

## Study of the Thermodynamic Parameters of $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8-x}$ by Using Hao and Clem's Model

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Magnetization  $M(T)$  of grain-aligned  $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8-x}$  high-T<sub>c</sub> superconductor was measured for external magnetic fields parallel to the c-axis. The magnetization is field independent at  $T^* = 128$  K, which indicates a dominant vortex fluctuation. Except for this vortex fluctuation region, reversible magnetization could be described by using the Hao and Clem's model, and this model was used to extract thermodynamic critical field  $H_c(T) = 5877$  Oe and Ginzburg-Landau parameter  $\kappa = 118$ . We also obtained such superconducting parameters such as the penetration depth  $\lambda_{ab}(0) = 2060 \text{ \AA}$ , coherence length  $\xi_{ab}(0) = 18 \text{ \AA}$ , and upper critical field  $H_{c2}(0) = 108$  T.

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The London model [1] for high  $\kappa$  type II superconductors has been widely used to describe reversible magnetization. This model accounts for electromagnetic energy density  $F_{em}$  for the region outside the vortex core to calculate the free energy. However the core energy density  $F_{core}$  due to suppression of the order parameter in vortex is ignored in this model. In a high field region, this core effect is important because the increase in magnetization is mainly associated with  $F_{core}$ . Therefore, the London model is valid in a restricted field region of  $H_{c1} \ll H \ll H_{c2}$ . This model predicts logarithmic behavior of magnetization ( $M(H) \sim \ln H$ ). On the contrary, the Abrikosov model [1] is valid at the high field region ( $H \sim H_{c2}$ ). This model is based on the Ginzburg-Landau theory and predicts  $M(H) \sim H$  at high field region where the order parameter is highly suppressed. The Hao and Clem's model [2,3] is an integrated version of above two models and explain reversible magnetization more reasonably for entire field range of mixed state ( $H_{c1} \leq H \leq H_{c2}$ ).

This paper is a report on the experimental results of reversible magnetization for a grain aligned  $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8-x}$  (Hg1223) sample which have the highest transition temperature known for the layered Cu-O materials [4,5]. We have analyzed reversible magnetization curves in the range  $110 \text{ K} \leq T \leq 124 \text{ K}$  by using the Hao and Clem's model and found that both the magnetization curves  $M(H)$  and the normalized superfluid density  $\langle f^2 \rangle$  can be scaled to a single universal curve. From these analyses, the various superconducting parameters, such as critical fields  $H_{c1}(0)$ ,  $H_{c2}(0)$ , penetration depth  $\lambda(0)$ , coherence length  $\xi(0)$ , and  $\kappa(T)$  were obtained.

Details on the preparations of  $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8-x}$  are given elsewhere [6-8]. Details of the Raman spectra [9], thermoelectric power measurement [10] and tunneling measurement [11] for this sample are published. To obtain a c-axis aligned sample, Farrell's method [12] was employed. The powders were aligned in epoxy with an external magnetic field of 7 T. The alignment was confirmed by XRD experiment. The full width at half maximum (FWHM) of the rocking curve was less than 1 degree. The temperature dependence of magnetization was measured using a SQUID magnetometer (MPMS, Quantum Design). We repeated the measurement in ZFC and FC at various magnetic fields. Weak temperature-dependent contributions originated from the epoxy and the paramagnetic impurities were subtracted from the observed values by fitting the magnetization curve at a high temperature region of  $200 \text{ K} < T < 230 \text{ K}$  by  $C/T + \chi_0$ .

The dimensionless Ginzburg-Landau free energy [13] per unit volume over cross-sectional area  $A$  in a plane perpendicular to the vortices, measured relative to that of the Meissner state can be expressed as

$$F = \frac{1}{A} \int d^2\rho \left[ \frac{1}{2}(1-f^2)^2 + \frac{1}{\kappa^2}(\nabla f)^2 + f^2 \left( a + \frac{1}{\kappa} \nabla \gamma \right)^2 + b^2 \right]. \quad (1)$$

In this integral, the first two terms represent the core energy density and the last two terms represent electromagnetic energy density.  $f$  and  $\gamma$  are the normalized magnitude and phase of the order parameter  $\Psi = \Psi_0 f e^{i\gamma}$ . In this dimensionless equation, length  $\rho$ , magnetic field, and free energy are normalized by penetration depth  $\lambda$ ,  $\sqrt{2}H_c$ , and  $H_c^2/4\pi$  respectively.

Hao et al. assume the trial order parameter as follows:

$$f(\rho) = \frac{\rho}{(\rho^2 + \xi_v^2)^{1/2}} f_\infty, \quad (2)$$

where  $\xi_v$  and  $f_\infty$  are variational parameters representing the effective core radius of a vortex and the order parameter far from core respectively. The dimensionless magnetization  $-4\pi M'$  is given by

$$-4\pi M' = \frac{1}{2} \frac{\partial}{\partial B} (F - B^2)_{f_\infty, \xi_v} \quad (3)$$

$$\begin{aligned}
&= \frac{\kappa f_{\infty}^2 \xi_v^2}{2} \left[ \frac{1 - f_{\infty}^2}{2} \ln \left( \frac{2}{B\kappa\xi_v^2} + 1 \right) - \frac{1 - f_{\infty}^2}{2 + B\kappa\xi_v^2} + \frac{f_{\infty}^2}{(2 + B\kappa\xi_v^2)^2} \right] \\
&+ \frac{f_{\infty}^2 (2 + 3B\kappa\xi_v^2)}{2\kappa(2 + B\kappa\xi_v^2)^3} + \frac{f_{\infty}}{2\kappa\xi_v K_1(f_{\infty}\xi_v)} \left[ K_0(\xi_v(f_{\infty}^2 + 2B\kappa)^{1/2}) \right. \\
&\left. - \frac{B\kappa\xi_v K_1(\xi_v(f_{\infty}^2 + 2B\kappa)^{1/2})}{(f_{\infty}^2 + 2B\kappa)^{1/2}} \right], \tag{4}
\end{aligned}$$

where  $K_n(x)$  is a modified Bessel function of  $n$ th order. The first two terms denote the contribution of core energy to magnetization. The last term denotes the contribution of electromagnetic energy.

Two suitable variational parameters  $f_{\infty}$  and  $\xi_v$  to minimize the free energy for arbitrary  $B$  and  $\kappa$  are approximately written as

$$f_{\infty}^2 = 1 - \left[ \frac{B}{\kappa} \right]^4 \tag{5}$$

$$\left[ \frac{\xi_v}{\xi_{v0}} \right]^2 = \left[ 1 - 2 \left( 1 - \frac{B}{\kappa} \right)^2 \frac{B}{\kappa} \right] \left[ 1 + \left( \frac{B}{\kappa} \right)^4 \right] \tag{6}$$

for the cases of  $\kappa > 10$  with  $\kappa\xi_{v0} = \sqrt{2}$ . This model was further extended to include anisotropy by using the effective-mass tensor.

Figure 1 shows  $M(T)$  of Hg1223 for the various external magnetic fields parallel to the  $c$ -axis. The field independent magnetization  $M^*$  appears at  $T^* = 128$  K, which indicates a dominant vortex fluctuation [14,15]. This effect is especially important for highly anisotropic superconductors. In our case, the vortex fluctuation is significantly diminished with decreasing temperature because Hg1223 is only a moderately anisotropic material with anisotropy ratio  $\gamma \simeq 9$  [15].

For theoretical analysis, we chose a set of data  $\{-4\pi M_i, H_i\} (i = 1, 2, \dots)$  at a fixed temperature in range of  $110 \text{ K} \leq T \leq 124 \text{ K}$  as shown in the inset of Fig. 2. If the proper value of  $\kappa$  is chosen, then  $-4\pi M(H)$  could be scaled to a universal curve with scaling factor  $\sqrt{2}H_c(T)$ , consistent with Eq. (4). Our best fit gives  $\kappa = 118 \pm 8$ . Figure 2 shows  $-4\pi M' = -4\pi M / \sqrt{2}H_c(T)$  versus  $H' = H / \sqrt{2}H_c(T)$  from the experimental data and theoretical fitting. All data collapse onto a single curve.

Figure 3 shows  $II_c(T)$  obtained from the above analysis. This data fit to the BCS result [16]

$$\frac{H_c(T)}{H_c(0)} = 1.7367 \left[ 1 - \frac{T}{T_c} \right] \left[ 1 - 0.2730 \left( 1 - \frac{T}{T_c} \right) - 0.0949 \left( 1 - \frac{T}{T_c} \right)^2 \right], \tag{7}$$

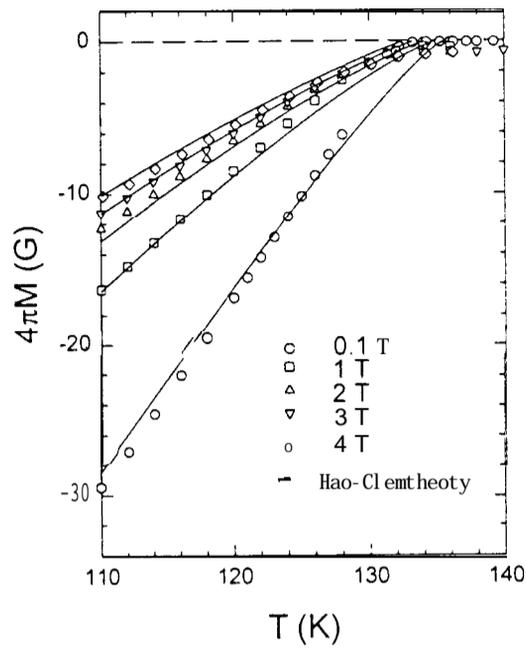


FIG. 1. Temperature dependence of reversible magnetization  $4\pi M(T)$  with the theoretical curves (solid line) for  $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8-x}$  for  $H \parallel c$ .

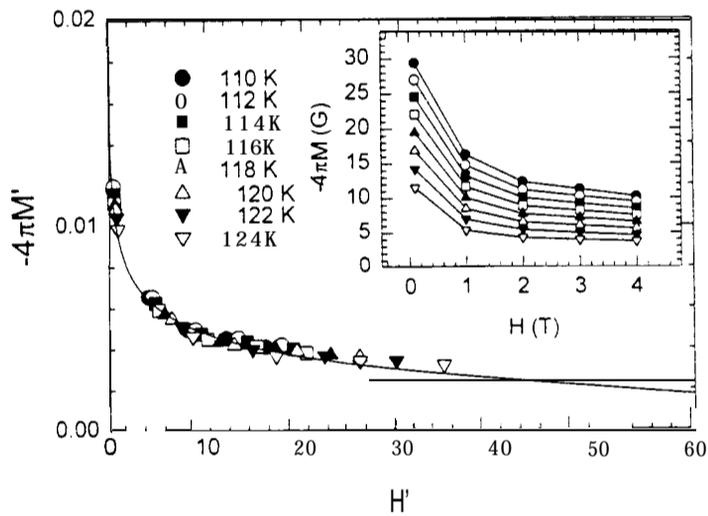


FIG. 2. The curves of  $-4\pi M(H)$  scaled by  $\sqrt{2}H_c(T)$ . Solid line represents the universal curve of the Hao and Clem' model with  $\kappa=118$ . Inset; Magnetization versus magnetic field at various temperatures.

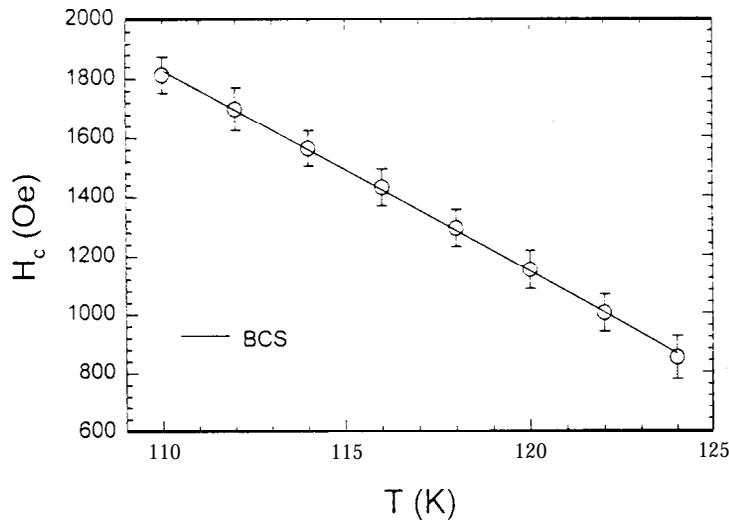


FIG. 3. Temperature dependence of thermodynamic critical field  $H_c(T)$  obtained from the theoretical fitting. Solid line represents the BCS temperature dependence of  $H_c(T)$ .

which yields  $H_c(0) = 5.877 \times 10^3 \pm 52$  Oe with  $T_c = 135.7 \pm 0.2$  K.

From these analyses, the theoretical curve of  $M(T)$  could be obtained. The solid line of Fig. 1 shows calculated  $-4\pi M(T)$ . We note that the experimental data deviates from the theoretical curve for the temperature range  $124 \text{ K} \leq T \leq T_c$ . This may originate from the vortex fluctuation effect, which is important near  $T_c$  [14]. In the vortex fluctuation region of  $124 \text{ K} \leq T \leq T_c$ , the contribution of the distorted vortices to the free energy should be taken into consideration, however for  $110 \text{ K} \leq T \leq 124 \text{ K}$ , the vortex fluctuation effect is not important.

According to the relation  $H_{c2}(T) = \kappa H_c(T)$ , the upper critical field slope  $(\partial H_{c2} / \partial T)_{T_c} = -1.15 \pm 0.02$  T/K was estimated. The slope can be used to estimate the upper critical field at  $T = 0$  by using the WHH formula [17]  $H_{c2}(0) = 0.5758(\kappa_1/\kappa)T_c(dH_{c2}/dT)_{T_c}$ . In the dirty limit  $\kappa_1(0)/\kappa = 1.20$  and in the clean limit  $\kappa_1(0)/\kappa = 1.26$  [17]. From this formula,  $H_{c2}(0)$  is estimated  $108.1 \pm 1.6$  T for the dirty limit but  $113.5 \pm 1.7$  T for the clean limit. The  $\xi_{ab}(0)$  calculated from  $H_{c2}(0) = \phi_0 / 2\pi\xi_{ab}^2(0)$  is  $17.5 \pm 0.14$  Å in the dirty limit and  $\lambda_{ab}(0) = 2059 \pm 15$  Å is extracted from  $\kappa = \lambda/\xi$ . The thermodynamic parameters obtained from this analysis are consistent with scaling results for the high field region [18].

The Hao and Clem' model offers not only above useful parameters, but also information for the field-induced suppression of order parameter. Within the framework, the normalized superfluid density  $\langle f^2 \rangle$  [19] is given by

$$\langle f^2 \rangle = \frac{8}{a^2} \int_0^{a/2} f^2(\rho) \rho d\rho, \quad (8)$$

where  $a$  is a lattice parameter of the vortex array,  $a = 1.075(\phi_0/H)^{1/2}$ . In this calculation, it is assumed that the unit cell of the vortex array is circle. Figure 4 shows the reduced field  $H/H_{c2}$  dependence of  $\langle f^2 \rangle$ . The solid line represents the theoretical curve from Fig. 2. All data follows the universal curve.

Our derived  $\kappa = 118$  for the Hg1223 is much larger than  $\kappa = 57$  of Y123[3]. It is known that  $\kappa$  is proportional to  $\lambda/\xi$  in the clean limit and  $\lambda/l$  in the dirty limit [20]. As Eilenberger [21] has pointed out, the  $\kappa$  might depend not only on  $\lambda/\xi$ , but also on the degree of anisotropy in the impurity scattering. In an other sense,  $\kappa$  reflects the different degree of sensitivity of the various magnetic properties of a superconductor to the degree of nonlocality. Therefore, more detailed theories and experiments are needed to clarify the difference between the two systems.

In summary, we measured the high magnetic field magnetization for the grain-aligned HgBa<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>8-x</sub> for field parallel to the  $c$ -axis. Using Hao and Clem's model, we obtained the Ginzburg-Landau parameter  $\kappa = 118$  and upper critical field slope  $(\partial H_{c2}/\partial T)_{T_c} = -1.15$  T/K, which show the  $\lambda_{ab}(0) = 2060 \text{ \AA}$ , and  $\xi_{ab}(0) = 18 \text{ \AA}$ . Our derived values are consistent with the results from the high magnetic field scaling.

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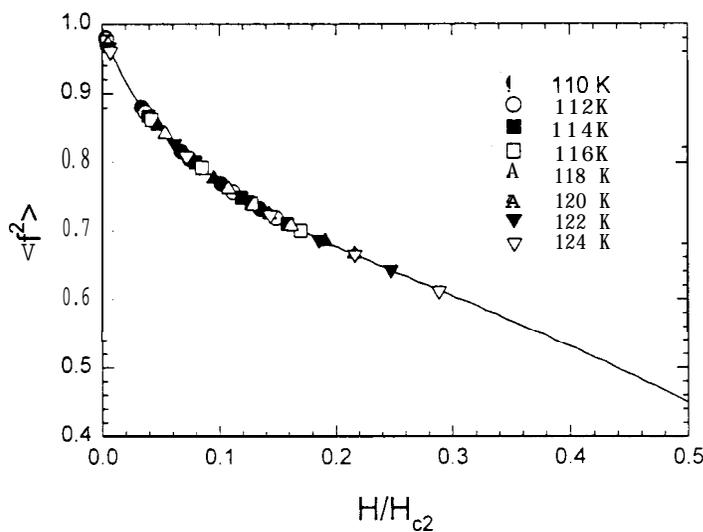


FIG. 4. Magnetic field  $H/H_{c2}$  dependence of normalized superfluid density  $\langle f^2 \rangle$ . Solid line represents the universal curve of the Hao and Clem's model with  $\kappa = 118$ .

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