Enhanced anisotropic nature of $HgBa_2Ca_3Cu_4O_{10+\delta}$

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Reversible magnetization of grain-aligned $HgBa_2Ca_3Cu_4O_{10+\delta}$ with the external magnetic fields of 1 T $\leqslant H \leqslant$ 5 T parallel to the c axis has been measured. The application of the model of Hao *et al.* [Phys. Rev. Lett. **67**, 2371 (1991); Phys. Rev. B **43**, 2844 (1991)] to our data gives the anomalous temperature dependence of the Ginzburg-Landau parameter $\kappa(T)$, i.e., the increase of κ with T, in the entire reversible region 81 K $\leqslant T \leqslant$ 100 K. This implies that the thermal distortions of the vortices are not negligible even far below T_c and thus the system is a highly anisotropic. This is supported by the two-dimensional scaling behavior of the magnetization in the critical region. From the model of Hao *et al.* we obtained various superconducting parameters such as the critical fields, the coherence length, and the penetration depth. In particular, the zero-temperature penetration depth $\lambda_{ab}(0)$ is estimated to be 157 nm, which is the smallest of the Hg-based superconductors. We infer that this is due to the large hole density within the CuO_2 plane.
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I. INTRODUCTION

Within the homologous series of $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$, the value of T_c is known to increase with the number of the CuO_2 layers n up to $n=3$. The compound with $n=4$ (Hg-1234) shows the transition near $T=125$ K, which is lower than $T_c=135$ K of the compound with $n=3$ (Hg-1223), but is comparable to $T_c=127$ K of the compound with $n=2$ (Hg-1212). It is widely accepted that the charge carrier density within the CuO_2 plane and the interlayer coupling are responsible for determining the transition temperature of the layered superconductors.[1] Thus it is desirable to elucidate the variation of these factors with respect to n in Hg-based superconductors. The study of reversible magnetization and superconducting fluctuation effects could give crucial information for this purpose.

In this paper, we report the experimental results on the reversible magnetization of grain-aligned Hg-1234 with the external magnetic fields parallel to the c axis. The two dimensional (2D) nature is evidenced by the 2D scaling behavior of the magnetization around $T_c(H)$ and a strong vortex fluctuation effect well below T_c. These characteristics were analyzed quantitatively by the theoretical predictions of Tešanović *et al.*,[2,3] and Bulaevskii, Ledvij, and Kogan.[4,5] From these analyses the effective interlayer spacing $s=44.6$ was obtained. However, the physical meaning of s is ambiguous since the value is larger than the length of the crystallographic c axis. To elucidate this we also considered the model for the critical fluctuations proposed by Koshelev.[6]

In the temperature region 81 K $\leqslant T \leqslant$ 100 K where the contribution of the vortex fluctuation effect to the magnetization is relatively small, the magnetization was analyzed using the model of Hao *et al.*[7,8] From this, various thermodynamic parameters were obtained. Notably, the zero-temperature penetration depth $\lambda_{ab}(0)$, which contains the information about the charge carrier density, is compared with those of Hg-based superconductors with different n.

II. EXPERIMENTAL ASPECTS

The sample was synthesized by the solid-state reaction method in a high-pressure chamber with a specially designed anvil. The technique of the sample preparation is described elsewhere.[9] The x-ray-diffraction analysis showed the phase purity to be more than 90%. This compound has a tetragonal symmetry ($p4/mmm$) with the lattice parameters of $a=3.8516(2)$ Å and $c=18.988(2)$ Å. To obtain the c-axis aligned sample, the method of Farrell *et al.*[10] was employed. The fine powder of the sample was aligned in commercial epoxy (Hardman Inc.) with an external magnetic field of 7 T at room temperature. The size of the permanently aligned sample was approximately 9.5 mm long and 3 mm in diameter. From the x-ray rocking curve measurement the full width at half maximum of (006) reflection was estimated to be less than 2°. The low-field dc susceptibility measurement revealed the transition temperature $T_c \simeq$ 125 K, the transition width $\Delta T_c \simeq$ 6K, and the superconducting volume fraction $V_s \simeq 96\%$ of the sample.

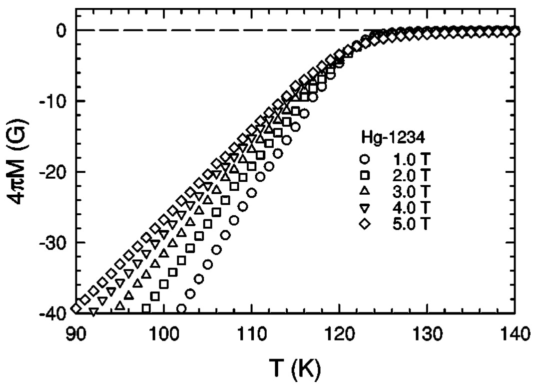

FIG. 1. Temperature dependence of the reversible magnetization $4\pi M(T)$ in the field range 1 T $\leqslant H \leqslant$ 5 T parallel to the c axis.

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the reversible magnetization $4\pi M(T)$ in the external magnetic fields of 1 T $\leqslant H \leqslant$ 5 T parallel to the c axis. For $H=1$ T, the lower boundary of the reversible temperature region is about 80 K. An intriguing feature in this figure is the existence of the crossing point of the magnetization curves at $T \simeq 121$ K. As Bulaevskii *et al.*[4] pointed out, in most cuprate superconductors, the thermal distortions of the vortices out of the straight stacks results in an extra contribution to the free energy of the system, i.e., the vortex fluctuation effect. This crossing feature is clear evidence of the vortex fluctuation effect.

Owing to the vortex fluctuation effect, the reversible magnetization $4\pi M(T,H)$ deviates from the prediction of the Ginzburg-Landau (GL) theory. Thus the quantities such as the upper critical field $H_{c2}(T)$ and the GL parameter $\kappa(T)$ evaluated by the application of the London model or the model of Hao *et al.* show an unphysical increase with temperature near the crossing temperature $T=T^*$. The anomalous temperature dependences of these quantities have been regarded as a qualitative barometer of the degree of the vortex fluctuations.[5]

Figure 2 shows the GL parameter with respect to the temperature, obtained from the model of Hao *et al.* The abrupt increase of κ with the temperature near $T=100$ K is clearly demonstrated. Furthermore, even though the rate is some-

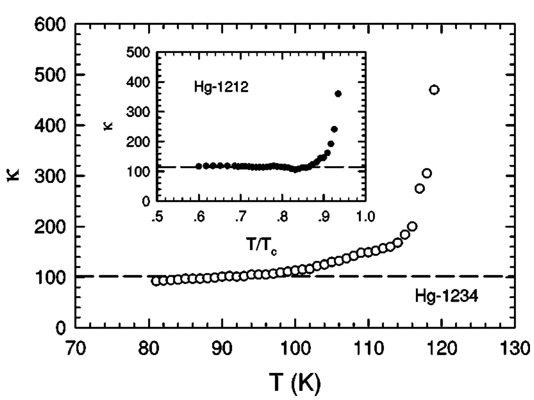

FIG. 2. Temperature dependence of the Ginzburg-Landau parameter $\kappa(T)$ extracted from the model of Hao *et al.* Inset: temperature vs the Ginzburg-Landau parameter for Hg-1212.

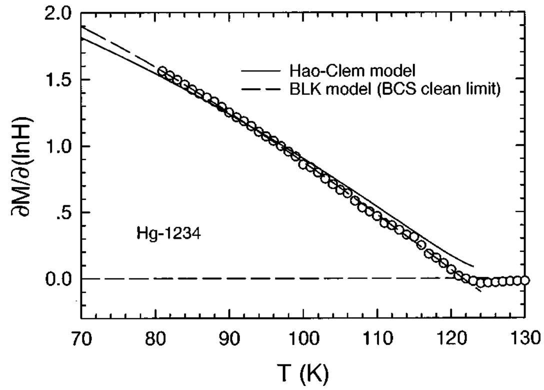

FIG. 3. Temperature dependence of the slope $\partial M/\partial(\ln H)$. Solid and dashed lines represent the theoretical curves deduced from the model of Hao *et al.* and the BLK model, respectively.

what diminished, the upward trend of κ is extended to the lower boundary of the reversible temperature region, which indicates the extensive vortex fluctuation effect. This is in sharp contrast to the case of the Hg-1212 showing 3D superconducting nature, where the anomalous behavior is limited only for $T/T_c \geqslant 0.86$, as shown in the inset of Fig. 2. Since the vortex fluctuation effect is severe for large anisotropic materials, the dominant vortex fluctuations of Hg-1234 is inferred to originate from its strong anisotropic nature.

For a quantitative analysis of the vortex fluctuation effect in Hg-1234, the Bulaevskii-Ledvij-Kogan (BLK) model[4,5] was applied. In this model, the contribution of the vortex fluctuations to the magnetization is given by $M_{\rm VF} = \ln(16\pi k_B T\kappa^2/\alpha\phi_0 s H\sqrt{e})k_B T/\phi_0 s$, where s is the effective interlayer spacing, ϕ_0 is the flux quantum, $e = 2.718\ldots$, and α is constant of order unity. Thus the slope $\partial M/\partial(\ln H) = \partial(M_{\rm rev}+M_{\rm VF})/\partial(\ln H)$ is

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

where $M_{\rm rev}$ is the reversible magnetization in the frame of the London model and the fluctuation term $g(T)$ is given by

$$g(T) = \frac{32\pi^2 k_B}{\phi_0^2 s} T\lambda_{ab}^2(T). \tag{2}$$

Figure 3 shows the temperature dependence of the slope $\partial M/\partial(\ln H)$ for Hg-1234. The solid line represents our attempt to fit using Eq. (1), assuming the BCS clean limit.[11] From this analysis, we obtained $s=44.6$ Å and $\lambda_{ab}(0)=158$ nm. The dashed line represents the theoretical curve from the model of Hao *et al.*, which deviates somewhat from the data.

In the BLK model, the temperature T^* is defined as $g(T^*)=1$ and the magnetization at this temperature is field independent

$$-M(T^*) \equiv -M^* = \frac{k_B T^*}{s\phi_0}\ln\frac{\eta\alpha}{\sqrt{e}}, \tag{3}$$

where η is the constant of order unity. The high-field scaling theory on quasi-2D systems proposed by Tešanović *et al.*[2,3] predicts the same relation as Eq. (3) without the ln factor, i.e., $\ln(\eta\alpha/\sqrt{e})=1$. Using $M^*=0.18$ G and $T^*=121.8$ K taken from the data, a constant $\ln(\eta\alpha/\sqrt{e})$ was estimated to be $\sim$ 0.99 using $s=44.6$ Å, which means that both the BLK model and the high-field scaling theory give the same results, even though their approaches are different from each other.

It is noteworthy that the effective interlayer spacing $s=44.6$ Å is much larger than the lattice parameter $c=19$ Å. Thus the physical implication of s is ambiguous. Recently, Koshelev[6] claimed that the exact scaling formula proposed by Tešanović *et al.* is a good approximation at the critical region, but the magnetization M^* at $T=T^*$ is beyond the applicability of the scaling law because the contribution of the weak fluctuations at higher Landau levels is not negligible. If the weak fluctuations are treated as the Gaussian approximation, then the field-independent magnetization is given by

$$M^* = -m_\infty \frac{k_B T^*}{\phi_0 s}, \tag{4}$$

where $m_\infty \approx 0.346$. From this we obtained a different value $s=15.4$ Å, which is between the spacing of the CuO_2 layers separated by the Hg-O layer (9.6 Å) and the lattice parameter $c=19$ Å. This value is close to $s=15.3$ Å of $Bi_2Sr_2CaCu_2O_8$, but is significantly larger than $s=11.7$ Å of $YBa_2Cu_3O_{7-\delta}$. This relatively large value of s might explain why Hg-1234 has a strong 2D nature.

Further strong evidence supporting this is that the magnetization at the critical region follows the 2D scaling behavior. In the high-field limit, according to Tešanović *et al.*,[2,3] the exact scaling function for the 2D system is given by

$$\frac{M(H,T)}{\sqrt{HT}} \frac{s\phi_0 H'_{c2}}{A} = x - \sqrt{x^2+2}, \tag{5}$$

where A is a constant, $x=A[T-T_c(H)]/(TH)^{1/2}$, $H'_{c2} = dH_{c2}/dT|_{T_c}$, and s is the effective interlayer spacing. To compare the scaling function with the data, it is convenient to reduce Eq. (5) as

$$\frac{M}{M^*} = \frac{1}{2}\{1-\tau-h+\sqrt{(1-\tau-h)^2+4h}\}, \tag{6}$$

where $\tau=(T-T^*)/(T_c-T^*)$ and $h=H/H_{c2}(T^*)$. The upper part of Fig. 4 shows our attempt to fit the fluctuation-induced magnetization using Eq. (6) with the experimental values of M^* and T^*. This analysis gives $T_c=124.2$ K and $-dH_{c2}/dT|_{T_c}=2.4$ T/K. The scaled magnetization curves that are addressed in the different fields are shown in the lower part of Fig. 4. The solid line represents the theoretical curve.

As mentioned above, the extra free energy due to the distortion of vortices should be considered to describe the reversible magnetization properly. However, since the vortex fluctuation effect is less important at low temperatures, the change of $\kappa(T)$ well below T_c is smaller than that near T_c. We take $\kappa_{avg}=102$ as the average value of $\kappa(T)$ in the limited temperature range of 81 K $\leqslant T \leqslant$ 100 K. Using this fixed GL parameter, $-4\pi M(H)$ curves could be represented as a universal curve with the scaling factor $\sqrt{2}H_c(T)$, consistent with the model of Hao *et al.*[8] Figure 5 shows

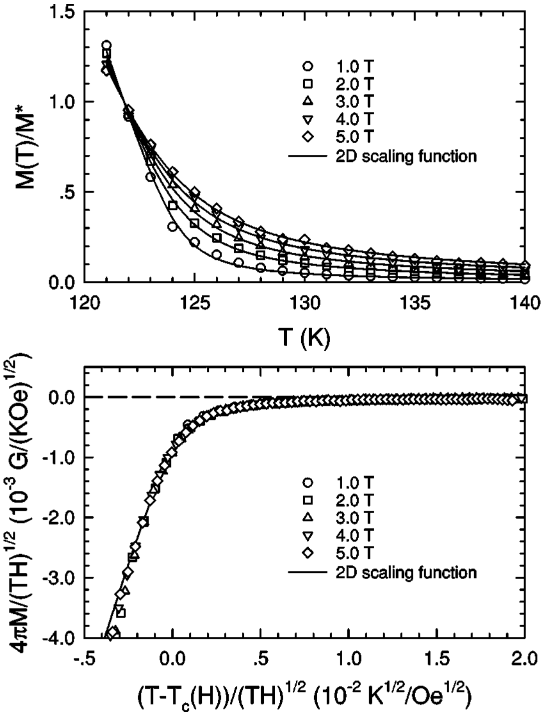

FIG. 4. Temperature vs the normalized magnetization M/M^* around T_c (upper part) and 2D scaling of the fluctuation-induced magnetization $4\pi M(T,H)$ (lower part). Solid lines represent theoretical curves obtained from the exact scaling function proposed by Tešanović.

$-4\pi M' = -4\pi M/\sqrt{2}H_c(T)$ vs $H' = H/\sqrt{2}H_c(T)$ of the experimental data and the theoretical curve.

The inset of Fig. 5 shows the thermodynamic critical field $H_c(T)$ obtained from the model of Hao *et al.* employing κ_{avg}. The solid and dashed lines represent the BCS temperature dependence of H_c and the empirical two-fluid model,[12] respectively. The BCS result for $H_c(T)$ yields $H_c(0)=1.13$ T and $T_c=126$ K. These results as well as various thermodynamic parameters such as the critical fields $[H_c(0)$ and $H_{c2}(0)]$ and the characteristic lengths $[\lambda_{ab}(0)$ and $\xi_{ab}(0)]$

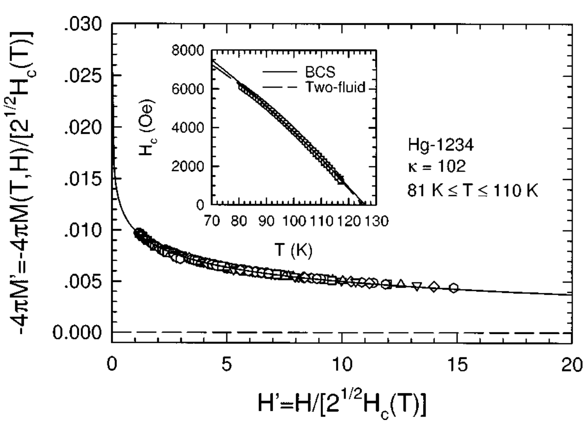

FIG. 5. Magnetization $-4\pi M' = -4\pi M/\sqrt{2}H_c(T)$ vs external magnetic field $H' = H/\sqrt{2}H_c(T)$. The solid line represents the universal curve derived from the model of Hao *et al.* with $\kappa=102$. Inset: temperature dependence of the thermodynamic critical field $H_c(T)$. Solid and dashed lines represent the BCS temperature dependence and the two-fluid model, respectively.

TABLE I. Ginzburg-Landau parameter, critical fields, and characteristic lengths of $HgBa_2Ca_3Cu_4O_{10+\delta}$.

κ	$dH_{c2}/dT\|_{T_c}$ (T/K)	$H_c(0)$ (T)	$H_{c2}(0)$ (T)	$\lambda_{ab}(0)$ (nm)	$\xi_{ab}(0)$ (Å)
102 [a]	−2.2 [a]	1.13 [c]	205 [e]	157 [e]	12.7 [e]
	−2.4 [b]	1.06 [d]	195 [f]	172 [f]	13.0 [f]
				158 [g]	

[a] Hao-Clem model.
[b] 2D scaling function.
[c] BCS temperature dependence of H_c.
[d] Two-fluid model.
[e] BCS clean limit.
[f] BCS dirty limit.
[g] BLK model.

are summarized in Table I. The total errors of the superconducting parameters presented here from all sources including the sample quality and the experimental errors are estimated to be much less than 10%.

Employing the $H_c(T)$ and κ_{avg} obtained from the above, the penetration depth $\lambda_{ab}(T)$ was evaluated by using the relation $\lambda(T)=\kappa[\phi_0/2\pi H_{c2}(T)]^{1/2}$, as shown in Fig. 6. In this figure, the solid and dashed lines show the best fit of the BCS clean and dirty limits, respectively. From this analysis, $\lambda_{ab}(0)$ was estimated to be 157 nm for the clean limit and 172 nm for the dirty limit. The value of $\lambda_{ab}(0)$ for the clean limit is fairly consistent with 158 nm from the BLK model. For comparison, the value is plotted in Fig. 6 with earlier reported values[13–16] for Hg-1201, Hg-1212, and Hg-1223.

It is important to notice that $\lambda_{ab}(0)$ decreases with the number of CuO_2 layers in the unit cell n. Since $\lambda_{ab}(0)$ is proportional to $(m^*_{ab}/n_s)^{1/2}$, where m^*_{ab} is the electronic effective mass in the ab plane and n_s is the charge carrier density, the decrease in λ with n reflects an increase of n_s and/or a decrease of m^* with n. It is widely accepted that the hole density is correlated with the bond length between the Hg and apical oxygen O_{apical} in the Hg-based superconductors. The hole density of $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ with $\delta=0$ ($n=1$, 2, and 3), i.e., the fully stoichiometric compounds, is nearly zero or so small that it falls within the limit of electron localization.[17] The oxygen doping into the HgO_δ plane results in the charge transfer from CuO_2 planes, and this feature is reflected by the shorter Hg-O_{apical} bond length.

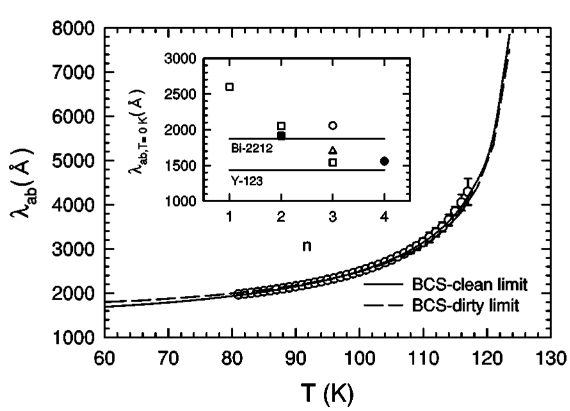

FIG. 6. Temperature dependence of the penetration depth $\lambda_{ab}(T)$ obtained from the model of Hao *et al.* Solid and dashed lines represent the BCS clean and dirty limits, respectively. The inset shows the variation of the penetration depth $\lambda_{ab}(0)$ with respect to the number of CuO_2 layers n per unit cell for Hg-based superconductors: filled symbols, this work; open squares, Ref. 14; open triangles, Ref. 13; open circles, Ref. 15.

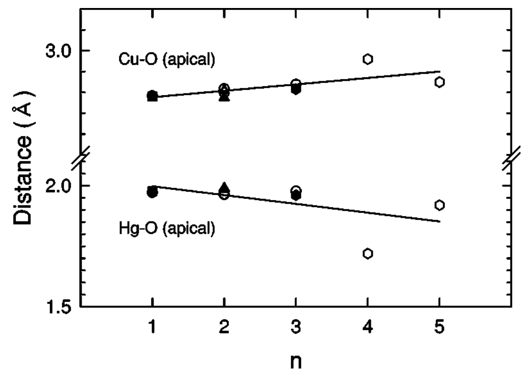

FIG. 7. Variation of Hg-O_{apical} and Cu-O_{apical} bond lengths with respect to the number of CuO_2 layers n per unit cell for Hg-based superconductors: open squares, Ref. 18; filled circles, Ref. 19; open hexagons, Refs. 20 and 21; filled triangles, Ref. 23; open triangles, Ref. 22; open circles, Ref. 24; filled hexagons, Ref. 25.

For Hg-based compounds, the Hg-O_{apical} bond length[18–25] is known to decrease with n as shown in Fig. 7. As mentioned above, this may imply an increase of the hole density with n. This is supported by the calculation of the electronic band structure by Gupta and Gupta.[17] They reported that the values of hole density per a CuO_2 are ∼0.21 hole per CuO_2 ($\delta\sim0.09$), ∼0.26 hole per CuO_2 ($\delta\sim0.22$), and ∼0.27 hole per CuO_2 ($\delta\sim0.35$) for Hg-1201, Hg-1212, and Hg-1223, respectively.

The bond length ($\simeq$ 1.72 Å) of Hg-1234 (Ref. 20) is the shortest among the known Hg-based superconductors. Recalling that the short Hg-O_{apical} bond length is caused by the large charge transfer from the CuO_2 planes, we can infer that the small penetration depth of Hg-1234 is due to the large hole density within the plane. From this point of view, for Hg-based superconductors, a decrease in penetration depth with n can be explained by the same reason.

It is well known that there is a reasonable correlation between the transition temperature and the charge carrier density within the CuO_2 plane. This explains the variation of T_c with n up to $n=3$. However, the fact that Hg-1234 has a lower T_c ($\simeq$ 125 K) than the T_c ($\simeq$ 135 K) of Hg-1223 even though it has the highest hole density suggests that there is a difficulty in inferring the trend in T_c from the carrier density alone. It is also claimed that the high-T_c superconductivity can be enhanced by the coupling or tunneling between the superconducting layers.[26–29] If one recalls that Hg-1234 shows a strong 2D nature, the relatively low T_c is inferred to originate from the weak interlayer coupling.

IV. SUMMARY

We found that 2D fluctuations are dominant due to weak interlayer coupling in Hg-1234. The 2D nature of Hg-1234 induced strong positional fluctuations of the vortices below T_c and the reversible magnetization deviated remarkably from the prediction of the mean-field theory. Except for the temperature region of dominant vortex fluctuations, the magnetization was described approximately using the model of Hao *et al.* From this analysis, the various thermodynamic parameters such as the penetration depth $\lambda_{ab}(0)$, the coherence length $\xi_{ab}(0)$, the Ginzburg-Landau parameter κ, and the critical fields $[H_c(0)$ and $H_{c2}(0)]$ were extracted. In particular, if the BCS clean limit is assumed, $\lambda_{ab}(0)$ is estimated to be 157 nm, which is the smallest among the Hg-based superconductors. We infer that this is due to its large hole density within the CuO_2 plane, which is consistent with the results of electronic band-structure calculations for Hg-based superconductors. However, the fact that T_c of Hg-1234 is lower than that of Hg-1223 contradicts the claim that the transition temperature correlates with the charge carrier density. Therefore, there is difficulty in inferring the trends in T_c from the charge carrier density alone and a number of theories, which assume that the high-T_c superconductivity is enhanced by the interlayer coupling or the interlayer tunneling, should be considered seriously. In this sense, we can infer that the relatively low T_c of Hg-1234 originates from the weak interlayer coupling.

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