First-Order Vortex-Lattice Phase Transition in $(La_{1-x}Sr_x)_2CuO_4$ Single Crystals: Universal Scaling of the Transition Lines in High-Temperature Superconductors

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The first-order vortex-lattice phase transition was observed in $(La_{1-x}Sr_x)_2CuO_4$ single crystals ($0.046 \leq x \leq 0.077$). A scaling law, $H_{pt}(T)[\mathrm{Oe}] = 2.85\gamma^{-2}s^{-1}(T_c/T - 1)$, was found to universally hold for the phase transition lines not only in the present system but also in $YBa_2Cu_3O_y$ and $Bi_2Sr_2CaCu_2O_y$. Here γ^2 and s are the anisotropy factor and the superconducting layer spacing. This remarkable scaling provides sound evidence that the first-order phase transition in high-T_c superconductors manifests itself as the vortex-lattice *sublimation* rather than *melting*. [S0031-9007(98)06110-9]

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In the high-temperature superconductors (HTSC), a large thermal fluctuation dramatically changes the equilibrium vortex state and the vortex dynamics. One of the vortex matter physics recently attracting a lot of theorists as well as experimentalists is the vortex-lattice phase transition, which has been experimentally confirmed for its nature to be the first-order thermodynamic transition in $YBa_2Cu_3O_y$ (Y123) and $Bi_2Sr_2CaCu_2O_y$ (Bi2212).

This phenomenon has been generally attributed to the *melting* transition of the vortex lattice; that is, the triangular vortex lattice loses its shear modulus and turns into a vortex liquid state. In Y123, these phase transition lines obtained from resistivity [1,2], magnetization [2–4], and calorimetric measurements [5] were well consistent with each other and have been claimed to be well fitted by a formula derived from the *melting* theory [6]:

$$H_m(T) = H_{m0}(1 - T/T_c)^n, \tag{1}$$

with $H_{m0} \sim 1000$ kOe and $n \sim 1.35$. Equation (1) has been supported also in Bi2212 by magnetization [7–11] and resistivity measurements [11]. However, the magnitude of entropy change associated with this first-order phase transition and its temperature dependence was reported to deviate significantly from what was predicted from the *melting* theory [2,3,8,10,12]. In this context, an alternative explanation such as the *sublimation* of vortex lines has been attempted. In this scenario, even a tilt modulus of the vortex lattice is lost and the vortex lines split up into decoupled vortex-pancake gases on the phase transition. If the transitions are these simultaneous *melting* and *decoupling* processes, the transition line is given by the *decoupling* theory [13] as

$$H_d(T) = H_{d0}(T_c/T - 1). \tag{2}$$

Indeed, several groups have claimed [8,9,14,15] that their results are better fitted by Eq. (2) rather than by Eq. (1). In particular, the *sublimation* scenario seemed to be positively supported in the experiments in which the carrier doping level, or electromagnetic anisotropy, was explicitly taken into account [9,15].

Both the *melting* and the *decoupling* theories are based on the strong anisotropy, which originates from the layered crystal structures intrinsic to all HTSC. In this context, looking at the vortex-lattice phase transition more systematically in materials with various degrees of anisotropy should provide a deeper insight into the nature of the phenomenon. However, the experiments as well as discussions so far have been confined to each particular material system, not covering a wide range of anisotropy.

In this study, we chose $(La_{1-x}Sr_x)_2CuO_4$ (La214), because the value of anisotropy factor $\gamma^2(\equiv m_c^*/m_{ab}^*)$ lies intermediate ($\gamma^2 = 2 \times 10^2 \sim 4 \times 10^3$) [16,17] between those for Y123 ($\gamma^2 = 25 \sim 1 \times 10^2$) [18] and Bi2212 ($\gamma^2 = 3 \times 10^3 \sim 3 \times 10^4$) [19]. In addition, the value γ^2 can be controlled systematically with Sr composition x in La214. Although only one report has shown reasonably clear evidence for the vortex-lattice phase transition in La214 [20], the Sr content in their study was fixed, and hence the above mentioned features with respect to the anisotropy have not been addressed. Then we carefully prepared several La214 single crystals with systematic variation of the Sr composition. In these crystals, magnetization step changes associated with the first-order phase transition were observed both in field-scan and temperature-scan magnetization measurements. It was found that a scaling law, $H_{pt}(T)[\mathrm{Oe}] = 2.85\gamma^{-2}s^{-1}(T_c/T - 1)$, where s is the distance between superconducting planes, could describe the transition lines $H_{pt}(T)$ not only for the present La214 with different γ^2 values but also those for the Y123 and Bi2212 systems. We conclude that the *sublimation* (the simultaneous *melting* and *decoupling*) is the more likely scenario to describe the first-order vortex phase transition universally in all of the HTSC compounds.

$(La_{1-x}Sr_x)_2CuO_{4-\delta}$ single crystals with nominal Sr compositions of $x = 0.05$, 0.075, and 0.080 were prepared by the traveling-solvent-floating-zone method [17]. Crystal orientations were determined by an x-ray back-reflection Laue technique. Rectangular crystals with a typical dimension of $2.0 \times 0.7 \times 3.0$ mm^3, having long edges along $a(b)$ axis (ab crystal) and c axis (c crystal), were sectioned from grown boules by a diamond wheel. The Sr contents x were determined by EPMA (JEOL JXA-8600) to be 0.046, 0.068, and 0.077, respectively. These crystals were sealed in quartz ampoules under controlled initial oxygen pressures ($P_{O2} = 100$, 600, and 700 torr, respectively, for the $x = 0.046$, 0.068, and 0.077 crystals), and annealed at 900 °C for 10 days, followed by rapid quenching to room temperature in order to remove oxygen defects ($\delta \sim 0$) [21]. Both resistive components, ρ_{ab} or ρ_c, were measured, respectively, on ab crystals and c crystals by the ac four-probe technique using a Quantum Design PPMS-6000. The critical temperatures T_c and the anisotropy factor γ^2 were determined as the midpoint of the resistive transition and as the ratio of the out-of-plane to in-plane resistive components at 50 K, respectively. These parameters together with s, the distance between superconducting planes, are summarized in Table I. Temperature-scan magnetization measurements were performed using a SQUID susceptometer (Hoxan HSM-2000X), while field-scan measurements were done using both a dc magnetometer (PPMS-6000, ACMS-option) and a SQUID magnetometer (Quantum Design MPMS, RSO-option). All of the magnetization measurements were performed on c crystals with the external fields of up to 50 kOe applied parallel to the c axis.

Figure 1 shows M-H curves for the $x = 0.046$ crystal at several temperatures. Two H-linear parts are observed in the reversible magnetization, one corresponding to the magnetization of vortex solid, and the other to that of vortex liquid or gas. We defined the phase transition field H_{pt} as the middle point of the transition between the two linear parts. It should be noted that the vortex density increased as the phase changed from the vortex solid into liquid or gas. This is the same behavior observed in Y123 [2–4] and Bi2212 [7–11] and has been regarded as the thermodynamic evidence of the first-order phase transition. The similar magnetization step structure was observed in the temperature-scan measurements, and these independently obtained phase transition lines, $H_{pt}(T)$ and $T_{pt}(H)$, agreed with each other. As shown in the inset of Fig. 1, $H_{pt}(T)$ was located higher than the irreversibility line $H_{irr}(T)$. The disappearance of the first-order phase transition at the higher field regions, as reported in Y123 [4] and Bi2212 [8,9], was not observed in La214 within the field range examined in this study.

TABLE I. Measured characteristic parameters for La214, Y123 [2], and Bi2212 [11] crystals.

Sample	T_c/K	s/Å	ρ_{ab}(50 K)	ρ_c(50 K)	γ^2
La214 (0.046)	25.6	6.6	0.65	1900	2900
(0.068)	34.9	6.6	0.32	280	880
(0.077)	36.6	6.6	0.16	66	410
Y123 [2]	92.9	11.7	⋯	⋯	(50)
Bi2212 [11]	78.3	15.4	⋯	⋯	(10 000)

The similar magnetization anomaly was observed also in the $x = 0.068$ and 0.077 crystals. Shown in Fig. 2(a) are the $H_{pt}(T)$ lines obtained for the three crystals plotted in the H-T phase diagram, where we included the phase transition lines reported for Y123 ($T_c = 92.9$ K) [2] and Bi2212 ($T_c = 78.3$ K) [11]. It was found that $H_{pt}(T)$ shifted toward higher fields as x increased in La214. However, as can be seen among La214, Bi2212, and Y123, the location of $H_{pt}(T)$ in the H-T phase diagram is dependent on T_c of each sample. The present La214 crystals were underdoped ($x = 0.046$, 0.068) to optimum-doped ($x = 0.077$) ones. Therefore, one may attribute the upward shift of $H_{pt}(T)$ with x to the enhancement of T_c. However, such interpretation of the x dependence in $H_{pt}(T)$ is not appropriate in our case as discussed below.

Figure 2(b) is the magnetic phase diagram in which the temperature axis is normalized against the respective T_c. The transition lines shift still upward with an increase of x, strongly suggesting the involvement of other factors to shift the transition lines. It was reported in Bi2212 that $H_{pt}(T)$ shifts upward by reducing the anisotropy [9,15,22]. Since the anisotropy factor γ^2 as shown in Table I decreases similarly with increasing x in La214, the upward trend in $H_{pt}(T)$ with reducing the anisotropy can be regarded as a universal behavior. Furthermore, a clear relationship between γ^2 and $H_{pt}(T)$ is found also among different material systems; the lines for La214 are located between those for Bi2212 and Y123, where γ^2 for Bi2212, La214, and Y123 decreases in this order. Therefore, we can conclude that (1) the first-order vortex phase transition is a universal phenomenon observed in the HTSC covering a wide range of anisotropy, and (2) γ^2

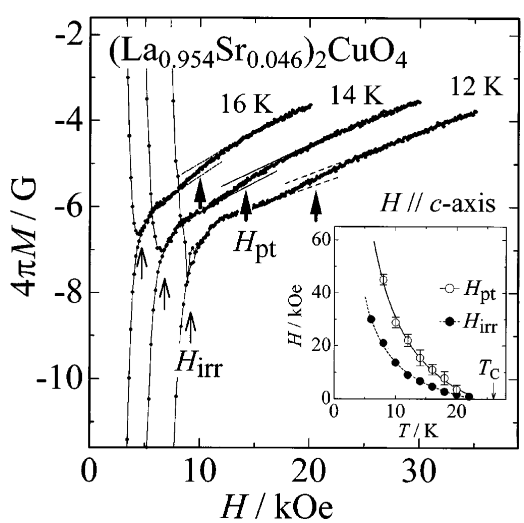

FIG. 1. Field dependence of the magnetization in $(La_{0.954}Sr_{0.046})_2CuO_4$ single crystal at various temperatures. The thick arrows indicate the phase transition fields $H_{pt}(T)$, while the thin arrows indicate irreversibility fields $H_{irr}(T)$. The inset shows the H-T phase diagram obtained from the magnetization measurements. The solid and dashed lines are guides to the eye.

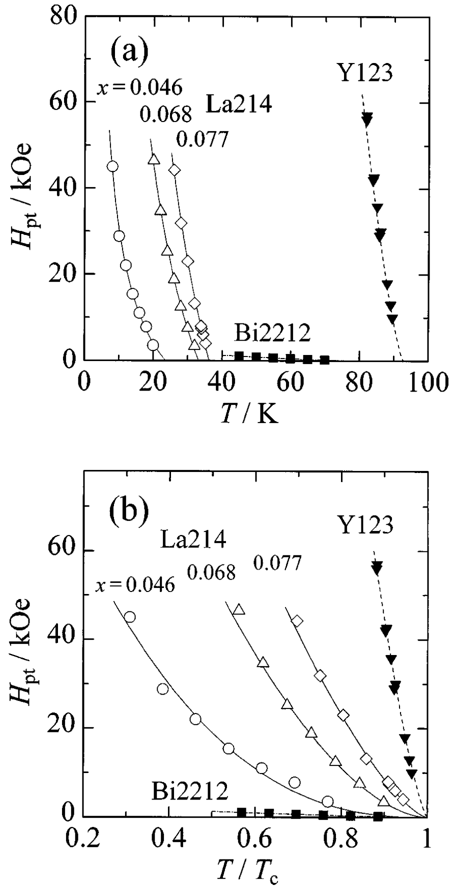

FIG. 2. The phase transition lines $H_{pt}(T)$ for La214 crystals with different Sr compositions. For comparison, the results of Y123 [2] and Bi2212 [11] are also shown. Curves are guides to the eye. (a) The horizontal axis is the normal temperature scale. (b) The temperature is normalized by each T_c.

is an important determining factor of the transition lines in the phase diagram in addition to T_c.

As described at the beginning, the origin of the first-order vortex-lattice phase transition has been proposed to be either the simple *melting* or the simultaneous *melting* and *decoupling* process. We, therefore, attempted to fit the observed $H_{pt}(T)$ to both Eqs. (1) and (2). Although the fitting curves are not shown here, the fits of $H_{pt}(T)$ were fairly good both to $H_m(T)$ and to $H_d(T)$. As for the fitting to Eq. (1), however, the resulted fitting parameter n was found to increase with decreasing x ($n = 1.43$, 1.73, and 2.48 for the $x = 0.077$, 0.068, and 0.046 crystals, respectively). The variation of n with x (or γ^2) is hardly explained within the framework of the existing theories. It is particularly noted that the fitting parameter $n = 2.48$ for the $x = 0.046$ crystal does not fall under the category of the *melting* theory ($n \leq 2$). Therefore, the *sublimation* scenario seems to be more favored in La214. A further support to this scenario, which is more convincing, may be obtained in the following arguments.

The scaling behavior of the phase transition lines was previously reported by our group in Bi2212, for which anisotropy γ^2 was systematically changed by its oxygen content. That is, the phase transition lines fell onto a single line when they were plotted as $\gamma^2 H_{pt}(T)$ vs 1-T/T_c [22]. This scaling should hold if Eq. (1) is the appropriate expression for the transition lines and the prefactor H_{m0} depends almost only on γ^2. In fact, considering $H_{m0} \propto \gamma^{-2} T_c^{-2} \lambda_{ab}^{-4}(0)$ [$\lambda_{ab}(0)$ is the in-plane penetration depth] [6], $\gamma^2 H_{pt}(T)$ vs 1-T/T_c scaling is thought to be valid when $T_c^{-2} \lambda_{ab}^{-4}(0)$ is almost constant. This assumption has been semiquantitatively established by μSR (muon-spin-relaxation) studies which show that T_c is proportional to the muon-spin-relaxation rate σ [$\propto \lambda^{-2}(0)$] and the proportionality is constant in any compounds [23]. Now the question arises whether this kind of scaling holds to different HTSC material systems. Therefore, the plot of $\gamma^2 H_{pt}(T)$ vs 1-T/T_c will serve as a test of the *melting* scenario whether or not applicable as a model for the first-order phase transition. Similarly, because of $H_{d0} \propto \gamma^{-2} s^{-1} T_c^{-1} \lambda_{ab}^{-2}(0)$ in Eq. (2) [13], $\gamma^2 s H_{pt}(T)$ vs $T_c/T - 1$ plots give a test to the *sublimation* scenario.

The results of the test plots for *melting* and *sublimation* are shown in Figs. 3(a) and 3(b), respectively. The values in Table I were used for the plots. In the test plots

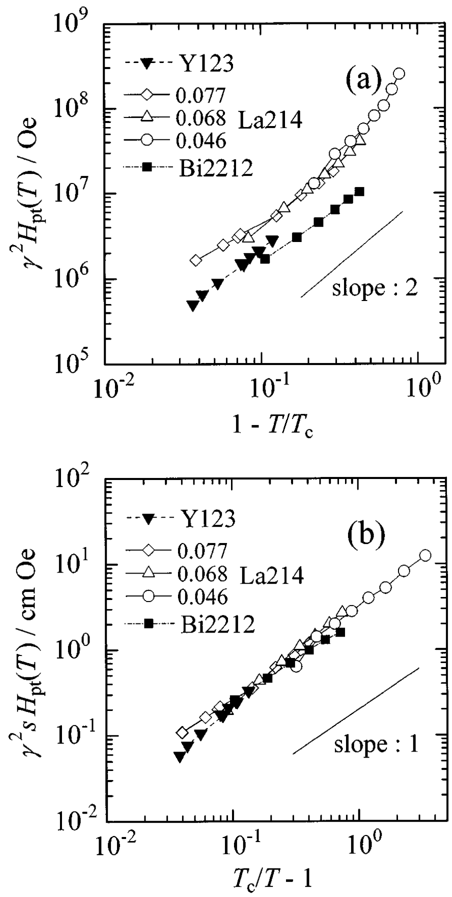

FIG. 3. Test of the universality of the first-order vortex phase transition in HTSC. (a) $\gamma^2 H_{pt}(T)$ vs 1-T/T_c plots testing *melting* transition scenario. (b) $\gamma^2 s H_{pt}(T)$ vs $T_c/T - 1$ plots testing *sublimation* transition scenario.

of the *melting*, the scaling feature was not seen among the different material systems. If we force these lines to be scaled, the values of γ^2 should be changed at least 200%. In this case, nevertheless, the slope of the scaling line, corresponding to n in Eq. (1), ranged from 1.3 to 2.5 as mentioned before. On the other hand, all of the transition lines for the three different material systems fell on a single line in the test plots for the *sublimation* and have a single slope of approximate unity. It is rather amazing that such a scaling behavior of the transition lines universally holds for three material systems having the extreme differences in γ^2, s, and T_c. The reason that the scaling law holds indicates that all of the HTSC materials are composed of CuO_2 planes of the same nature but only with a different coupling constant, which is determined by the interlayer spacing and by the doping level. Then the first-order phase transition can be interpreted to take place when thermal energy surpasses interlayer coupling of the pancake vortices as well as elastic energy of the vortex lattice. In this simultaneous *melting* and *decoupling* picture, pancake-vortex gases above $H_{\rm pt}(T)$ should result in the loss of the superconducting phase coherence in all of the directions. Experimental evidence for such a loss of the phase coherence was found in resistivity measurements in Lorentz force-free configuration [24], namely, significant resistive broadening was seen in the same temperature range, not only in ρ_{ab} but also in ρ_c, when external magnetic fields were applied parallel to the c axis. Recently, a more striking result was reported in multiterminal transport measurements [25], that is, the resistive components both along the a axis and along the c axis disappeared concurrently in the vicinity of the vortex-lattice phase transition. Furthermore, the Josephson-plasma-resonance study, in which direct information of the phase coherence between CuO_2 planes is obtained, has revealed the drastic decrease of the interlayer coherence above $H_{\rm pt}(T)$ [26]. Consequently, the observed remarkable scaling feature in this study, together with several experimental facts reported, strongly supports that the simultaneous *melting* and *decoupling* process is the origin of the first-order vortex-lattice phase transition in HTSC. Based on the *sublimation* scenario, we obtained

$$H_{\rm pt}(T)\,[\mathrm{Oe}] = 2.85\gamma^{-2}s^{-1}(T_c/T - 1) \qquad (3)$$

as a rule of universal application to the transition lines.

In conclusion, we have studied the first-order vortex-lattice phase transition in La214 single crystals with various doping levels and anisotropies. The obtained phase transition lines were compared among different compositions ($x = 0.046$–0.077 in La214) and among different material systems (Y123, La214, and Bi2212) from the viewpoints of both the *melting* and the *sublimation* transitions. This comparative analysis indicated that the underlying mechanism of the first-order phase transition is the simultaneous *melting* and *decoupling* rather than simple *melting*, and the obtained Eq. (3) was proposed to be a material-independent universal expression for the transition lines in HTSC compounds.

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[1] H. Safar *et al.*, Phys. Rev. Lett. **69**, 824 (1992); W. K. Kwok *et al.*, Phys. Rev. Lett. **69**, 3370 (1992).
[2] U. Welp *et al.*, Phys. Rev. Lett. **76**, 4809 (1996).
[3] R. Liang *et al.*, Phys. Rev. Lett. **76**, 835 (1996).
[4] T. Nishizaki *et al.*, Physica (Amsterdam) **282-287C**, 2117 (1997); K. Deligiannis *et al.*, Phys. Rev. Lett. **79**, 2121 (1997).
[5] A. Schilling *et al.*, Nature (London) **382**, 791 (1996); M. Roulin *et al.*, Science **273**, 1210 (1996).
[6] A. Houghton *et al.*, Phys. Rev. B **40**, 6763 (1989); E. H. Brandt, Phys. Rev. Lett. **63**, 1106 (1989); G. Blatter and B. I. Ivlev, Phys. Rev. B **50**, 10 272 (1994).
[7] H. Pastoriza *et al.*, Phys. Rev. Lett. **72**, 2951 (1994).
[8] E. Zeldov *et al.*, Nature (London) **375**, 373 (1995).
[9] B. Khaykovich *et al.*, Phys. Rev. Lett. **76**, 2555 (1996); Y. Yamaguchi *et al.*, *Advances in Superconductivity VIII*, edited by H. Hayakawa and Y. Enomoto (Springer-Verlag, Berlin, 1996), p. 209.
[10] T. Hanaguri *et al.*, Physica (Amsterdam) **256C**, 111 (1996).
[11] S. Watauchi *et al.*, Physica (Amsterdam) **259C**, 373 (1996).
[12] N. Morozov *et al.*, Phys. Rev. B **54**, R3784 (1996).
[13] L. I. Glazman and A. E. Koshelev, Phys. Rev. B **43**, 2835 (1991); L. L. Daemen *et al.*, Phys. Rev. Lett. **70**, 1167 (1993); Phys. Rev. B **47**, 11 291 (1993); R. Ikeda, J. Phys. Soc. Jpn. **64**, 1683 (1995).
[14] R. A. Doyle *et al.*, Phys. Rev. Lett. **75**, 4520 (1995).
[15] Y. Ando *et al.*, Phys. Rev. B **52**, 3765 (1995).
[16] T. T. M. Palstra *et al.*, Phys. Rev. B **41**, 6621 (1990); T. Shibauchi *et al.*, Phys. Rev. Lett. **72**, 2263 (1994); T. Kimura *et al.*, *Advances in Superconductivity VII*, edited by K. Yamafuji and T. Morishita (Springer-Verlag, Berlin, 1995), p. 539.
[17] T. Kimura *et al.*, Physica (Amsterdam) **192C**, 247 (1992).
[18] D. E. Farrell *et al.*, Phys. Rev. Lett. **61**, 2805 (1988); D. E. Farrell *et al.*, Phys. Rev. Lett. **64**, 1573 (1990); K. Takenaka *et al.*, Phys. Rev. B **50**, 6534 (1994).
[19] D. E. Farrell *et al.*, Phys. Rev. Lett. **63**, 782 (1989); J. C. Martinez *et al.*, Phys. Rev. Lett. **69**, 2276 (1992); Y. Kotaka *et al.*, Physica (Amsterdam) **235-240C**, 1529 (1994).
[20] T. Naito *et al.*, Czech. J. Phys. **46**, 1585 (1996).
[21] T. Sasagawa *et al.*, J. Low Temp. Phys. **105**, 1201 (1996); M. Okuya *et al.*, Physica (Amsterdam) **271C**, 265 (1996).
[22] J. Shimoyama *et al.*, Physica (Amsterdam) **282-287C**, 2055 (1997).
[23] Y. J. Uemura *et al.*, Phys. Rev. Lett. **62**, 2317 (1989); Y. J. Uemura *et al.*, Phys. Rev. Lett. **66**, 2665 (1991).
[24] K. Kitazawa *et al.*, Jpn. J. Appl. Phys. **28**, L555 (1989).
[25] D. T. Fuchs *et al.*, Phys. Rev. B **55**, R6156 (1997).
[26] Y. Matsuda *et al.*, Phys. Rev. Lett. **75**, 4512 (1995); Y. Matsuda *et al.*, Phys. Rev. Lett. **78**, 1972 (1997).