Enhanced two-dimensional properties of the four-layered cuprate high-T_c superconductor $TlBa_2Ca_3Cu_4O_y$

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We have measured the temperature dependence of the reversible magnetization of a four-layered cuprate $TlBa_2Ca_3Cu_4O_y$ (Tl-1234) for the $H \| c$ axis, which has the highest superconducting transition temperature T_c of almost 130 K among the four-layered cuprates. The reversible magnetization was analyzed by using the high-field scaling theory and the Hao-Clem model. It was found that in the critical region around T_c, two-dimensional fluctuations dominate. This enhanced 2D behavior can be naturally explained by selective doping model, where doping levels of the inner and outer CuO_2 planes in a unit cell are different. From these analyses, various thermodynamic parameters, such as the coherence length, the penetration depth, the critical fields, and the Ginzburg-Landau parameter were obtained.

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I. INTRODUCTION

Experimentally, it is known that the superconducting transition temperature (T_c) for multilayered high-T_c superconductors does not monotonically increase with the number of CuO_2 layers n. The T_c increases with n up to $n=3$, but then starts to drop at $n=4$.[1,2] This is in sharp contrast to the general expectation that T_c increases while n increases, suggesting that the charge carriers were not distributed equally in every CuO_2 plane within a unit cell.[3]

The inequivalent distribution of the charge carriers in different CuO_2 planes was confirmed by the Cu-NMR experiment[4] for Hg- and Cu-based high- T_c cuprates with $n=3$, 4, and 5. According to this experiment, the local-hole doping levels N_h can be different on the inner square planes (IP) and the outer pyramidal planes (OP). For compounds with $n=3$ where CuO_2 planes in a unit cell consist of one IP and two OP's, N_h(IP) and N_h(OP) are almost the same and both exhibit nearly optimal hole contents. On the other hand, for compounds with $n=4$ or 5, OP is predominantly in an overdoped state while IP remains either optimal or underdoped. Consequently, "microscopic" T_c of the OP and of the IP differ from each other by this selective doping. Thus, the compound with $n=3$ shows the maximum T_c and the compounds with $n=4$ have lower T_c, along with a reduced interlayer coupling. This proposes that a reduction in T_c with increasing n is associated with an increase in $\Delta N_h = N_h(\mathrm{OP}) - N_h(\mathrm{IP})$. Quite recently, the coexistence of the superconductivity and the antiferromagnetism in Hg-1245 was observed in the NMR experiment. This revealed that one of the IP and OP became extremely underdoped.[5]

Tl-based cuprates constitute one of the largest chemical families of high-T_c superconductors, forming two distinct structure series with the general formulas $TlBa_2Ca_{n-1}Cu_nO_{2n+3}$ $[\mathrm{Tl}-12(n-1)n]$ for single-Tl-O-layered and $Tl_2Ba_2Ca_{n-1}Cu_nO_{2n+4}$ $[\mathrm{Tl}-22(n-1)n]$ for double-Tl-O-layered compounds ($n=1$–4). So far it is known that critical current density (J_c) and irreversibility field (H_{irr}) of the single Tl-O layer compounds are relatively higher than that of the double Tl-O layers for $n=1$–3.[6] Therefore for the application, the single Tl-O layer compound is more promising compared to the double Tl-O layer compound. However, these single Tl-O layered compounds have not been studied in detail due to the lack of phase pure samples. The toxicity and high vapor pressure of the Tl_2O_3 has prevented one from synthesizing phase pure Tl-based compounds. Using a high pressure technique, Iyo *et al.* successfully synthesized high quality Tl-1234 with $T_c=128$ K.[7] The T_c of this sample is higher than the T_c of 115 K of the earlier samples.[8] The increase of T_c was due to very careful preparations of a carbon-free precursor. Because of the above-mentioned reasons, even the basic superconducting parameters of a Tl-1234 superconductor, such as the characteristic lengths (ξ and λ), the critical fields (H_c and H_{c2}), and the Ginzburg-Landau parameter κ, have not been reported on the phase pure Tl-1234.

In this paper, basic superconducting parameters are obtained after the analysis of the reversible magnetization of the grain-aligned high quality Tl-1234 in magnetic fields applied parallel to the c axis. Ullah-Dorsey's scaling theory[9] and the Hao-Clem model[10] were used in the analyses. From these analyses, we found that a two-dimensional nature is pronounced for this four-layered Tl-1234, which is a natural consequence of the selective doping.

II. EXPERIMENTS

The Tl-1234 was prepared in high-pressure conditions. The detailed process of sample preparation is given elsewhere.[7] This compound has a tetragonal structure ($P4/mmm$) with lattice parameters $a=b=3.849$ Å and $c=19.005$ Å. The Farrell method[12] was employed in order to make a c-axis-aligned sample. The powder was aligned in an

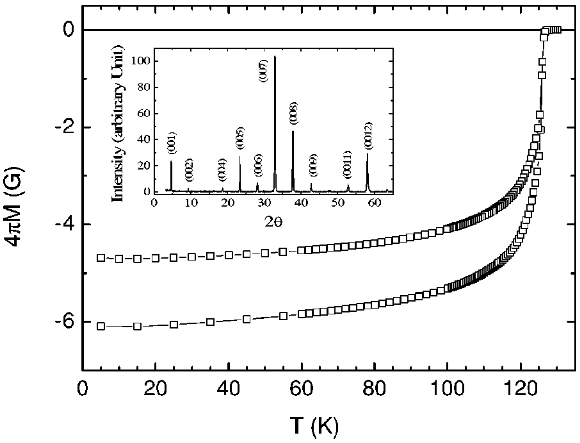

FIG. 1. Temperature dependence of zero-field-cooled (lower curve) and field-cooled magnetization (upper curve) at 10 Oe. Inset: XRD for grain-aligned T1-1234. The (00*l*) reflections are predominant in the XRD pattern, which confirms that these grains are well oriented along the *c* axis.

epoxy with an external magnetic field of 7 T at room temperature. The alignment was confirmed by the x-ray diffraction. All the peaks in the inset of Fig. 1 were indexed as (00*l*), indicating that the grains are well aligned along the *c* axis. Figure 1 shows the low field magnetization at 10 Oe parallel to the *c* axis. The transition temperature T_c was found to be 128 K.

The temperature dependence of the magnetization was measured by using a superconducting quantum interference device magnetometer (MPMS-XL, Quantum design). The zero-field-cooled and field-cooled magnetization for the external field range of $1\ \mathrm{T} \leqslant H \leqslant 5\ \mathrm{T}$ were measured.

III. RESULTS AND DISCUSSION

Figure 2 shows the temperature dependence of the reversible magnetization $4\pi M(T)$ for various magnetic fields applied parallel to the *c* axis. As shown in this figure, the crossover point of the magnetization that is a typically feature of high-T_c, cuprate[11,13–16] was observed. The crossover point occurs at $T^*=125.2$ K and $4\pi M^*(T)=-2.04$ G. and this was due to the enhanced positional fluctuation of vortices, as predicted by Bulaevskii *et al.*[17]

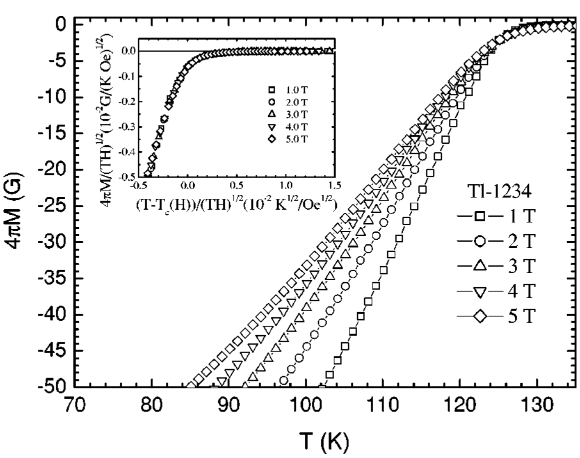

FIG. 2. Temperature dependence of the reversible magnetization in the field range of $1\ \mathrm{T} \leqslant H \leqslant 5\ \mathrm{T}$ applied parallel to the *c* axis. Inset: Two-dimensional scaling of the magnetization around $T_c(H)$.

In the vortex fluctuation model, free energy consists of two parts. One is due to the static structure of vortices and the other is the contribution from the positional fluctuation of vortices. In this framework, the derivative of magnetization is

$$\frac{\partial M}{\partial \ln H} = \frac{\phi_0}{32\pi^2\lambda_{ab}^2(T)}[1-g(T)] \text{ with } g(T) = \frac{32\pi^2 k_B}{\pi_0^2 s} T\lambda_{ab}^2(T), \tag{1}$$

where s is the effective interlayer spacing and ϕ_0 is the flux quantum. The temperature T^* in this model is determined as $g(T^*)=1$ and the corresponding magnetization is

$$M^*(T^*) = \frac{k_B T^*}{s\phi_0} \ln \frac{\eta\alpha}{\sqrt{e}}, \tag{2}$$

where η and α are constants of order unity.

The high-field scaling theory suggested by Tešanović *et al.*[18,19] also predicted $M^*(T^*)=k_B T^*/s\phi_0$ in a quasi-2D system. However, the calculated value s in this theory was found to be larger than the crystallographic interlayer spacing. This discrepancy was due to the high field approximation and could be mediated by using a theoretical result of Koshelev,[20] which calculated the contribution of the fluctuation to the magnetization by considering higher Landau levels. In this case, the magnetization at the crossing point becomes $M^*(T^*)=(k_B T^*/s\phi_0)m_\infty$, where $m_\infty \approx 0.346$. Using the values of M^* and T^* from our data, we calculated $s=1.78$ nm, which is comparable to the crystallographic interlayer spacing. The large s indicates that Tl-1234 has a strong two-dimensional (2D) superconducting character.

Another indication of the 2D nature of the four layer Tl-1234 is from the high field scaling law proposed by Ullah-Dorsey. According to Ullah-Dorsey,[9] in the critical region around T_c the scaling form for magnetization is given by

$$\frac{4\pi M}{(TH)^n} = F\left[A\frac{T-T_c(H)}{(TH)^n}\right], \tag{3}$$

where F is a scaling function, A is the temperature and field independent coefficient, and the exponent n is $2/3$ for 3D and $1/2$ for 2D. The inset of Fig. 2 shows the data scaled by the 2D scaling. For each field, all data collapsed onto a single curve, consistent with the 2D nature of Tl-1234. This analysis gives $T_c=128$ K and the slope $dH_{c2}/dT_c=-2.24$ T/K near T_c. The 3D scaling was not satisfactory.

Far away from T_c, the effects of thermal fluctuations are less important. In this regime, we used the Hao-Clem model to analyze the reversible magnetization. Using this model, several important thermodynamic parameters were obtained. A detailed description of the method was given in Ref. 10.

Figure 3 and the inset of Fig. 3 show the temperature dependence of the thermodynamic critical field (H_c) and the Ginzburg-Landau parameter (κ), respectively. The solid-line

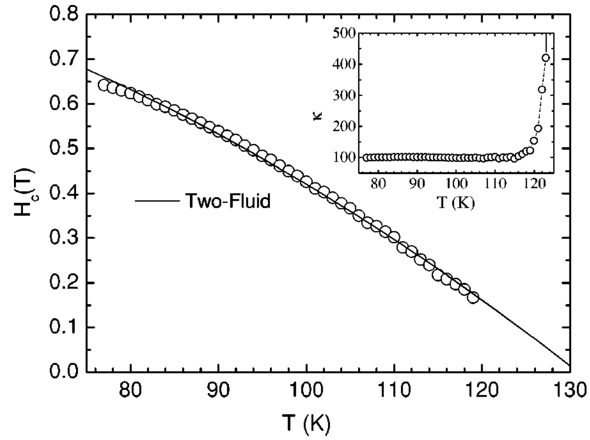

FIG. 3. Temperature dependence of thermodynamic critical field $H_c(T)$ from theoretical fitting. The solid line represents the two-fluid model. The inset shows temperature dependence of the κ extracted from the Hao-Clem model.

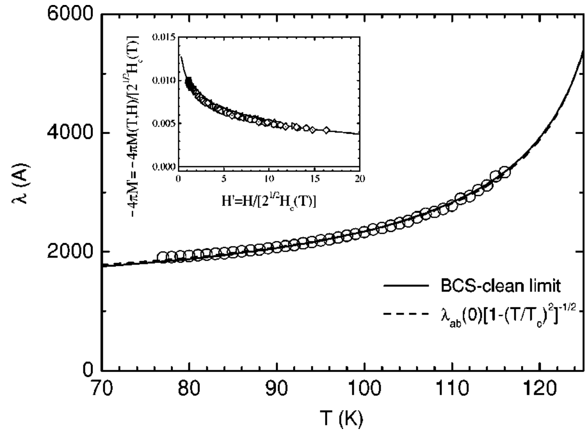

FIG. 4. Temperature dependence of the penetration depth $\lambda_{ab}(T)$ for T1-1234 obtained from the experiment. Dashed and solid lines represent $\lambda(T)=\lambda(0)/[1-(T/T_c)^4]^{1/2}$ and the BCS clean, respectively. Magnetization $-4\pi M'(H)=-4\pi M/\sqrt{2}H_c(T)$ versus $H' = H/\sqrt{2}H_c(T)$. Solid line represents the universal curve derived from the model of Hao *et al.* with $\kappa=99.7$.

represents the two-fluid model for H_c, which yields $H_c(0) = 1.01$ T and $T_c = 130.8$ K. The $\kappa(T)$ is nearly constant at $\kappa = 99.7$ up to $T = 116$ K ($\approx 0.91\,T_c$) but rapidly increases above this temperature due to the large thermal fluctuations. This anomalous behavior is consistent with the high field scaling by Ullah-Dorsey.

If the value of κ is correctly chosen, the Hao-Clem model[21–23] predicts that $-4\pi M(H)$ curves are scaled by $\sqrt{2}H_c(T)$. The inset of Fig. 4 shows the experimental data and the theoretical curve of $-4\pi M'(H)=-4\pi M/\sqrt{2}H_c(T)$ versus $H' = H/\sqrt{2}H_c(T)$ with $\kappa=99.7$. All the data are clearly collapsed onto a single curve.

Employing the values of $H_c(0)$ and κ obtained from the Hao-Clem model, we calculated several thermodynamic parameters. Using the relation of $H_{c2}(T)=\sqrt{2}\kappa H_c(T)$, $(dH_{c2}/dT)_{T_c} = 2.23$ T/K was obtained. According to the Werthamer-Helfand-Hohenberg formula[24] $H_{c2}(0) = 0.5758(\kappa_1/\kappa)T_c|dH_{c2}/dT|_{T_c}$, $H_{c2}(0)$ can be estimated, where κ_1/κ is 1.26 in the clean and 1.20 in the dirty limit. $H_{c2}(0)$ was 207 T, which corresponds to $\xi_{ab}(0) = 1.26$ nm in clean limit and 197 T and $\xi_{ab}(0) = 1.29$ nm in the dirty limit. These values are very much consistent with that of high field scaling.

Figure 4 shows the magnetic penetration depth $[\lambda_{ab}(T)]$ in Tl-1234 that was obtained using the relation $\lambda(T) = \kappa[\phi_0/2\pi H_{c2}(T)]^{1/2}$, along with the theoretical calculations of different models. The solid line and the dotted line represent the BCS clean limit calculation and an empirical formula, defined by $\lambda(T)=\lambda(0)/[1-(T/T_c)^2]^{1/2}$, respectively. As shown in this figure, these models reasonably fit the experimental data. As a result, the derived value of $\lambda_{ab}(0)$ is 156 nm with $T_c = 130.5$ K for the BCS clean limit and 150 nm with $T_c = 130$ K for the empirical formula.

Table I summarizes superconducting parameters of Tl-1234 as well as those of other four-layer superconductors

TABLE I. Thermodynamic parameters of three four-layer superconductors deduced from the reversible magnetization.

	$TlBa_2Ca_3Cu_4O_y$	$HgBa_2Ca_3Cu_4O_{10+\delta}$	$CuBa_2Ca_3Cu_4O_{12-\delta}$
T_c (K)	128 (Low field) 130.5 (BCS)	125	117
κ	99.7	102	127
$-(dH_{c2}/dT_c)_{Tc}$ (T/K)	2.23 2.24 (scaling)	2.2	2.3
$H_c(0)$ (T)	1.02	1.1	0.9
$H_{c2}(0)$ (T)	207	205	196
$\xi_{ab}(0)$ (nm)	1.26	1.27	1.28
$\lambda_{ab}(0)$ (nm)	156	157	198

Hg-1234 (Ref. 25) and Cu-1234 (Ref. 11) for comparison. Before the synthesis of the high quality Tl-1234 superconductor, the T_c and H_{c2} of Hg-1234 were the best among the four layer superconductors. However, as shown the Table I, the T_c of T1-1234 is now the highest. The enhanced T_c in T1-1234 may be due to the high charge carrier density deduced from the penetration depth as shown in the table.

IV. SUMMARY

The equilibrium magnetization for a well-aligned T1-1234 was measured at various fields up to 5 T and various superconducting parameters were obtained from the Hao-Clem model and scaling of the high field magnetization. The fluctuation-induced magnetization shows a pronounced two-dimensional behavior, which is a natural consequence of the inequivalent charge distribution within a unit cell.

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[1] K. Tanaka *et al.*, Phys. Rev. B **63**, 064508 (2001).
[2] Y. Zhuo *et al.*, Phys. Rev. B **60**, 13 094 (1999).
[3] S. L. Cooper and K. E. Gray, in *Physical Properties of High Temperature Superconductors IV* (World Scientific, Singapore, 1994), p. 61.
[4] H. Kotegawa *et al.*, Phys. Rev. B **64**, 064515 (2001).
[5] H. Kotegawa *et al.*, Phys. Rev. B **69**, 014501 (2004).
[6] D. H. Kim *et al.*, Physica C **177**, 431 (1991).
[7] A. Iyo, Y. Tanaka, Y. Ishiura, M. Tokumoto, K. Tokiwa, T. Watanabe, and H. Ihara, Supercond. Sci. Technol. **14**, 504 (2001); A. Iyo, Y. Aizawa, Y. Tanaka, M. Tokumoto, K. Tokiwa, T. Watanabe, and H. Ihara, Physica C **357–360**, 324 (2001); A. Iyo, Y. Ishiura, Y. Tanaka, P. Badica, K. Tokiwa, T. Watanabe, and H. Ihara, *ibid.* **370**, 205 (2002).
[8] H. Ihara *et al.*, Phys. Rev. B **38**, 11 952 (1988); D. M. Ogborne and M. T. Weller, Physica C **230**, 153 (1994).
[9] S. Ullah and A. T. Dorsey, Phys. Rev. Lett. **65**, 2066 (1990).
[10] Z. Hao and J. R. Clem, Phys. Rev. Lett. **67**, 2371 (1991); Z. Hao, J. R. Clem, M. W. McElfresh, L. Civale, A. P. Malozemoff, and F. Holtzberg, Phys. Rev. B **43**, 2844 (1991).
[11] M.-S. Kim, C. U. Jung, S.-I. Lee, and A. Iyo, Phys. Rev. B **63**, 134513 (2001).
[12] D. E. Farrell, B. S. Chandrsekhar, M. R. McGuire, M. M. Fang, V. G. Kogan, J. R. Clem, and D. K. Finnemore, Phys. Rev. B **36**, 4025 (1987).
[13] J. H. Choi *et al.*, Phys. Rev. B **58**, 538 (1998).
[14] M.-K. Bae, M.-S. Kim, S.-I. Lee, N.-G. Park, S.-J. Hwang, D.-H. Kim, and J.-H. Choy, Phys. Rev. B **53**, 12 416 (1996).
[15] A. Wahl, V. Hardy, F. Warmont, A. Maignan, M. P. Delamare, and Ch. Simon, Phys. Rev. B **55**, 3929 (1997).
[16] H.-J. Kim, P. Chowdhury, I.-S. Jo, and S.-I. Lee, Phys. Rev. B **66**, 134508 (2002).
[17] L. N. Bulaevskii, M. Ledvij, and V. G. Kogan, Phys. Rev. Lett. **68**, 3773 (1992).
[18] Z. Tešanović and A. V. Andreev, Phys. Rev. B **49**, 4064 (1994).
[19] Z. Tešanović, L. Xing, and L. Bulaevskii, Phys. Rev. Lett. **69**, 3563 (1992).
[20] A. E. Koshelev, Phys. Rev. B **50**, 506 (1994).
[21] M.-S. Kim, T. R. Lemberger, C. U. Jung, J.-H. Choi, J. Y. Kim, H.-J. Kim, and S.-I. Lee, Phys. Rev. B **66**, 214509 (2002).
[22] Y. Zhuo, J.-H. Choi, M.-S. Kim, W.-S. Kim, Z. S. Lim, and S.-I. Lee, Phys. Rev. B **55**, 12 719 (1997).
[23] M.-S. Kim, S.-I. Lee, S.-C. Yu, and N. H. Hur, Phys. Rev. B **53**, 9460 (1996).
[24] N. R. Werthamer, E. Helfand, and P. C. Hohenberg, Phys. Rev. **147**, 295 (1966).
[25] M.-S. Kim, S.-I. Lee, S.-C. Yu, I. Kuzemskaya, E. S. Itskevish, and K.A. Lokshin, Phys. Rev. B **57**, 6121 (1998).