## Magnetic measurements of the upper critical field, irreversibility line, anisotropy, and magnetic penetration depth of grain-aligned YBa<sub>2</sub>Cu<sub>4</sub>O<sub>8</sub>

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We have measured the upper critical field and the irreversibility line of grain-aligned YBa<sub>2</sub>Cu<sub>4</sub>O<sub>8</sub>, with the magnetic field oriented perpendicular to the CuO<sub>2</sub> planes. The upper critical field's slope,  $dH_{c2}/dT$ , is -1.57 T/K, corresponding to a zero-temperature Ginzburg-Landau (GL) coherence length of 19.5 Å. The irreversibility line obeys a power-law behavior similar to that of 90-K YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>. Using the lower-critical-field data, we obtain the zero-temperature magnetic penetration depth  $\lambda_{ab}(0) = 1960$  Å and GL parameter  $\kappa_c = 100$ .

There is no generally accepted theory for high- $T_c$  superconductors, and so experimental determinations of fundamental physical parameters are needed to clarify the basic mechanism of high- $T_c$  superconductivity. Among high- $T_c$  superconductors, YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> (Y 1:2:3) is one of the most widely studied materials. From its transport, optical, and thermodynamic properties, one can explore the role of the CuO chains in Y 1:2:3,<sup>1-5</sup> which contains two CuO<sub>2</sub> planes and one CuO chain per unit cell. Another system containing CuO chains is YBa<sub>2</sub>Cu<sub>4</sub>O<sub>8</sub> (Y 1:2:4), having two CuO<sub>2</sub> planes and two CuO chains per unit cell. Y 1:2:4 also has a relatively high superconducting transition temperature ( $T_c \sim 80$  K) and excellent thermal stability against the loss of oxygen.<sup>6</sup> Although Y 1:2:4 has important potential applications, there are only a few publications<sup>7-9</sup> on its fundamental properties, such as magnetic anisotropy, critical fields, coherence lengths, and penetration depth.

We report the magnetic anisotropy below  $T_c$ , the upper critical field, magnetic penetration depth, and irreversibility line of grain-aligned Y 1:2:4. All our data were taken with the magnetic field oriented either perpendicular or parallel to the CuO<sub>2</sub> planes. We show that the properties of Y 1:2:4 are similar to those of 90-K Y 1:2:3. The magnetic penetration depth of Y 1:2:4, however, is larger than that of 90-K Y 1:2:3, but is smaller than 60-K Y 1:2:3. In addition, we discuss the possible role of CuO chains in Y 1:2:4, 60-K Y 1:2:3, and 90-K Y 1:2:3.

The superconducting Y 1:2:4 sample was powder, synthesized by a solid-state reaction method. The wellground mixture (with the stoichiometric ratio [Y]:[Ba]:[Cu]=1:2:4) was annealed in 1 atm of oxygen gas at 880°C for two weeks with several intermediate grindings. From x-ray diffraction data, the annealed powder was identified as a mixed phase of Y 1:2:3 and a small amount of CuO. This prereacted powder was pressed into pellets and fired in 185 atm of O<sub>2</sub> at 1000°C for 10 days with several intermediate grindings. This material showed almost entirely a Y 1:2:4 phase with a trace of CuO, but no Y 1:2:3 or Y 2:4:7, determined by x-ray diffraction. We mixed this powder sample, which had an average grain size of 1.5  $\mu m$  in diameter, with epoxy, and cured it in a magnetic field of 8 T at room temperature. The resulting aligned sample was examined by x-ray diffraction. The full width at half maximum (FWHM) of the rocking curve was ~ 1° for the (0012) peak, indicating excellent alignment.

With use of a superconducting quantum interference device (SQUID), magnetic measurements were carried out in an applied field oriented either perpendicular or parallel to the CuO<sub>2</sub> planes. The Meissner and shielding effects were measured in 2 mT for both magnetic-field directions. The upper critical field and irreversibility line were measured with both zero-field cooling (ZFC) and field cooling (FC) in applied magnetic fields between 0.1 and 5 T. An almost temperature-independent background contribution originated from the epoxy; it was subtracted from the observed values to yield the intrinsic M(T, H), which we present and discuss below.

Figure 1 shows the Meissner and shielding effects for a magnetic field of 2 mT oriented both perpendicular and parallel to the CuO<sub>2</sub> planes. The superconducting transition temperature is 79.2 K for both field directions; it was determined from the intercept of the extended line from the maximum slope in M(T) with the M=0 line. At T=10 K, the shielding and Meissner fractions are 67% and 51% for the H  $\parallel c$  axis and 12% and 9% for the H  $\parallel ab$ plane, respectively, without demagnetization factor corrections. Defining the superconducting transition temperature width  $\delta T_c$  as the difference between the temperatures at which the Meissner effect attained 10% and 90% of its maximum value, we find  $\delta T_c \sim 10$  K for both field directions. The magnetic susceptibility anisotropy ratio  $\chi_c(T)/\chi_{ab}(T)$ , where  $\chi_c(T)$  is for the **H**  $\parallel c$  axis and  $\chi_{ab}(T)$  is for the **H** ab plane, is plotted in Fig. 2. The  $\chi(T)$  anisotropy ratio is almost constant below 70 K, with a value of about 5.7, and this value is similar to that of Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub>,<sup>10</sup> but it is larger than that of 90-K Y 1:2:3 (Ref. 11) and the anisotropy of the lower critical field Y 1:2:4.9 The anisotropy ratio  $\chi_c(T)/\chi_{ab}(T)$  increases sharply at temperatures above 70 K, and this anomaly is also observed for grain-aligned Y 1:2:3 and

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FIG. 1. Meissner (solid symbols) and shielding (open symbols) effects for grain-aligned  $YBa_2Cu_4O_8$  samples, for magnetic fields perpendicular (circles) and parallel (squares) to the  $CuO_2$  planes.

 $\operatorname{Bi}_2\operatorname{Sr}_2\operatorname{CuO}_6$ .<sup>10,11</sup> As the temperature approaches  $T_c$  from either direction, the ratio  $\chi_c(T)/\chi_{ab}(T)$  goes to zero. This behavior, which is consistent with previous results,<sup>11,12</sup> shows that  $\chi_c(T) < \chi_{ab}(T)$  for  $T < T_c$  and  $\chi_c(T) > \chi_{ab}(T)$  for  $T > T_c$ .

The ZFC high-magnetic-field magnetization near  $T_c$  for the  $H \parallel c$  axis is shown in Fig. 3. The magnetization has a linear temperature dependence below  $T_c$ , as expected from the high-field Abrikosov theory.<sup>13</sup> M(T,H) has a clearly rounded behavior in the vicinity of  $T_c$ , which can be attributed to the thermodynamic fluctuations or to possible inhomogeneity of the sample. To determine  $T_c(H)$ , we used the reversible regimes in high-field ZFC and FC magnetizations to indicate the interception of the linear fit and the normal-state background line. As pointed out by Hao *et al.*, <sup>14</sup> the exact solution for the magnetization, including the contribution of the wortex cores, is not an exactly linear behavior of the magnetization.



FIG. 2. ZFC (open symbols) and FC (solid symbols) magnetic susceptibility anisotropy ratio  $\chi_c(T)/\chi_{ab}(T)$ , where  $\chi_c(T)$  and  $\chi_{ab}(T)$  are for magnetic fields perpendicular and parallel to CuO<sub>2</sub> planes, respectively.



FIG. 3. Temperature dependence of the ZFC magnetization with the magnetic field perpendicular to the CuO<sub>2</sub> planes;  $H=0.1 \text{ T}(\odot)$ , 0.25 T ( $\bigcirc$ ), 0.5 T ( $\bigtriangledown$ ), 1 T ( $\bigtriangledown$ ), 2 T ( $\square$ ), 3 T ( $\blacksquare$ ), 4 T ( $\triangle$ ), and 5 T ( $\blacktriangle$ ).

versus-temperature plot close to  $T_c$ , but the difference between the two methods<sup>13,14</sup> can be neglected in practice (see below).

In Fig. 3, we regard the zero-magnetization line as a normal-state base line. We find  $T_c(H)$  to be suppressed as the magnetic field increases. Figure 4 shows the  $H_{c2}(T)$  curve for the **H**  $\parallel c$  axis, derived from Fig. 3 by using the interception points between a linear fit to M(T,H) below  $T_c$  and a normal-state background. Because the signal-to-noise was low for the  $H \parallel ab$  plane, we could not repeat the same measurements for that orientation of H. In Fig. 4, there is a strong suppression of  $T_c(H)$  at low fields, which shows an upward curvature, and which is consistent with previous results.<sup>15-17</sup> For H > 0.5 T, we observe almost linear behavior in  $H_{c2}(T)$ up to 5 T. With the magnetic field perpendicular to the CuO<sub>2</sub> planes, a linear fit above H=0.5 T shows a critical field slope  $dH_{c2}/dT$  of  $-1.57\pm0.11$  T/K. Using the Werthamer, Heelfand, and Hohenberg (WHH) formula



FIG. 4. The upper-critical-field slope for H perpendicular to the CuO<sub>2</sub> planes, determined from Fig. 3. The solid line is a linear fit for H > 0.5 T. The inset is a semilogarithmic plot of the slopes of the linear part of the magnetization data vs field, from Fig. 3.

 $H_{c2}(0) \sim 0.7T_c \ [dH_{c2}/dT]$  at  $T = T_c$ ,<sup>18</sup> the extrapolated zero-temperature upper critical field  $H_{c2}(0) \sim 87$  T for the H|| c axis. This upper critical field,  $H_{c2}^c(0) \sim 87$  T for the H|| c axis. This upper critical field,  $H_{c2}^c(0) \sim 87$  T for the H|| c axis. This upper critical field,  $H_{c2}^c(0) \approx 87$  T for the H|| c axis. This upper critical field,  $H_{c2}^c(0) \approx 87$  T for the H|| c axis. This upper critical field,  $H_{c2}^c(0) \approx 87$  T for the H|| c axis. This upper critical field,  $H_{c2}^c(0) \approx 87$  T for the H|| c axis. This upper critical field,  $H_{c2}^c(0) \approx 87$  T for the relation  $H_{c2}^c(0) \approx \phi_0/2\pi\xi_{ab}^2(0)$ , we get the zero-temperature coherence length  $\xi_{ab}(0) \sim 19.5$  Å, which is similar to that of Y 1:2:3.<sup>15,16</sup> Using the high-field Abrikosov theory, we can derive the zero-temperature London penetration depth  $\lambda_{ab}^L(0)$  from the dM/dT versus  $\ln(H)$  relation close to  $T_c$ .<sup>19</sup> In Fig. 4 we plot the dM/dT versus  $\ln(H)$  in the inset. From the relation

$$-M = \phi_0 / [32\pi^2 \lambda^2(T)] \ln(\beta H_{c2}/H)$$

with  $\lambda(t)=0.7\lambda^{L}(0)(1-t)^{-1/2}$ , where  $\lambda(t)$  is the Ginzburg-Landau (GL) penetration depth,  $\lambda^{L}(0)$  is the London penetration depth, and  $\beta$  is a constant of order unity depending on the vortex structure, we use the observed value of dM/dT to obtain  $\lambda^{L}(0) \sim 2470$  Å in the CuO<sub>2</sub> planes.

From the high-magnetic-field ZFC and FC data below  $T_c$ , we determine the irreversibility temperatures for the  $\mathbf{H} \parallel c$  axis, as shown in Fig. 5. We defined the irreversibility temperature  $T^{irr}(H)$  as the temperature at which the difference in the magnetizations between ZFC and FC are less than  $10^{-4} \mu T$ , which is just above the limit of our system's resolution. Excluding the low-magneticfield data, we fit the data points to  $H^{*}(T) = H_0[1 - T^{irr}(H)/T_c(0)]^n$ , and the solid line shows that n=1.51 (see the log-log plot in Fig. 6). This value is close to that of Y 1:2:3.<sup>20,21</sup>

In Fig. 4, we use the high-magnetic-field Abrikosov theory to extrapolate the temperature dependence of  $H_{c2}$ . In Ref. 14, the authors pointed out that the linear-fit method overestimates the upper critical field by about 10%, compared with a more accurate calculation that includes vortex core contributions to the magnetization. Such a 10% error is almost comparable to our uncertainty (7% of the slope in Fig. 4). In addition to that, the



FIG. 5. The irreversibility temperatures vs H for YBa<sub>2</sub>Cu<sub>4</sub>O<sub>8</sub>, where H is perpendicular to the CuO<sub>2</sub> planes. The solid line is a least-squares fit (see text).



FIG. 6. The log-log plot from Fig. 5. The slope of the solid line is n = 1.51.

 $Bi_2Sr_2CaCu_2O_8$  sample shows different behavior<sup>22,23</sup> for M(T,H) close to  $T_c$ , compared to those of other high- $T_c$ superconductors. The origin of the difference is still not clear. It has been reported that  $dH_{c2}/dT \sim -1.1 \text{ T/K}$ , from magnetization measurements<sup>7</sup> on polycrystalline samples, and that  $dH_{c2}^c/dT \sim -0.35$  T/K from resistivity measurements<sup>8</sup> on thin films. Such values are significantly smaller in magnitude than our result. Using the values  $H_{c2}^{c}(0) \sim 87$  T and  $H_{c1}^{c}(0) = 19.7$  mT,<sup>9</sup> and the relation between the upper critical field  $H_{c2}$  and lower critical field  $H_{c1} [H_{c1}/H_{c2} = \ln(\kappa_3)/2\kappa_1\kappa_3]$ , where  $\kappa_1$  and  $\kappa_3$  are the first and third GL parameters, and where  $\kappa_1$ and  $\kappa_3$  are assumed to be approximately the same for high- $\kappa$  superconductors such as high- $T_c$  superconductors,<sup>19</sup> we obtain  $\kappa_3 = \kappa_c \sim 100$  for the **H**  $\parallel c$  axis. Using this  $\kappa_c$  and the relation  $H_{c1}^c \sim \phi_0 / (4\pi \lambda_{ab}^2) \ln \kappa_c$ , we estimate the  $\lambda_{ab}(0) \sim 1960$  Å. This  $\lambda_{ab}(0)$  is much smaller than the value calculated from the slope in Fig. 4, but is similar to the  $\mu$ SR result<sup>24</sup> for polycrystals. The difference between the value from Fig. 4 and the value  $\lambda_{ab}(0) \sim 1960$  Å might arise from the use of the threedimensional (3D) GL theory in the analysis of Fig. 3, even though one might expect the 3D region in Y 1:2:4 to be less than that in Y 1:2:3 because of larger anisotropy.

The estimated value  $\lambda_{ab}(0) \sim 1960$  Å for Y 1:2:4 is between those of 60-K Y 1:2:3 (Ref. 25) and 90-K Y 1:2:3.<sup>26</sup> If we assume the same effective mass  $m_{ab}$  for Y 1:2:3 and Y 1:2:4 and use the Hall-effect-measurement result to determine carrier density,<sup>27</sup> then the  $\lambda_{ab}^{Y1:2:4}(0) \sim 0.9 \lambda_{ab}^{Y1:2:3}(0)$ , which is inconsistent with our result. All three of these systems are almost identical except for CuO chains. We might therefore expect to see almost the same  $\lambda_{ab}(0)$  's if we could neglect the role of CuO chains in the superconductivity. The clear difference among the three  $\lambda_{ab}(0)$  values could therefore be interpreted as showing that the CuO chain is related to the superconductivity. Other evidence for the possible role of the CuO chains in superconductivity comes from Raman-scattering results.<sup>28</sup>

The irreversibility line of Y 1:2:4, which is shown in

Fig. 5, is consistent with the results from the singlecrystal and the grain-aligned 90-K Y 1:2:3, in spite of the different values of the prefactor  $H_0$ , which may be dependent on the sample's quality. The behavior  $H^*(t) \propto [1 - T^{\text{irr}}(H)/T_c(0)]^n$  with  $n \sim 1.51$  is similar to that of Y 1:2:3,<sup>20</sup> but different from those of Tl<sub>2</sub>Ba<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>10</sub>,<sup>29</sup> Bi<sub>2</sub>Sr<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>10</sub>,<sup>30</sup> and PbMo<sub>6</sub>S<sub>8</sub>.<sup>31</sup> Comparing our data with those for 90-K Y 1:2:3, we can speculate that Y 1:2:3 and Y 1:2:4 are very similar in their vortex behavior.

In summary, we have determined the upper critical field, zero-temperature coherence length, magnetic penetration depth, and irreversibility line for a grainaligned  $YBa_2Cu_4O_8$ . In most of these parameters,

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YBa<sub>2</sub>Cu<sub>4</sub>O<sub>8</sub> is similar to 90-K YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>. The magnetic penetration depth, however, is much larger than that of 90-K YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>. Using the lower-critical-field result, we determined the Ginzburg-Landau parameters  $\kappa_c \sim 100$ .

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