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## Upper critical field and resistivity of single-crystal EuBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub>: Direct measurements under high field up to 50 T

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A wide-temperature-range profile of the  $H_{c2}$ -T curve for the superconducting 1:2:3 compound is presented. The data were obtained from a EuBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub> single crystal under a pulsed magnetic field up to 50 T along the c axis.  $H_{c2}$  at 4.2 K is 27.5 ± 2.5 T for the onset of the resistivity. The residual resistivity is roughly estimated to be about 50  $\mu$  Ω cm. The magnetoresistance is positive and the general features of the superconducting properties except  $T_c$  are well sketched by the Bardeen-Cooper-Schrieffer dirty-limit model. The anisotropies in  $H_{c2}$  and the resistivity are also discussed.

It is generally accepted that high magnetic-field studies on the high- $T_c$  superconductors will give much information about the intrinsic properties of the transport phenomena.<sup>1</sup> In practice, however, this has been difficult because the expected  $H_{c2}(T)$  near 0 K is very high and much work has been done without looking at the whole profile of the  $H_{c2}$ -T curve. The experimental studies for single crystals<sup>2-9</sup> and sintered powder samples<sup>10-12</sup> show only the extrapolated  $H_{c2}(T)$  and resistivity curves based on data near  $T_c$ . As the present status of electronic band calculations<sup>13,14</sup> and other various theories<sup>15</sup> do not give a sufficient explanation of the intrinsic mechanism of high- $T_c$  superconductivity, it is highly desirable to have complete information about the electrical and magnetic responses of high- $T_c$  superconducting materials.

The present paper gives the wide-temperature-range profile of the  $H_{c2}$ -T curve and the residual resistivity of 1:2:3 compound. The experiment was performed using a EuBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub> single crystal under a pulsed magnetic field up to 50 T down to liquid-helium temperature. A clear change from the superconducting state is found even at 4.2 K when the magnetic field is applied along the *c* axis. Then, the coherent length  $\xi$  is determined with high accuracy and one can also check the validity of the Werthamer-Helffand-Hohenberg (WHH) theory and the anisotropic Ginzburg-Landau (GL) theory in the 1:2:3 compounds.

 $EuBa_2Cu_3O_y$  single crystals with a mean size of

 $1100 \times 560 \times 22 \ \mu m$  were grown from molton Eu-Ba-Cu-O compounds.<sup>16,17</sup> As-grown crystals were annealed in an oxygen atmosphere at 900-950°C for 5-10 h and then cooled down slowly to room temperature. Annealed crys-tals show a twinning structure.<sup>8,18,19</sup> The largest facet is the a-b plane which is perpendicular to the c axis. Sample resistivity was measured using the usual four-terminal method except for measurement along the c direction. The electrical contact was obtained by using gold wires of 25  $\mu$ m in diameter with conductive silver paste on the gold films evaporated on the largest facet of the crystal. Contact resistances were between  $10^{-2}$  and  $10^{-1} \Omega \text{ mm}^2$  for the area of  $560 \times 170 \ \mu m^2$ . Resistivity measurements without magnetic field were made by using the lowfrequency (73 Hz) ac four-terminal method. Temperature was measured with a calibrated platinum resistance thermometer. A highly qualified single crystal of  $EuBa_2Cu_3O_y$  with a superconducting transition temperature of 94 K was obtained and the transition width was within 1 K. Magnetoresistance was measured by using the dc four-terminal method in a temperature range from 4.2 K to room temperature under a pulsed magnetic field up to 50 T at the Research Center for Extreme Materials in Osaka University where the sample temperature was measured with a calibrated thermocouple of Au-Fe/Ag. The pulsed width of the magnetic field was 0.4 msec. The measurement conditions in the pulsed magnetic field are described in detail in an earlier paper.<sup>11</sup>

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Magnetic-field dependence of the parallel resistance in the *a*-*b* plane with the magnetic field *H* along the *c* axis from 310 to 4.2 K is shown in Fig. 1.<sup>20</sup> The magnetoresistance  $MR_{\perp}$  defined by  $\rho_{\parallel}(T,H) - \rho_{\parallel}(T,0)$  is positive and is approximately linear up to 50 T in the temperature range above  $T_c$ . Figure 2 shows the corresponding  $\rho_{\parallel}$ when *H* is parallel to the *a*-*b* plane. The magnetoresistance in this case,  $MR_{\parallel}$ , is slightly smaller than  $MR_{\perp}$ . However, the negative magnetoresistance observed by Oussena *et al.* in sintered powder YBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub> (Ref. 12) sample was not found in this experiment.

Figure 3 shows the temperature dependence of  $H_{c2\perp}(T)$  and  $H_{c2\parallel}(T)$  obtained from Figs. 1 and 2 where ⊥ and ∥ represent the magnetic-field directions referred to the *a*-*b* plane. The  $H_{c2}(\rho = 0)$  is defined as magnetic field at the onset point of the resistance shown by notation A in Fig. 2. The estimated  $H_{c2}(\rho = \frac{1}{2})$ , which is normally used for sintered powder samples, is also shown.<sup>21</sup> The  $H_{c2}(\rho = \frac{1}{2})$  is defined as notation B shown in Fig. 2. The  $H_{c2\perp}(T)$  curve exhibits a slightly upward curvature near  $T_c$  which is very often observed in layered superconductors.<sup>22,23</sup> The value of  $H_{c2\perp}(0)$  is estimated to be  $27.5 \pm 2.5$  T. This value is close to 26.5 T which is evaluated by the WHH theory, i.e., the dirty-limit Bardeen-Cooper-Schrieffer (BCS) relation of  $H_{c2}(0) = 0.69T_c$  $\times (dH_{c2}/dT)$  (Ref. 24) with the observed  $dH_{c2\perp}/dT$  of 0.41 T/K obtained around  $T/T_c = 0.65 - 0.85$ . This value is quite close to the value estimated by Worthington et al.<sup>3</sup> This means that the WHH theory is effective in the case of this 1:2:3 compound. On the other hand,  $H_{c2\parallel}(T)$ drastically increases with decreasing temperature and is measured only above 79 K. The extrapolated  $H_{c2\parallel}(0)$  using the same relation is  $245 \pm 20$  T where  $dH_{c2\parallel}/dT$  is 3.8 T/K around  $T/T_c = 0.8 - 0.9$ . The  $H_{c2\parallel}(T)$  also exhibits upward curvature near  $T_c$ .

The  $dH_{c2\parallel}/dT$  value of 3.8 T/K in the present EuBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub> crystal is almost the same as the previously reported values of 3.8 T/K,<sup>7</sup> 3.3 T/K,<sup>6</sup> and 3.9 T/K (Ref.



FIG. 1. Magnetic-field dependence of the resistivity  $\rho$  in the *a-b* plane at various temperatures. The field is applied along the *c* axis.



FIG. 2. Magnetic-field dependence of the resistivity in the a-b plane, when the field is in the plane. Definitions of the onset point A and the midpoint B are schematically shown.

9) for single-crystal YBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub> measurements while  $dH_{c2\perp}/dT$  obtained in the present study is 0.41 T/K which differs from the previous values of 0.54 T/K (Ref. 7) and 0.56 T/K,<sup>6</sup> and 1.1 T/K.<sup>9</sup> According to the WHH theory,  $dH_{c2}/dT$  is strongly dependent on resistivity. The difference of  $dH_{c2\perp}/dT$  in various single crystals seems to depend on  $\rho$ . The observed  $\rho$  in the present study near  $T_c$  is 170  $\mu$   $\Omega$  cm (-0.41 T/K) which is lower than the previously reported value of 800  $\mu$   $\Omega$  cm (-1.1 T/K) for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub>.<sup>8</sup> It may be said that highly qualified material shows a low  $dH_{c2\perp}/dT$  value. However, Orlando *et al.* investigated the relation between  $\rho$  and  $dH_{c2}/dT$  by using sintered powder samples and concluded that the



FIG. 3. Upper-critical field  $H_{c2}$  vs temperature T for singlecrystal EuBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub>.

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FIG. 4. Temperature dependence of the normal resistivity in the *a-b* plane. Error bars come from the ambiguity of the substraction of magnetoresistance. The inset shows the resistivity near  $T_c$  without magnetic field. The dashed straight line is a linear extrapolated value from the higher-temperature data above 170 K.

resistivities were not an intrinsic property of the superconductivity because the  $dH_{c2}/dT$  ws not a function of resistivity.<sup>10</sup> The detailed measurements using single crystals will be necessary for understanding the relation between  $dH_{c2}/dT$  and  $\rho$ .

The coherent length tensor is calculated from the estimated  $H_{c2\perp}(0)$  and  $H_{c2\parallel}(0)$  data using the relations given by  $H_{c2\perp} = \Phi_0/2\pi\xi_{\parallel}^2$ ,  $H_{c2\parallel} = \Phi_0/2\pi\xi_{\parallel}\xi_{\perp}$ , where  $\Phi_0$  is the flux quantum. The obtained values are  $\xi_{\parallel}(0) = 35$  Å,  $\xi_{\perp}(0) = 3.8$  Å.

It should be noted that two-dimensional behavior is not ruled out because  $\xi_{\perp}$  is much smaller than the unit-cell length along the *c* axis.<sup>25</sup> The observed  $\xi_{\perp}$  value is critical<sup>26</sup> even if the Cu-O chain were responsible for the superconductivity. Although three-dimensional behavior is reported by Freitas *et al.*,<sup>27</sup> the possibility of twodimensional character and dimensional crossover should be investigated in the future. The observed anisotropic ratio of  $H_{c2\parallel}/H_{c2\perp}$  is 9.3 at 80 K in accordance with previously reported data<sup>28</sup> of the reciprocal ratio,  $H_{c1\perp}/H_{c1\parallel} = 10$ . This fact means that the anisotropic GL theory can be applicable to the 1:2:3 compounds.

The zero-field normal resistivity curve in the *a-b* plane can be roughly estimated by combining the high-field results with the data above  $T_c$  as is shown in Fig. 4, where the observed linear and positive magnetoresistance is substracted from the high-field data and the residual resistivity is estimated as about 50  $\mu\Omega$  cm.<sup>29</sup> With the exception of magnetoresistance the outline of  $\rho(T)$  can be under-



FIG. 5. Temperature dependence of the resistivity tensor measured by the Montgomery method where terminals B, C, E, and F are used for the cross measurement. The dashed line is obtained by the four-terminal method where terminals A, B, C, and D are used.

stood by the normal-metal model. The linear magnetoresistance is a little unusual but is sometimes found in magnetic materials.<sup>30</sup> The spin-fluctuation effect, i.e., negative magnetoresistance<sup>31</sup> is not found.

The anisotropic resistivity above  $T_c$  is also measured and the results are shown in Fig. 5. The Montgomery method<sup>32</sup> was used to separate each resistivity. The temperature dependence roughly agrees with the observed data of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub> single crystal given by Tozzer *et al.*<sup>33</sup> The resistivity along the *c* axis is semiconductive near  $T_c$ . This observed peculiarity should be investigated in the future.

In conclusion, it should be noted that a high-quality single crystal of EuBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub> shows a clear type-II superconductivity under a strong magnetic field up to 50 T. The wide-temperature-range profile of the  $H_{c2}$ -T curve shows that the WHH theory based on the BCS dirty-limit model well explained the experimental results whose coherent lengths were  $\xi_{\parallel}=35$  Å and  $\xi_{\perp}=3.8$  Å. The estimated in-plane normal resistivity down to liquid-helium temperature does not show any peculiarity and the magnetoresistance is positive. The remaining or rather enhanced problem is why the superconducting critical temperature  $T_c$  is so high in such a "normal" metal.

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- <sup>20</sup>For checking upon the critical current, the applied current of 81 and 45 mA was used for measurements at 77 K. Both results coincided in experimental accuracy. Therefore, the lower current was used in the measurements at the other temperature. The resistivity limit is less than 1  $\mu \Omega$  cm for the measurements.
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