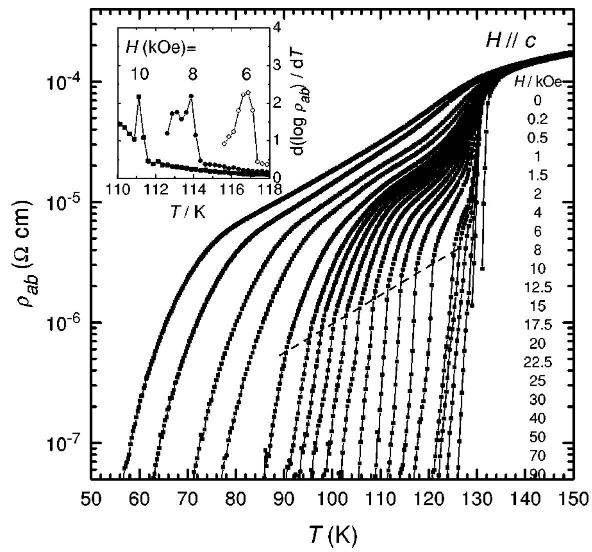

FIG. 2. Temperature dependence of ρ_{ab} of Hg(Re)1223 single crystal annealed at $P_{O_2}=1$ atm under various magnetic fields applied along the c axis. Inset shows temperature dependence of temperature derivative of logarithmic ρ_{ab} for $H=6,8$, and 10 kOe.

Figure 2 shows the temperature dependence of ρ_{ab} of the Hg(Re)1223 crystal annealed at $P_{O_2}=1$ atm under various magnetic fields up to 90 kOe. The superconducting transition was substantially broadened by applying strong fields. For comparison on irreversibility field H_{irr} with Re-undoped Hg1223, zero resistivity point T_{c0} was determined from a criterion reported by Carrington *et al.*;[13] T_{c0} is defined as the temperature at which normalized resistivity ρ/ρ (150 K) decreases to below 10^{-3}. In our case, a value of ρ at T_{c0} corresponds to roughly 10^{-7} Ω cm, and the T_{c0} was found to be systematically decreased down to 56 K under $H=90$ kOe from 130 K at self-field. Using this criterion, the H_{irr} at 77 K was estimated to be ~40 kOe, which is remarkably larger in comparison to $H_{irr}=20$ kOe in the Re-free Hg1223.[13] Since the flux-pinning property was drastically improved by the Re doping in polycrystalline Hg(Re)1223,[14] the observed higher H_{irr} in the present single crystals guarantees incorporation of a certain amount of rhenium. This result is qualitatively consistent with compositional analysis by WDX.

Another important feature in Fig. 2 is the abrupt drop in ρ_{ab} at $10^{-5}\sim10^{-6}$ Ω cm under applied fields below 30 kOe (see the broken line in Fig. 2). To make the details in the resistivity drops clearer, $d(\log\rho_{ab})/dT$ vs T plots under $H=6,8$, and 10 kOe are shown in the inset of Fig. 2. Discontinuous change in the T derivative of log ρ_{ab} can be clearly seen at ~117,~114, and ~111 K for $H=6,8$, and 10 kOe, respectively. These resistivity drops are quite similar to observed behaviors in Bi2212 (Ref. 21) and $(La_{1-x}Sr_x)_2CuO_4$ [La(Sr)214] (Refs. 8,9) systems, thus the observed resistivity drops are clearly considered to originate from the FOT. In the present system, the FOT behavior has been picked up successfully, strongly indicating excellent quality of our crystals with respect to homogeneity and crystallinity. Here, when the normalized resistivity, $\rho/\rho(150\ \mathrm{K})$, was introduced, the resistivity anomalies due to the FOT appeared in a region around ρ/ρ (150 K) $\sim10^{-2}$. (See the broken line in Fig. 2) Sasagawa *et al.*[9] reported that the normalized resistivity of the FOT has a correlation with the magnitude of the anisotropy using the Bardeen-Stephan model.[22] Intriguingly, the value of ρ/ρ (150 K) in the present study ($\sim10^{-2}$) roughly coincides with that in optimally doped or slightly underdoped La(Sr)214 systems[9]. This suggests that the anisotropy factors γ^2 were comparable to each other, indeed consistent with our results of behaviors of H_{irr} as a function of T/T_c (Ref. 15) and ratios of ρ_c/ρ_{ab} for these systems.

In order to quantitatively discuss the behaviors of the FOT in Hg(Re)1223, the phase-transition fields, H_{pt}'s, of the Hg(Re)1223 crystal annealed at $P_{O_2}=1$ atm are plotted as a function of $T_c/T-1$ in logarithmic scale as shown in Fig. 3. This is because our result in Hg(Re)1223 was revealed to well follow the scaling law from other monolayered and bilayered HTSC's and a layered organic supercoductor as proposed by Sasagawa *et al.*[8,9] For comparison with the behaviors of FOT with other cuprates, results in Y123 ($T_c=92.9$ K),[23] optimally doped and heavily overdoped Bi2212 ($T_c=84.2$ and 75.2 K)[21] and slightly underdoped and optimally doped La(Sr)214 ($T_c=36.6$ and 34.9 K) (Ref. 8) are also plotted. In case of the annealed crystal under $P_{O_2}=1$ atm, a series of data was successfully obtained as shown in Fig. 2. For other crystals annealed under $P_{O_2}=0.1$ and 0.01 atm, only few data were obtained, because the crystals were mechanically broken during the measurements. Incidentally, these results for 0.01 and 0.1 atm were roughly overlapped on that for 1 atm annealed crystal. As can be seen, the H_{pt} plot in the nearly optimally doped Hg(Re)1223 was located intermediately between those in the La(Sr)214 samples. As mentioned above, magnitude of γ^2 for Hg(Re)1223 and these two La214 crystals are comparable, which is consistent with the suggestion[8] relevant to a systematic relationship between H_{pt} and the anisotropy factor γ^2 of

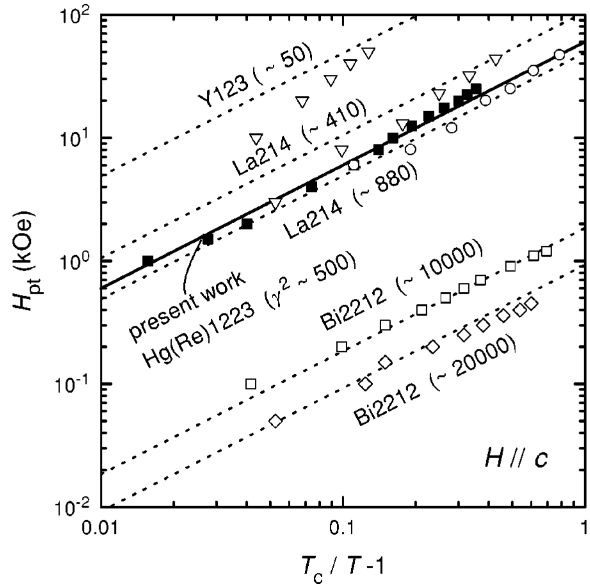

FIG. 3. Plot of H_{pt} (T) vs $T_c/T-1$ for Hg(Re)1223 crystal annealed at $P_{O_2}=1$ atm. A solid line is drawn using Eq. (1) (Ref. 8) with $\gamma^2=500$. For comparison, $H_{pt}(T)$ vs $T_c/T-1$ plots for La(Sr)214 (Ref. 8), Y123 (Ref. 23), and Bi2212 (Ref. 21) were also presented. Each broken line is drawn using Eq. (1).

Y123, Bi2212, and La(Sr)214. This strongly indicates that behaviors of H_{pt} are independent of the number of CuO_2 planes in the unit structure.

For more quantitative discussion on the behaviors of H_{pt} in multilayered HTSC materials, universally scaled lines for the above materials were shown as broken lines in Fig. 3 using an empirical equation[8] as follows:

$$H_{pt}(T)[\mathrm{Oe}]=2.85\gamma^{-2}\ \ s^{-1}(T_c/T-1) \tag{1}$$

This equation is based on the sublimation model of the vortex system, in which even a tilt modulus of the vortex lattice is lost and the vortex lines split up into decoupled vortex-pancake gases above the phase-transition temperature. One can obviously see that the behaviors of H_{pt}'s for these materials having monolayer and bilayer obey Eq. (1). In order to estimate the anisotropy factor γ^2 in the superconducting state in Hg(Re)1223, the obtained H_{pt} data in the present study were fitted by Eq. (1) with $s=9.5$ Å. As represented by a solid line in Fig. 3, the estimated value of γ^2 was found to be ~500, which is comparable with that of nearly optimally doped La(Sr)214, despite the layer spacing of CuO_2 plane, s, in Hg(Re)1223 (~9.5 Å) being much larger than that in La(Sr)214 (~6.6 Å). This can be explained by the improvement of conductivity along the c axis direction at the Hg-O layer by the Re doping.[24] Moreover, it was found that our result followed well the scaling law even for the CuO_2 trilayered system, suggesting that the vortex states in such multi-CuO_2-layered cuprates can be also well explained by the sublimation model proposed for HTSC's having CuO_2 monolayer and bilayers.

In conclusion, we have studied in-plane and out-of-plane resistivities for high-quality Hg(Re)1223 single crystals. The electromagnetic anisotropy factors γ^2 are roughly 1000 in the normal state, independent of annealing conditions, which reflects suppression of range of oxygen contents by Re doping. We have successfully probed the first-order vortex-lattice phase transition from in-plane resistivity measurement under $H\|c$. The normalized resistivity at which the FOT occurs and the behavior of $H_{pt}(T)$ are intriguingly similar to those in slightly underdoped La(Sr)214, which is originated only from similar γ^2 values The behavior of $H_{pt}(T)$ observed in Hg(Re)1223 was found to be also described by the universal sublimation model proposed for Y123, Bi2212, and La(Sr)214, indicating that H_{pt}'s for HTSC are satisfactorily scaled in wide ranges of γ^2, and T_c and in multilayered systems including triple CuO_2 layers.

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[1] D.R. Nelson, Phys. Rev. Lett. **60**, 1973 (1988); A. Houghton, R.A. Pelcovits, and A. Sudbo, Phys. Rev. B **40**, 6763 (1989).
[2] M.P.A. Fisher, Phys. Rev. Lett. **62**, 1415 (1989); D.S. Fisher, M.P.A. Fisher, and D.A. Huse, Phys. Rev. B **43**, 130 (1991).
[3] L.I. Glazman and A.E. Koshelev, Phys. Rev. B **43**, 2835 (1991).
[4] L.L. Daemen *et al.*, Phys. Rev. Lett. **70**, 1167 (1993); Phys. Rev. B **47**, 11 291 (1993).
[5] H. Safar *et al.*, Phys. Rev. Lett. **69**, 824 (1992); W.K. Kwok *et al.*, *ibid.* **69**, 3370 (1992).
[6] A. Schilling *et al.*, Nature (London) **382**, 791 (1996).
[7] E. Zeldov *et al.*, Nature (London) **375**, 373 (1995).
[8] T. Sasagawa *et al.*, Phys. Rev. Lett. **80**, 4297 (1998).
[9] T. Sasagawa *et al.*, Phys. Rev. B **61**, 1610 (2000).
[10] Y. Tokunaga *et al.*, Phys. Rev. B **61**, 9707 (2000).
[11] K. Magishi *et al.*, J. Phys. Soc. Jpn. **64**, 4561 (1995).
[12] A. Schilling *et al.*, Nature (London) **363**, 56 (1993).
[13] A. Carrington *et al.*, Physica C **234**, 1 (1994).
[14] J. Shimoyama *et al.*, Adv. Supercond., 1983 **7**, 287 (1994).
[15] S. Ueda *et al.*, in *Materials for High-Temperature Superconductor Technologies*, edited by M.P. Paranthaman, N.W. Rupich, K. Salama, J. Mannhart, and T. Hasegawa, Symposia Proceedings No. 689 (Materials Research Society, Pittsburgh, 2002), p. 101.
[16] T. Ito, K. Takenaka, and S. Uchida, Phys. Rev. Lett. **70**, 3995 (1993).
[17] K. Takenaka *et al.*, Phys. Rev. B **50**, 6534 (1994).
[18] T. Ito *et al.*, Nature (London) **350**, 596 (1991).
[19] K. Kishio *et al.*, J. Low Temp. Phys. **105**, 1359 (1996).
[20] A. Serquis *et al.*, Phys. Rev. B **63**, 014508 (2000).
[21] S. Watauchi *et al.*, Physica C **259**, 373 (1996); Phys. Rev. B **64**, 064520 (2001).
[22] J. Bardeen and M.J. Stephan, Phys. Rev. **140**, A1197 (1965).
[23] C. Marcenat *et al.*, Physica C **341-348**, 949 (2000).
[24] T. Tanaka *et al.*, J. Phys. C **9**, 1235 (1976).