Fourfold symmetry in the ab plane of the upper critical field for single-crystal Pb2Sr2Y0.62Ca0.38Cu308: Evidence for dx2-y2 pairing in a high-Tc superconductor

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Fourfold symmetry in the *ab* plane of the upper critical field for single-crystal $Pb_2Sr_2Y_{0.62}Ca_{0.38}Cu_3O_8$: Evidence for $d_{x^2-y^2}$ pairing in a high-T_c superconductor

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We have found clear anisotropy in the *ab* plane with fourfold symmetry of the upper critical field for single-crystal $Pb_2Sr_2Y_{0.62}Ca_{0.38}Cu_3O_8$, which is very similar to that formerly observed in $La_{1.86}Sr_{0.14}CuO_4$ [Physica B **165&166**, 1449 (1990)]. This is neither attributed to the twin structure in the orthorhombic phase nor to the anisotropy of the Fermi surface but is explained as being mainly due to the anisotropy of the superconducting energy gap owing to $d_{x^2-y^2}$ pairing, which supports $d_{x^2-y^2}$ pairing in a high-T_c superconductor. [S0163-1829(96)51426-8]

I. INTRODUCTION

Recently, the symmetry of the pairing state of Cooper pairs has attracted great interest in the high-T_c cuprates, because it is related to the mechanism of high-T_c superconductivity. Many kinds of experimental studies, such as NMR,[1] low-temperature specific heat,[2] Raman spectroscopy,[3] and neutron inelastic scattering,[4] have been devoted to the symmetry. According to these studies, it appears that $d_{x^2-y^2}$ pairing is superior to d_{xy} or s pairing. As for the upper critical field H_{c2}, clear anisotropy in the *ab* plane with fourfold symmetry has been observed for the La-based superconductor $La_{1.86}Sr_{0.14}CuO_4$ by Hanaguri *et al.*[5] Very recently, it has been theoretically pointed out by Takanaka and Kuboya[6] that the fourfold symmetry of H_{c2} is consistent with the $d_{x^2-y^2}$ pairing.

In this paper, in order to confirm the generality of the fourfold symmetry of H_{c2} in the *ab* plane among the high-T_c cuprates, we investigate the anisotropy of H_{c2} in the *ab* plane for the Pb-based superconductor $Pb_2Sr_2Y_{0.62}Ca_{0.38}Cu_3O_8$, whose crystal structure is characterized by double CuO_2 pyramidal layers and a thick blocking layer consisting of the stack of SrO-PbO-Cu-PbO-SrO sheets along the *c* axis. Here, the anisotropy of H_{c2} is estimated by measuring the resistive superconducting transition under magnetic fields applied in the *ab* plane.

II. EXPERIMENT

Single crystals of $Pb_2Sr_2Y_{0.62}Ca_{0.38}Cu_3O_8$ were grown from molten materials using excess PbO and NaCl as flux. The details are described in our previous paper.[7,8] Typical dimensions of the single crystals obtained were $1\times1\times0.1$ mm^3.

The single crystals were orthorhombic and cloven on the (001) plane and also on the (100) and (010) planes in the orthorhombic index. The single crystals were twinned with the twin boundary on the (110) plane,[9,10] but the twin boundary could not be seen so clearly using a polarizing microscope as seen in $YBa_2Cu_3O_{7-\delta}$. This may be due to a small quantity of orthorhombicity in $Pb_2Sr_2Y_{0.62}Ca_{0.38}Cu_3O_8$, compared with that in $YBa_2Cu_3O_{7-\delta}$.

The electrical resistivity along the *c* axis, ρ_c, was measured by a dc four-point probe method, in order to study the resistive superconducting transition. Two lead wires were attached to each surface (the *ab* plane) of a platelike single crystal with silver paste; one wire was for the current and the other for the voltage. The single crystal was rotated with the rotational axis of the *c* axis under magnetic fields so that magnetic fields were always applied in the *ab* plane perpendicular to the current direction. Temperature measurements were made with a Pt thermometer calibrated in the absence and presence of magnetic field. In the measurements under magnetic fields at a constant temperature, the temperature was controlled within ±20 mK by a capacitance temperature controller.

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of ρ_c in zero field. The superconducting transition temperature T_c, defined as the midpoint of the superconducting transition curve, is 75.5 K.

Figure 2 displays the angular dependence of ρ_c, normalized by the maximum value of ρ_c in zero field $\rho_{c,n}$ (77.6 K), under various magnetic fields H at various temperatures,

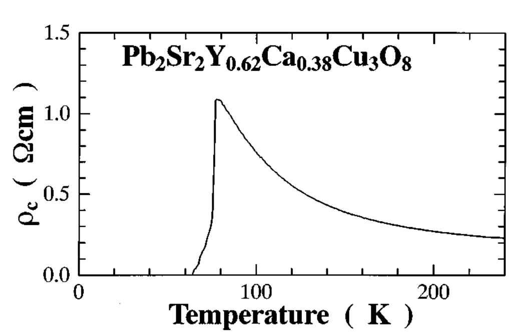

FIG. 1. Temperature dependence of the electrical resistivity along the *c* axis, ρ_c, in zero field for $Pb_2Sr_2Y_{0.62}Ca_{0.38}Cu_3O_8$.

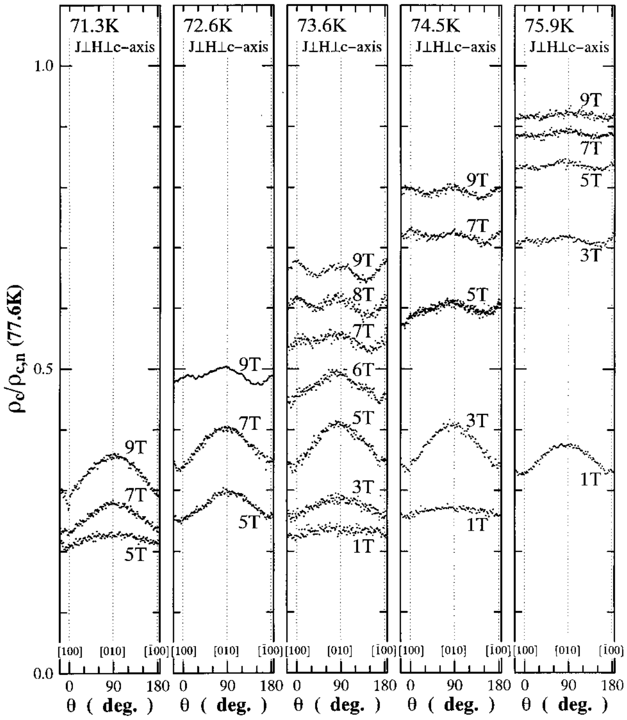

FIG. 2. Angular dependence of ρ_c, normalized by the maximum value of ρ_c in zero field $\rho_{c,n}$ (77.6 K), under various magnetic fields at various temperatures for $Pb_2Sr_2Y_{0.62}Ca_{0.38}Cu_3O_8$, where θ is the angle in the ab plane between the [100] direction in the orthorhombic index and the field direction.

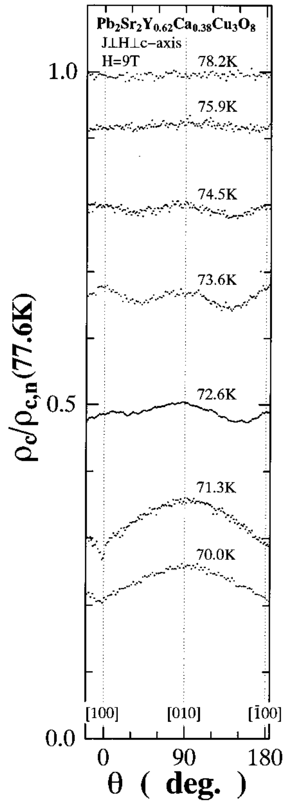

FIG. 3. Angular dependence of ρ_c, normalized by the maximum value of ρ_c in zero field $\rho_{c,n}$ (77.6 K), in 9 T at various temperatures for $Pb_2Sr_2Y_{0.62}Ca_{0.38}Cu_3O_8$, where θ is the angle in the ab plane between the [100] direction in the orthorhombic index and the field direction.

where θ is the angle in the *ab* plane between the [100] direction and the field direction. Only the data in 9 T are again shown in Fig. 3. It is found that twofold symmetry is observed in the low-resistance region, namely, in the tail region of the superconducting transition, On the other hand, fourfold symmetry is observed in the middle-resistance region, namely, in the middle region of the superconducting transition. Furthermore, ρ_c is almost independent of θ in the high-resistance region, namely, in the onset region of the superconducting transition. It is found that the symmetry is not so dependent on the field strength but on the resistance.

In the high-T_c cuprates, H_{c2} is not regarded as a phase transition point but a crossover point on account of thermal disorder, so that it is difficult to determine H_{c2} experimentally. However, it is empirically known that the mean-field value of H_{c2} is in approximate agreement with the H_{c2} value defined as the midpoint in the resistive superconducting transition.[11,12] Therefore, the fourfold symmetry of ρ_c in the middle region of the superconducting transition is regarded as the fourfold symmetry of H_{c2}. Then, it is found that H_{c2} takes maxima for $H \| [\pm 1, \pm 1, 0]$ and minima for $H \| [\pm 1,0,0]$ and $[0,\pm 1,0]$ in the orthorhombic index. Using the tetragonal index where directions along Cu-O-Cu in the CuO_2 plane are expressed as $[\pm 1,0,0]_t$ and $[0,\pm 1,0]_t$, H_{c2} takes maxima for $H \| [\pm 1,0,0]_t$ and $[0,\pm 1,0]_t$ and minima for $H \| [\pm 1,\pm 1,0]_t$, where the suffix t means the tetragonal index. This fourfold symmetry is qualitatively similar to that formerly observed in $La_{1.86}Sr_{0.14}CuO_4$.[5] The magnitude of the fourfold component of H_{c2}, $\Delta H_{c2}/H_{c2}$, defined as $2(H_{c2}^{max}-H_{c2}^{min})/(H_{c2}^{max}+H_{c2}^{min})$, can be estimated from the temperature dependence of ρ_c under constant magnetic fields to be 0.060 at $T/T_c = 0.96$, on the reasonable assumption that H_{c2} is proportional to $1-T/T_c$ near T_c. This value is not far from 0.086 at $T/T_c = 0.95$ in $La_{1.86}Sr_{0.14}CuO_4$. Therefore, it appears that the origin of the fourfold symmetry in $Pb_2Sr_2Y_{0.62}Ca_{0.38}Cu_3O_8$ is the same as in $La_{1.86}Sr_{0.14}CuO_4$.

Concerning the origin of the fourfold symmetry of H_{c2} in the *ab* plane, three possibilities are considered at present. The first is due to the twin structure in the orthorhombic phase, as mentioned in Sec. II. If H_{c2} in the orthorhombic phase has twofold symmetry in the *ab* plane, fourfold symmetry of H_{c2} resulting from the superposition of the twofold symmetry is expected from the twin structure. However, this inevitably leads to maxima of H_{c2} for $H \| [\pm 1,\pm 1,0]_t$, which is contradictory to the experimental results. The second is due to pseudofourfold symmetry of the Fermi surface. Although the Fermi surface of the high-T_c cuprates is still controversial, it is supposed to have pseduofourfold symmetry more or less when it exists. Formerly, in fact, the fourfold symmetry of H_{c2} resulting from the fourfold symmetry of the Fermi surface was observed for very pure single-crystals of Nb and V, which are regarded as conventional superconductors with *s* pairing.[13,14] However, it was found that the magnitude of $\Delta H_{c2}/H_{c2}$ is at most ~0.1 at a very low temperature of $T/T_c = 0.1$ and that it drastically decreases with increasing temperature. Therefore, it appears rather hard to ascribe the large anisotropy near T_c for the high-T_c cuprates to only the anisotropy of the Fermi surface. The third is due to the fourfold symmetry of the superconducting energy gap owing to *d* pairing, which has been theoretically pointed out as mentioned in Sec. I.[6] According to the theory, directions where H_{c2} takes maxima are different between $d_{x^2-y^2}$ pairing and d_{xy} pairing. The experimental results are consistent with $d_{x^2-y^2}$ pairing. Moreover, the theory gives the relation that $\Delta H_{c2}/H_{c2}$ increases with decreasing temperature as expressed as $\Delta H_{c2}/H_{c2} \sim 0.2(1-T/T_c)$. This is qualitatively consistent with the experimental results that $\Delta H_{c2}/H_{c2}$ at 73.6 K (H=9 T) is larger than that at 75.9 K (H=3 T). Quantitatively speaking, the theoretically expected value of $\Delta H_{c2}/H_{c2}$, 0.008 at T/T_c=0.96, is somewhat smaller than the experimental value. This may be attributed to oversimplification of the theory where the Fermi surface is assumed to be isotropic in the *ab* plane. It will be possible that the fourfold symmetry of H_{c2} is explained not only qualitatively but also quantitatively, taking into account the anisotropy of the Fermi surface in the *ab* plane.

Here, it may be worthwhile pointing out that the fourfold symmetry of ρ_c in the middle region of the superconducting transition does not seem to be related to the flux lattice melting transition. This is because the melting transition is usually observed in the tail region of the superconducting transition and does not affect the resistivity in the middle region, even if it exists.[15] For the present crystal, in fact, no drastic change of the resistivity due to the melting transition has been observed.

In the onset region of the superconducting transition, the fourfold symmetry may be smeared out because of superconducting fluctuation.

As for the twofold symmetry in the tail region of the superconducting transition, it is due to misalignment of the setting of a single crystal under magnetic fields, because the position of the dip of ρ_c as a function of θ changes, depending on the crystal setting. In general, when anisotropy between in-plane and out-of-plane properties is very large, twofold symmetry with respect to the angular dependence appears on account of such misalignment that the *ab* plane is not precisely parallel to the field direction. In the case of the resistive superconducting transition of the high-T_c cuprates, the resistance tail is known to be due to thermally activated flux creep[16] and to be very anisotropic between $H \parallel ab$ plane and $H \perp ab$ plane.[17] Accordingly, the twofold symmetry appears to be markedly observed in the tail region of the resistive transition. In fact, we have not succeeded in observing the fourfold symmetry even in the middle region of the superconducting transition for $Bi_2Sr_2CaCu_2O_{8+\delta}$, on account of much larger anisotropy between in-plane and out-of-plane properties than the present material and the La-based superconductor.

IV. CONCLUSIONS

We have found clear anisotropy in the *ab* plane with fourfold symmetry of the resistive superconducting transition under magnetic fields, namely, of H_{c2} for single-crystal $Pb_2Sr_2Y_{0.62}Ca_{0.38}Cu_3O_8$, which is not only qualitatively but also quantitatively similar to that formerly observed in $La_{1.86}Sr_{0.14}CuO_4$. This is neither attributed to the twin structure in the orthorhombic phase nor to the anisotropy of the Fermi surface, but is explained as being mainly due to the anisotropy of the superconducting energy gap owing to $d_{x^2-y^2}$ pairing. This supports $d_{x^2-y^2}$ pairing in the high-T_c superconductivity.

To be more conclusive, further investigations are necessary, for example, detailed studies on the temperature dependence of the anisotropy of H_{c2} in the *ab* plane using higher magnetic fields or experiments using other high-T_c cuprates such as $YBa_2Cu_3O_{7-\delta}$ and $Bi_2Sr_2CaCu_2O_{8+\delta}$.

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