Evidence of strong anisotropic behavior in the five-layered superconductor $HgBa_2Ca_4Cu_5O_{12+y}$ from equilibrium magnetization measurements

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The equilibrium magnetization of grain-aligned $HgBa_2Ca_4Cu_5O_{12+y}$ (Hg1245) with $T_c \simeq 108$ K was measured. The Ginzburg-Landau parameter (κ), obtained by using the Hao-Clem model, increased unphysically with increasing temperature for $T/T_c \geqslant 0.7$, which indicates that the fluctuation is greatly enhanced. The two-dimensional scaling behavior of the magnetization and the temperature variation of the irreversibility field $H_{irr}(T)$ from a power-law behavior at low fields to an exponential behavior at high fields indicate that Hg1245 is highly anisotropic, which seems to be affected by the appearance of antiferromagnetism at the inner CuO_2 planes while blocking the coupling of the superconductivity between the outer CuO_2 layers.

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One of the important questions about the homologous series of $HgBa_2Ca_{n-1}Cu_nO_{2n+2+y}$ [Hg12$(n-1)n$] high-T_c cuprates is why the superconducting transition temperature (T_c) does not increase continuously with increasing number of CuO_2 planes (n), but reaches its maximum value for $n=3$ and then decreases.[1,2] One possible clue is the weakening of the superconductivity or the appearance of antiferromagnetism in the inner CuO_2 layers which blocks the coupling of the interlayer superconductivity. The oxygen doping of CuO_2 planes, such as the inner CuO_2 plane (IP) with square coordinates of oxygen and the outer CuO_2 plane (OP) with pyramidal coordinates of oxygen, is known to be not the same for Hg12$(n-1)n$ with $n=4$ or 5. An extensive study of nuclear magnetic resonance (NMR) on Hg12$(n-1)n$ and $CuBa_2Ca_{n-1}Cu_nO_{2n+4-y}$ confirmed the different doping levels in the CuO_2 layer (N_h) for $n=3-5$.[2–4] It is worth noting that N_h(OP) is always larger than N_h(IP) and that the difference $\Delta N_h = N_h(\mathrm{OP}) - N_h(\mathrm{IP})$ becomes larger with increasing n.

It is natural to accept that the weakening of the superconductivity in the IP affects the equilibrium magnetization and makes the superconductor more anisotropic. It is quite interesting to notice that the reversible magnetization of Hg1234 is quite anisotropic.[5] If the CuO_2 plane is increased by 1 for Hg1234, then it becomes the case of Hg1245; we expect to have anisotropic behavior even higher than that of Hg1234. The previous NMR studies of Hg1245 revealed that while superconductivity with $T_c \sim 108$ K develops in the optimally doped OP, antiferromagnetism with $T_N \sim 60$ K develops in the underdoped IP.[3,4] This was also confirmed by the muon spin rotation.[6,7] Naturally, the superconductivity and, thus, the equilibrium magnetization of Hg1245 are inevitably affected by antiferromagnetism of the IPs. In this sense, investigating the general trend of the bulk magnetization and the changes in the basic thermodynamic parameters is very important, but these have not yet been studied in detail. Unfortunately, unlike the case of the NMR signal, this is not a direct way to observe the antiferromagnetic phase.

In this paper, the equilibrium magnetization of $HgBa_2Ca_4Cu_5O_{12+y}$ was measured and then analyzed by using both the Hao-Clem model[8,9] and the scaling theories developed first by Ullah and Dorsey[10] and then by Tešanović and Andreev.[11,12] This analysis showed that Hg1245 was highly anisotropic. Also, the enhanced thermal fluctuation of Hg1245, especially near the T_c, indicated that Hg1245 was highly anisotropic. The magnetization was also found to satisfy the two-dimensional (2D) scaling theories[10–12] in the critical region. Finally, the $H_{irr}(T)$ showed a crossover from a power-law behavior at low fields to an exponential behavior like that of $Bi_2Sr_2CaCu_2O_8$ at high fields,[13] which is a symbol of a highly anisotropic superconductor.

Polycrystalline Hg1245 was synthesized by using a high pressure technique.[14] We ground the polycrystalline Hg1245 into a fine powder and aligned it along the c axis in high magnetic field using the method of Farrel *et al.*[15] We obtained the rocking curve from the x-ray diffraction data, and the full width at half maximum of the (001) peak was less than 0.3°. The sizes of the grains were less than 30 μm and each was in the from of a single grain.

The magnetization $4\pi M(T)$ for $H \parallel c$ was measured for $H \leqslant 5$ T. Only the reversible magnetization is shown in Fig. 1. The inset of Fig. 1 shows a magnified $4\pi M(T)$ near the transition region. The magnetization curves cross over at (104.7 K, −4 G), which reflects that thermal distortion of the line vortices dominates nonthermal distortion.[16] Using the crossover point with $T^* \simeq 104.7$ K and $4\pi M(T^*) \simeq -4$ G, we determine the effective interlayer spacing $s \sim 7.6$ Å from the relation $M(T^*) = -m_\infty(k_B T^*/\phi_0 s)$ derived by Koshelev,[17] where $m_\infty = 0.346$, and k_B and ϕ_0 are the Boltzmann constant and the magnetic flux quantum, respectively. The obtained s is smaller than the length of the unit cell along the c axis (~22.2 Å); still, it is larger than the interlayer spacing (~3.2 Å) between the CuO_2 planes of Hg1245.[3] This indicates that the interlayer coupling of the superconductivity between CuO_2 planes is much weakened in the unit cell.

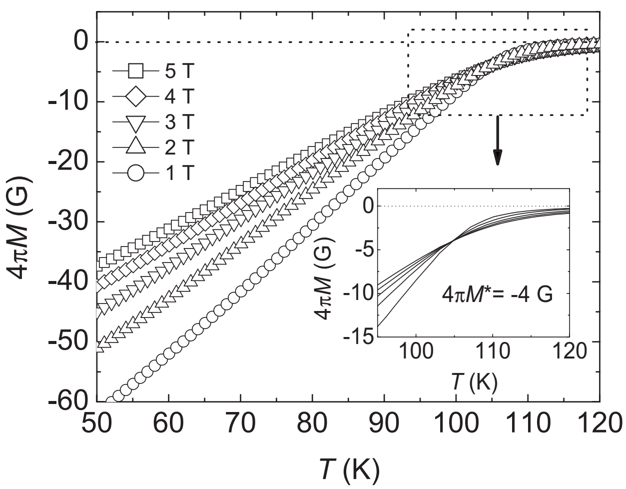

FIG. 1. Magnetization $4\pi M(T)$ obtained for values of H from 1 T (circles) to 5 T (squares) parallel to the c axis. Inset: Magnified $4\pi M(T)$ near the crossover point.

For the analysis of the reversible magnetization, the Hao-Clem model[8,9] based on the model of the Ginzburg-Landau free energy was used in the limit of low temperature where positional fluctuation of vortex is not important. This model describes the reversible magnetization for the entire mixed state of $H_{c1}<H<H_{c2}$, where H_{c1} and H_{c2} are the lower and upper critical fields, respectively. From this analysis, we determined the thermodynamic parameters, such as the Ginzburg-Landau parameter (κ) and the thermodynamic critical field (H_c).

From the Hao-Clem analysis, the temperature dependence of κ/κ_{min} for Hg1245 was obtained, as shown in Fig. 2, where κ_{min} is the lowest κ in the measured temperature range. The arbitrary values of κ/κ_{min} for the various materials are shifted for visualization. For comparison, the temperature dependences of κ/κ_{min} for $YBa_2Cu_4O_8$ (Y124),[18]

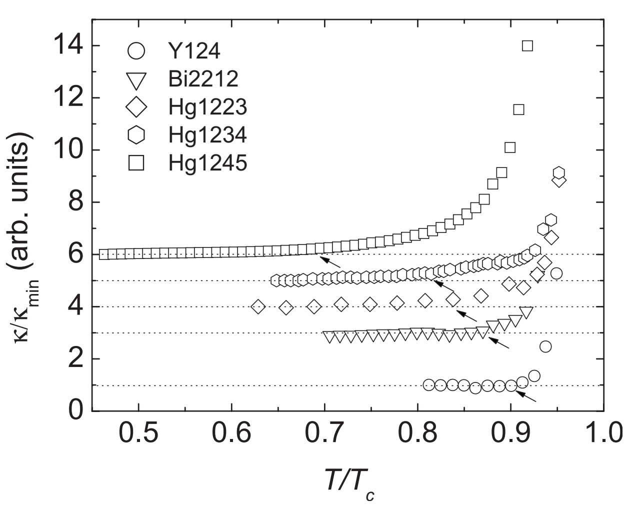

FIG. 2. Temperature dependence of the Ginzburg-Landau parameter κ/κ_{min} for Hg1245 (squares) obtained from the Hao-Clem model. For comparison, the temperature dependences of κ/κ_{min} for various cuprates are shown.

$Bi_2Sr_2CaCu_2O_8$ (Bi2212),[19] Hg1223,[20] and Hg1234 (Ref. 5) are also drawn in Fig. 2. The arrow denotes the point at which the κ starts to increase, which indicates that the vortex fluctuation becomes important from this point. The $\kappa(T)$ obtained from the Hao-Clem model, which increased with increasing T, is unphysical, indicating that the thermal fluctuation of the vortex position is not negligible.

As for a moderately anisotropic superconductor such as Y124, the κ starts to increase for $T/T_c\sim0.9$. Also, the κ below $T/T_c\sim0.9$ is almost temperature independent. On the other hand, for highly anisotropic superconductors such as Bi2212, Hg1223, and Hg1234, the points of arrows are somewhat below $T/T_c\sim0.9$, which indicates that the vortex fluctuation is strong and interlayer coupling is weak. For Hg1245, the starting point of the dominant increase in κ is $T/T_c\sim0.7$, which may be the lowest value ever reported among cuprate superconductors, even lower than the well-known highly anisotropic 2D superconductor Bi2212. The vortex fluctuation in Hg1245 is found to be even stronger than that in Hg1234.

The $H_{irr}(T)$ also gives valuable information on the anisotropy. The $H_{irr}(T)$ is influenced by the vortex pinning strength and by the strength of the interlayer coupling between the superconducting CuO_2 planes. Figure 3(a) shows $H_{irr}(T)$ vs T/T_c for Hg1245, where the irreversibility temperature T_{irr} is obtained from the criteria $M_{FC}/M_{ZFC}=0.98$. For comparison, the $H_{irr}(T)$ for various cuprate superconductors, such as $YBa_2Cu_3O_{7-\delta}$ (Y123),[21] $Sr_{0.9}La_{0.1}CuO_2$ [(Sr,La)112],[22] Hg1223,[23] Hg1234,[24] Bi2212,[23] and $Ba_2Ca_2Cu_3O_6(O,F)_2$ (0223F),[23] are shown in Fig. 3(a). For the well-known three-dimensional (3D) superconductors such as Y123 and (Sr,La)112, the H_{irr} is quite high, indicating strong interlayer coupling and, thus, strong flux pinning. In contrast, the value of the H_{irr} for very anisotropic superconductors such as Bi2212, 0223F, Hg1223, Hg1234, and Hg1245 is shifted to low temperatures. From this analysis, we notice that Hg1245 is highly anisotropic and the interlayer coupling of this compound is weaker than that of Hg1234.

In addition, the $H_{irr}(T)$ follows a scaling relation $H_{irr}(T)=H_0(1-T/T_c)^m$,[25] where H_0 is an adjustable parameter. An exponent of $m\approx3/2$ is observed for moderately anisotropic superconductors, such as Y123 (Ref. 26) and (Sr,La)112,[22] while $m\approx2$ is observed for highly anisotropic Bi2212 (Ref. 13) and $Tl_2Ba_2CaCu_2O_8$.[27] In quasi-2D superconductors, a 2D pancake vortex fluctuation becomes dominant and the $H_{irr}(T)$ shows an exponential behavior $\sim\exp(\alpha/T)$, where α is a constant at low temperatures.[13] Figure 3(b) shows a log-log plot of H_{irr} vs $1-T/T_c$. The H_{irr} shows a power-law behavior at low fields, but an exponential behavior of $\exp(\alpha/T)$ at high fields, as with 2D Bi2212.[13] The crossover point is about $H=0.7$ T, which is somewhat larger than both 0.07 T for 0223F (Ref. 23) and Bi2212,[13] and 0.2 T for Hg1223.[28,29] From this analysis, $m\approx1.98$ is obtained. The exponent m being close to 2 proves that Hg1245 belongs to the class of highly anisotropic 2D superconductors. As mentioned before, we think that the nonincreasing T_c for n larger than 3 for Hg12$(n-1)n$ is related to the reduction of the interlayer coupling because the anisotropy follows the same trend.

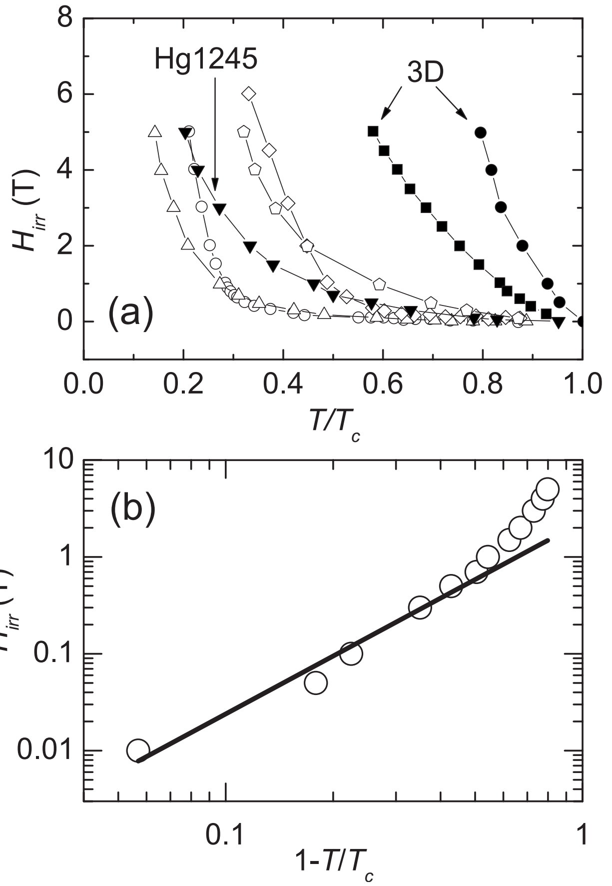

FIG. 3. (a) Irreversibility field $H_{irr}(T)$ for Hg1245 (solid down triangles) obtained from the $4\pi M(T)$ for $0.01\ \text{T} \leqslant H \leqslant 5$ T. For comparison, the irreversibility fields for Y123 (solid circles), (Sr,La)112 (solid squares), Hg1223 (diamonds), Hg1234 (pentagons), Bi2212 (circles), and 0223F (up triangles) are shown. Y123 and (Sr,La)112 are 3D superconductors, while Hg1223, Hg1234, Bi2212, and 0223F are quasi-2D superconductors. (b) A log-log plot of $H_{irr}(T)$ vs $1-T/T_c$. The solid line represents the fitting line ($H \leqslant 0.7$ T) obtained by using the relation $H_{irr}(T)=H_0(1-T/T_c)^m$.

We also investigated the scaling behaviors of the magnetization in the critical region. We began with the scaling theory of Ullah and Dorsey.[10] For T near $T_c(H)$ in the critical region of a high magnetic field, this theory predicts a scaling relation for the reversible magnetization, $4\pi M/(TH)^p = F(T - T_c(H)/(TH)^p)$, where F is a field- and transition-temperature-independent function and p corresponds to a critical exponent. A p of 2/3 indicates a 3D fluctuation, whereas a p of 1/2 indicates a 2D fluctuation. Figure 4(a) shows a 2D ($p=1/2$) scaling of the magnetization with $dH_{c2}(T)/dT|_{T=T_c} = -1.15 \pm 0.2$ T/K. The magnetization scales much better for a 2D curve rather than a 3D one. The 2D scaling behavior of the magnetization in the critical region is one more piece of support for the weakening of interlayer coupling in Hg1245. However, in this scaling analysis, the scaling function is not specified.

This scaling function was predicted by Tesanović and Andreev,[11,12] and the modified equation of the exact 2D scaling function in the high-field region is given by

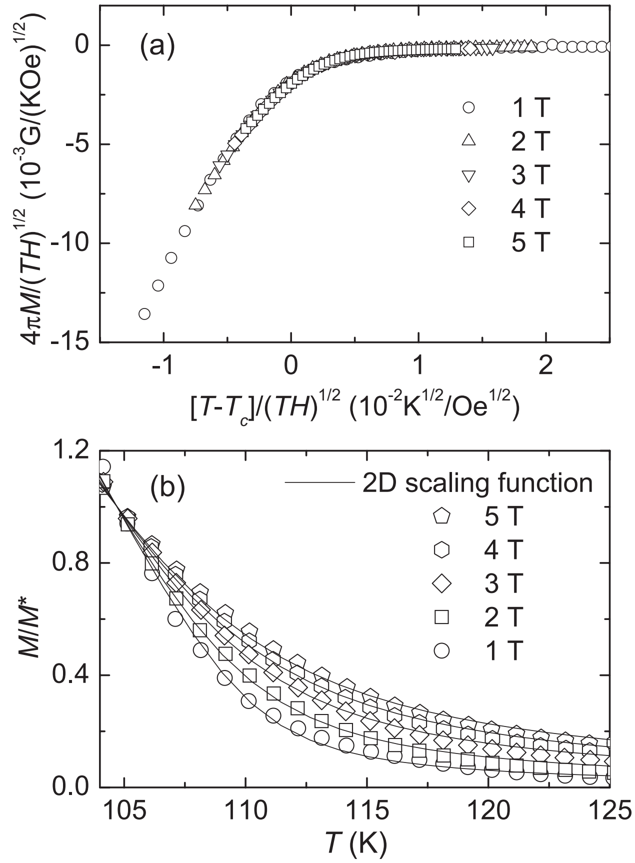

FIG. 4. (a) The Ullah-Dorsey 2D scaling theory: $4\pi M$ vs ($T - T_c$) scaled by $(TH)^{1/2}$. (b) The Tešanović-Andreev 2D scaling theory: M/M^* vs T(K).

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

where M^* is the field-independent magnetization at the crossover temperature T^*, $\tau=(T-T^*)/(T_c-T^*)$, and $h = H/H_{c2}(T^*)$. The $H_{c2}(T^*)$ is obtained from the relation $H_{c2}(T^*)=\sqrt{2}\kappa H_c(T^*)$, where the $H_c(T)$ given by the BCS theory is $H_c(T)/H_c(0)=1.7367(1-T/T_c)[1-0.2730(1-T/T_c)-0.0949(1-T/T_c)^2]$.[30] Figure 4(b) shows the results of the magnetization using Tešanović and Andreev's 2D scaling function. From this analysis, the above 2D scaling function is found to describe the magnetization very well and gives $T_c=110.4$ K and $dH_{c2}/dT|_{T=T_c} = -1.14$ T/K.

To date, the highly enhanced anisotropic behavior of the superconducting nature of Hg1245 has been confirmed as follows: (1) a derived s larger than the real interlayer spacing between two adjacent CuO_2 planes indicates that the interlayer coupling between these two CuO_2 planes is weakened; (2) the $\kappa(T)$ shows a weak interlayer coupling that is confirmed by the strong vortex fluctuation; (3) the $H_{irr}(T)$ shows a weak flux pinning strength, which originates from weak interlayer coupling, and the obtained exponent of $m \approx 1.98$ supports a highly enhanced anisotropic behavior; and (4) the magnetization in the critical region is well described by 2D scaling theories. All these results consistently support the highly anisotropic nature of the Hg1245 superconductor, which seems to be affected by a weakening of the superconductivity due to the development of antiferromagnetism at the inner CuO_2 layers. In other words, the antiferromagnetism at the IPs may affect the superconductivity and block the superconducting OPs of Hg1245. Consequently, the 2D superconducting nature of Hg1245 may be affected by the antiferromagnetism at the IPs.

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