

# Fourfold symmetric anisotropy in the $\text{CuO}_2$ planes of 60-K $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals

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(Received 29 October 2000; published 11 April 2001)

To study the anisotropy in the  $\text{CuO}_2$  planes, the in-plane resistivity  $\rho$  has been measured on 60 K  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) single crystal as a function of the angle  $\theta$  between the directions of a magnetic field  $H$  and of the  $b$  axis under magnetic fields precisely parallel to the  $\text{CuO}_2$  planes. Angular dependence of the resistivity  $\rho(\theta)$  is mainly described by the fourfold symmetry, and a dip structure appears in  $\rho(\theta)$  when the magnetic field is applied parallel to the  $a$  and  $b$  axes. The fourfold symmetry of the in-plane anisotropy of the resistivity is first observed in YBCO system. It is shown that the obtained results have a very close relation to the anisotropy which comes from the superconducting energy gap of the  $d$ -wave pairing with  $d_{x^2-y^2}$  symmetry and the effect of the directional vortex pinning.

DOI: 10.1103/PhysRevB.63.172506

PACS number(s): 74.72.Bk, 74.25.Fy, 74.60.Ge

## I. INTRODUCTION

In high- $T_c$  superconductors, the anisotropy in the  $\text{CuO}_2$  planes has been investigated by measuring the angular dependence of the resistivity in magnetic fields parallel to the  $\text{CuO}_2$  planes, because it has been expected that it would give information for the symmetry of Cooper pairs. Previous studies<sup>1-5</sup> indicate that the in-plane anisotropy of the resistivity  $\rho(\theta)$  depends strongly on the interplane anisotropy of the system defined by the anisotropic parameter  $\gamma$  ( $=\xi_{ab}/\xi_c$ ) as follows, where  $\xi_{ab}$  and  $\xi_c$  are the coherence length in the  $ab$  plane and along the  $c$  axis direction, respectively. In 90 K YBCO single crystal ( $\gamma\sim 6$ )  $\rho(\theta)$  showed clearly the twofold anisotropy.<sup>1</sup> On the other hand, no anisotropic behavior of  $\rho(\theta)$  was observed in  $c$  axis oriented  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  (BSCCO) thin film ( $\gamma\sim 200$ ).<sup>2</sup> Ikeda pointed out that the former behavior comes from the superconducting fluctuation<sup>6</sup> and the latter one is explained as that the system is practically in the zero field owing to the extremely high upper critical field  $H_{c2}$ .<sup>7</sup> In materials which have intermediate  $\gamma$ , for instance,  $\text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4$  single crystal ( $\gamma\sim 20$ ) (Ref. 3) and  $\text{Pb}_2\text{Sr}_2\text{Y}_{0.62}\text{Ca}_{0.38}\text{Cu}_3\text{O}_8$  one ( $\gamma\sim 12$ ),<sup>4</sup> the fourfold anisotropy clearly appeared. Very recently, Iwasaki *et al.*<sup>5</sup> found that  $\rho(\theta)$  of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO) single crystals with various  $\gamma$  is well described by the superposition of the twofold and fourfold symmetry and the contribution of the fourfold component correlates obviously with  $\gamma$ . Takanaka and Kuboya<sup>8</sup> theoretically proposed that the in-plane  $H_{c2}$  is proportional to  $\cos 4\theta$  in the  $d$ -wave pairing state; this gives the fourfold in-plane anisotropy of the resistivity. Therefore, it has been believed that the fourfold symmetry observed in  $\rho(\theta)$  could be understood by the anisotropy of the superconducting energy gap of the  $d$ -wave pairing state.

The results reported previously suggest a possibility that the fourfold anisotropy appears in  $\rho(\theta)$  of more anisotropic YBCO such as 60 K phase crystal ( $\gamma\sim 20$ ). Furthermore, this possibility is supported by that the fourfold in-plane an-

isotropy was observed in the magnetic torque measurements on 90 K YBCO,<sup>9</sup> though it has not been found in those of the resistivity. In this paper, we report on the in-plane anisotropy of the resistivity of 60 K YBCO in magnetic fields precisely parallel to the  $\text{CuO}_2$  planes. We succeeded in observation of the fourfold anisotropy in  $\rho(\theta)$  of the YBCO system. We show that the obtained fourfold symmetry in  $\rho(\theta)$  is basically described by the anisotropy originated from the  $d_{x^2-y^2}$  symmetry of the  $d$ -wave pairing state and that the directional vortex pinning occurs at  $H\parallel a$  and  $b$  axes.

## II. EXPERIMENT

YBCO single crystals were grown by a self-flux method in yttria crucibles. Twin planes of a crystal were removed by annealing at 450 °C for 24 h under an uniaxial stress of about  $1\times 10^{-7}$  N/m<sup>2</sup>; the detwinning process was observed by the *in situ* technique using polarized optical microscope. The detailed procedures of crystal growth and of evaluating the absence of twin planes were described elsewhere.<sup>10</sup> To obtain 60 K phase sample, the detwinned crystal was annealed at 690 °C for 7 days in flowing oxygen atmosphere, followed by quenching into liquid nitrogen. The sample dimensions are  $0.4\times 0.4\times 0.07$  mm<sup>3</sup> and the superconducting transition temperature  $T_c$ , defined as a temperature at the zero resistivity, is 65.1 K, respectively. The anisotropic parameter  $\gamma$  of the crystal is determined to be  $\sim 20$  from scaling of the set of the resistivity curves measured as a function of the angle  $\phi$  under several magnetic fields up to 90 kOe by the reduced field  $H_{\text{red}}=H(\sin^2\phi+\gamma^{-2}\cos^2\phi)^{1/2}$ , based on the effective mass model.<sup>11</sup>

The experimental setup of the resistivity measurements is schematically illustrated in the upper inset of Fig. 1. In-plane resistivity is measured by a conventional dc four-probe method and the direction of the applied current  $I$  is parallel to the  $b$  axis. We use the two-axis rotatable unit, and the angles  $\theta$  and  $\phi$  are defined between the field and the  $b$  axis and between the field and the  $c$  axis, respectively. To obtain the resistivity as a function of the angle  $\theta$  in magnetic fields

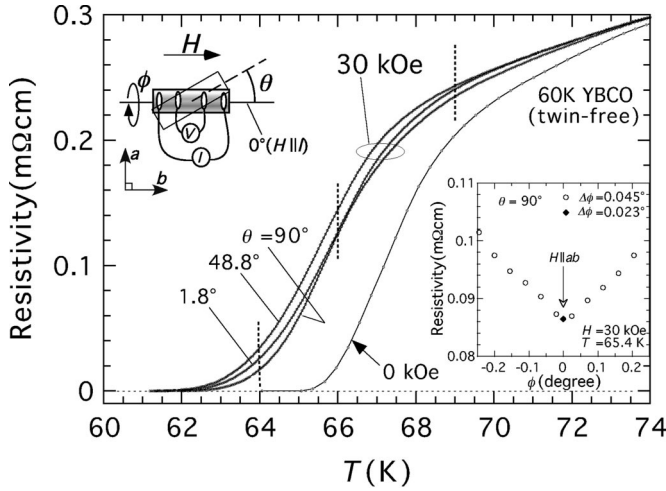


FIG. 1. Temperature dependence of the resistivity of the untwinned 60 K YBCO at  $H=30$  kOe for several angles  $\theta=1.8^\circ$ ,  $48.8^\circ$ , and  $90^\circ$  and at  $H=0$  kOe. The broken lines indicate the temperatures where we estimate  $\theta$  dependence of the resistivity shown in Fig. 2. The lower inset shows  $\phi$  dependence of the resistivity at  $H=30$  kOe and at  $T=65.4$  K for  $\theta=90^\circ$ ;  $\phi=0^\circ$  represents that the field is exactly parallel to the  $ab$  planes.

accurately parallel to the  $ab$  planes, we carried out the following procedure. We first measure the resistivity as a function of the angle  $\phi$  with a step  $\Delta\phi=0.023^\circ$  for finding the position of  $H\parallel ab$  planes, because the misalignment of the field with the  $ab$  planes gives rise to that  $\rho(\theta)$  is dominated by the interplane anisotropy; the interplane anisotropy is usually much larger than the in-plane one in the  $\text{CuO}_2$  plane, for instance, in an untwinned 90 K YBCO, the former  $\gamma$  is 8–9 and the latter  $\gamma$  is 1.2.<sup>9</sup> Next the temperature dependence of the resistivity is measured. This sequence is repeated for several angles  $\theta$ . Finally,  $\rho(\theta)$  curve at a constant temperature is given from the  $\rho(T)$  curves. The angle resolution in this study is small enough for adjusting the position, because it is well known that vortices are confined between the  $ab$  planes below the so-called lock-in angle, and it is less than of about  $0.2^\circ$  in 60 K YBCO.<sup>12</sup>

### III. RESULTS

In the lower inset of Fig. 1 the angular dependence of the resistivity  $\rho(\phi)$  of the untwinned crystal at  $H=30$  kOe and  $T=65.4$  K for  $\theta=90^\circ$  is shown as an example for finding the position of  $H\parallel ab$  planes.  $\rho(\phi)$  curve takes on a “V shape” in the narrow angle region, and its tip defined as  $\phi=0^\circ$  corresponds to the just parallel configuration; the final position is determined with the angle resolution  $\Delta\phi=0.023^\circ$ . Main panel of Fig. 1 shows the temperature dependence of the resistivity of the untwinned crystal at  $H=30$  kOe for several angles  $\theta$ . The obtained  $\rho(T)$  curves depend obviously on the angle  $\theta$ , though these show the similar temperature dependence. In the transition region the resistivity gradually approaches to the zero resistivity with decreasing temperature and the discontinuous feature due to the first-order melting transition cannot be observed at all.

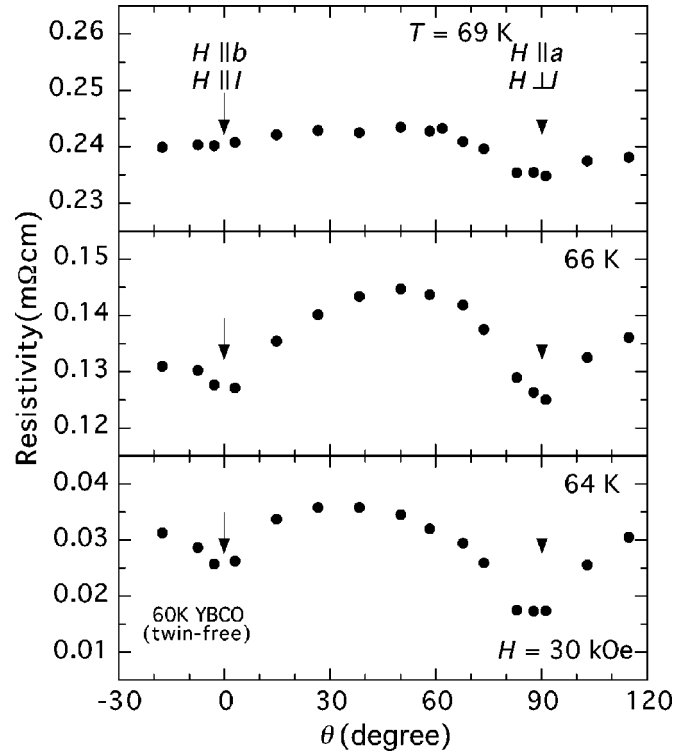


FIG. 2. Angular dependence of the resistivity of the untwinned 60 K YBCO at  $H=30$  kOe for several temperatures, from top to bottom,  $T=69$ ,  $66$ , and  $64$  K. The left scale of each window is the same.  $\theta=0^\circ$  and  $90^\circ$  indicate  $H\parallel I$  and  $H\perp I$ , respectively.

Figure 2 represents the angular dependence of the resistivity  $\rho(\theta)$  of the untwinned crystal at  $H=30$  kOe for several temperatures; these curves are estimated from the  $\rho(T)$  curves shown in Fig. 1. The  $\rho(\theta)$  curves take a maximum near  $\theta=45^\circ$  and minima at around  $\theta=0^\circ$  ( $H\parallel b$ ) and  $90^\circ$  ( $H\parallel a$ ), where  $\theta=0^\circ$  and  $90^\circ$  correspond to the configuration  $H\parallel I$  and  $H\perp I$ , respectively. Moreover, an observable dip structure, which was previously found in the LSCO system,<sup>5</sup> accompanies the resistivity minima. The structures at around  $\theta=0^\circ$  and  $90^\circ$  become more and more visible at lower temperatures, and the resistivity value at  $\theta=0^\circ$  is larger than that at  $\theta=90^\circ$  below 72 K.

### IV. DISCUSSION

The obtained  $\rho(\theta)$  curves seem to be mainly represented by a fourfold anisotropic mechanism. According to the previous reports,<sup>3–5,8</sup> the most plausible origin of the fourfold anisotropy of  $\rho(\theta)$  is the anisotropy due to the superconducting energy gap of the  $d$ -wave pairing. Takanaka and Kuboya<sup>8</sup> theoretically pointed out that the angular dependence of the upper critical field is described as  $H_{c2}(\theta) \propto \cos 4\theta$  in the  $d$ -wave pairing state. Because the midpoint of the resistive transition in a finite magnetic field has been empirically regarded as  $H_{c2}$ , the experimental results could be explained using this theory; a  $H_{c2}$  maximum and minimum give a resistivity minimum and maximum, respectively. If the  $d_{x^2-y^2}$  pairing is assumed,  $H_{c2}(\theta)$  takes maxima at  $H\parallel[\pm 1,0,0]$  and  $[0,\pm 1,0]$  and minima at  $H\parallel$

$[\pm 1, \pm 1, 0]$ , while the  $d_{xy}$  one gives  $H_{c2}$  maxima and minima at opposite configuration of the former. Therefore, the fourfold symmetry observed in our results are qualitatively consistent with the  $H_{c2}$  anisotropy due to the  $d_{x^2-y^2}$  pairing symmetry. However, it should be emphasized that the dip structure and the difference between  $\rho(\theta=0^\circ)$  and  $\rho(\theta=90^\circ)$  could not be represented by the  $\cos 4\theta$  dependence. Thus, let us discuss the origin of the dip structure in  $\rho(\theta)$  curves below.

When the directions of the magnetic field and of the current is different, the vortices are driven by the Lorentz force, which gives rise to the energy dissipation. Since the magnitude of the Lorentz force varies with the angle  $\theta$  in our experimental configuration, it should be considered that the angular dependence of the energy dissipation because it could affect the resistivity. It is well known that the Lorentz force picture gives the twofold anisotropy of the resistivity as  $\rho(\theta) \propto \sin^2 \theta$ . This  $\sin^2 \theta$  dependence surely succeeded in describing the  $\rho(\theta)$  behaviors of 90 K YBCO,<sup>1</sup> but failed to explain the angle independent behavior in BSCCO.<sup>2</sup> Therefore, the Lorentz force scenario seems to be inadequate to describe the  $\rho(\theta)$  behaviors.

According to a theory<sup>6</sup> based on the superconducting fluctuation, the resistivity as a function of the angle between the field and current directions is given as

$$\rho(\theta, I \| E) = \rho_{\parallel} + (\rho_{\perp} - \rho_{\parallel}) \sin^2 \theta, \quad (1)$$

where  $E$  is the electric field, and

$$\rho_{\perp} \equiv \rho(H \perp I) = [\sigma_b^N + \sigma_{\perp}^{\parallel}(H/\gamma)]^{-1},$$

$$\rho_{\parallel} \equiv \rho(H \parallel I) = [\sigma_b^N + \sigma_{\parallel}^{\parallel}(H/\gamma)]^{-1}.$$

Here  $\sigma_b^N$  is the contribution of the normal conductivity, and  $\sigma_{\perp}^{\parallel}$  and  $\sigma_{\parallel}^{\parallel}$  are the fluctuation conductivity for  $H \perp I$  and for  $H \parallel I$ , respectively. Equation (1) obviously gives the  $\sin^2 \theta$  dependence, i.e., twofold anisotropy, when both fluctuation components are effectively larger than the normal one and indicates that the contribution of the twofold anisotropy tends to decrease with increasing  $\gamma$ . Therefore, it is no wonder that the twofold anisotropy reduces in  $\rho(\theta)$  of relatively highly anisotropic materials such as 60 K YBCO. Consequently, this theory cannot also explain the nature of the dip structure, but gives the reason that the twofold component is absent.

The theories concerned with the  $d$ -wave pairing<sup>8</sup> and with the superconducting fluctuation<sup>6</sup> cannot explain the dip structure and the difference between  $\rho(\theta=0^\circ)$  and  $\rho(\theta=90^\circ)$ . Thus, we must introduce another mechanism for describing these features. This dip structure strongly suggests that a directional pinning potential exists along the directions of the  $a$  and  $b$  axes, because it is well known that the angular dependence of the resistivity often shows a dip structure as a result of the pinning effect caused by planar or linear objects such as the  $\text{CuO}_2$  planes,<sup>13</sup> twin planes,<sup>14</sup> and columnar defects.<sup>15</sup> Figure 3 shows the temperature dependence of the activation energy, which is given by the local slope in the Arrhenius plots  $d(\ln \rho)/d(T^{-1})$ , in the untwinned crystal at  $H=30$  kOe for several angles  $\theta$ . The activation energy at

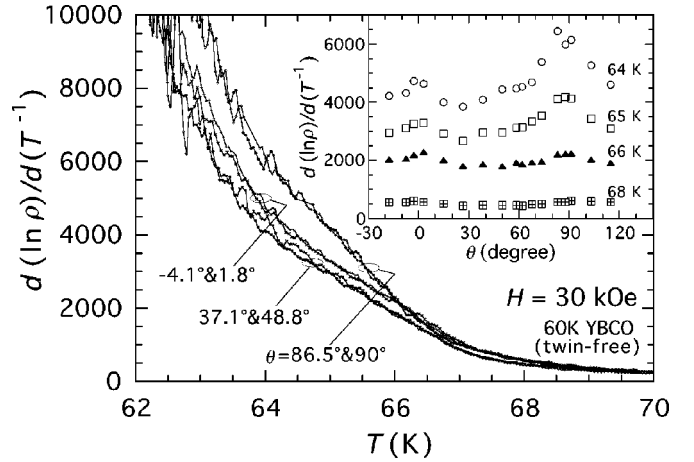


FIG. 3. Temperature dependence of the activation energy, given by  $d(\ln \rho)/d(T^{-1})$ , of the untwinned 60 K YBCO at  $H=30$  kOe for several angles  $\theta$ . Inset shows the angular dependence of the activation energy for several temperatures.

around  $\theta=0^\circ$  and  $90^\circ$  is always larger than that at around  $45^\circ$  except for high temperatures. With decreasing temperature, the activation energy gradually increases, and then rapidly increases below 67 K. Especially, near  $\theta=90^\circ$  which corresponds to  $H \parallel a$  axis, it begins to drastically increase in comparison with other configuration below 66 K. In the inset of Fig. 3 the angular dependence of the activation energy at several temperatures is represented. The activation energy depends clearly on the angle  $\theta$  and takes cusplike maxima at around  $\theta=0^\circ$  and  $90^\circ$ . These features obviously indicate that the vortex pinning is considerably effective when the magnetic field is applied parallel to the  $a$  and  $b$  axes; especially the former direction. The similar directional pinning effect in the  $ab$  planes was observed in the magnetic torque measurements on untwinned single crystals of 90 K YBCO.<sup>9</sup> We can conclude that the picture of the directional vortex pinning bring about the dip structure and the difference between  $\rho(\theta=0^\circ)$  and  $\rho(\theta=90^\circ)$ .

Finally, we make a comment on the relationship of the observation of the fourfold symmetry with  $\gamma$ . The fourfold anisotropy of the resistivity did not appear in 90 K YBCO,<sup>1</sup> but in 60 K YBCO. Contrary to the twofold one, thus, the fourfold one is enhanced with increasing  $\gamma$ , which is previously observed in the LSCO system.<sup>5</sup> One possible origin is  $\gamma$  dependence of the contribution of the fluctuation conductivity,<sup>6</sup> because we suppose that in the small  $\gamma$  systems the large twofold component masks the fourfold one. Another is attributed to the layered structure of high- $T_c$  materials. The enhancement of  $\gamma$  corresponds to that the coupling between the  $\text{CuO}_2$  planes weakens, therefore vortices which are aligned parallel to the  $\text{CuO}_2$  planes must more strictly be confined between them, i.e., the intrinsic pinning<sup>16</sup> becomes rather effective. Because the vortices cannot freely wander across the plane in such a situation, it is expected that the intrinsic nature of the in-plane anisotropy more visibly appears in  $\rho(\theta)$ , indicating that the fourfold symmetric behavior is easily observed in materials with relatively large  $\gamma$ . To confirm the validity of these scenarios, we need to

check whether the fourfold anisotropy appears in BSCCO using high quality single crystals.

### V. CONCLUSIONS

We have studied the anisotropy in the  $\text{CuO}_2$  planes from measuring the angular dependence of the resistivity when the magnetic field is applied just parallel to them. The angular dependence of the resistivity is mainly represented by the fourfold symmetric component which is qualitatively described by the  $H_{c2}$  anisotropy due to the superconducting energy gap of the  $d_{x^2-y^2}$  pairing symmetry. The dip structure in the resistivity curves was observed when the magnetic field is applied parallel to the  $a$  and  $b$  axes and we showed that the vortex pinning is considerably effective at  $H\parallel a$  and  $b$  axes in comparison with other configuration. The in-plane

fourfold symmetry of the resistivity was first observed in the YBCO system by using a 60 K YBCO single crystal. Therefore, we insist that the fourfold anisotropy in the  $\text{CuO}_2$  planes is the intrinsic phenomena of high- $T_c$  cuprates which have  $d$ -wave pairing with  $d_{x^2-y^2}$  symmetry.

### ACKNOWLEDGMENTS

The authors would like to thank the members of Low Temperature Science Division, Center for Low Temperature Science, Tohoku University. The experiments were performed at Institute for Materials Research, Tohoku University. This work was in part supported by a Grant for Basic Science Research Projects (No. 000539) from the Sumitomo Foundation.

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