Novel Anisotropy in the Superconducting Gap Structure of $Bi_2Sr_2CaCu_2O_{8+\delta}$ Probed by Quasiparticle Heat Transport

Yoichi Ando,* J. Takeya, Yasushi Abe, X. F. Sun, and A. N. Lavrov

Central Research Institute of Electric Power Industry, Komae, Tokyo 201-8511, Japan
(Received 15 November 2001; published 26 March 2002)

Since the nature of pairing interactions is manifested in the superconducting gap symmetry, the exact gap structure, particularly any deviation from the simple $d_{x^2-y^2}$ symmetry, would help in elucidating the pairing mechanism in high-T_c cuprates. Anisotropic heat transport measurement in $Bi_2Sr_2CaCu_2O_{8+\delta}$ reveals that the quasiparticle populations are different for the two nodal directions and thus the gap structure must be uniquely anisotropic, suggesting that pairing is governed by interactions with a rather complicated anisotropy. Intriguingly, it is found that the "plateau" in the magnetic-field dependence of the thermal conductivity is observed only in the b-axis transport.

It has been established [1] that in the high-T_c cuprates the superconducting gap Δ has essentially the $d_{x^2-y^2}$ symmetry, $\Delta(\mathbf{k}) = \Delta_0 \cos 2\phi$, which is depicted in Fig. 1 (Δ_0 is the maximum gap and ϕ is the azimuthal angle). Although this information is very important, it does not help much in clarifying the high-T_c mechanism, because all of the proposed theories of the high-T_c superconductivity have successfully adopted the d-wave symmetry. It is thus important to obtain more detailed information on the gap structure, because any deviation from the simple $d_{x^2-y^2}$ symmetry would point to the role of a particular interaction in the paring. However, not much is known about the precise gap structure of the high-T_c cuprates [2], partly because the experimental techniques to selectively probe the nodal structures are limited [3].

The most prominent feature of the d-wave gap is that it has four nodes where the gap magnitude vanishes. At finite temperature below T_c, quasiparticles (QPs) are always thermally excited near these nodes from the superconducting condensate; how many QPs are excited depends on the "slope" of the gap at the node [4], because a steeper gap allows less phase space available below a given energy of $k_B T$. Since these QPs carry heat (in contrast to the Cooper pairs that do not carry heat), one can investigate the gap structure of unconventional superconductors by measuring the QP heat transport in the superconducting state [5]. Moreover, because the QPs can travel only along the nodal directions [5], one can gain information about different nodes by tuning the direction of the heat current to the respective nodal directions; this makes it possible to study the detailed anisotropy of the gap structure in high-T_c cuprates.

In recent years, the heat transport has been conveniently used to probe the gap structure of the high-T_c cuprates. For example, it was reported [6,7] that the in-plane thermal conductivity showed a fourfold-symmetric change when the heat current was fixed along an antinodal direction and an applied magnetic field was 360° rotated in the CuO_2 plane, giving maxima for fields parallel to the nodal di-

rections. This feature helped in establishing the $d_{x^2-y^2}$ symmetry of the superconducting gap. The heat transport measurement was also employed in an intriguing proposal of a field-induced transition in the gap structure; namely, Krishana *et al.* reported [8] that at low temperatures the magnetic-field (H) dependence of the thermal conductivity of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO) showed a "plateau" in high fields, and they proposed that this plateau region may correspond to a fully gapped state where there are no QPs to carry heat. This experiment attracted a lot of attention because the field-induced phase transition in the superconducting state has a significant implication on the mechanism of the superconductivity; however, the origin of the plateau, as well as the reproducibility of the experimental
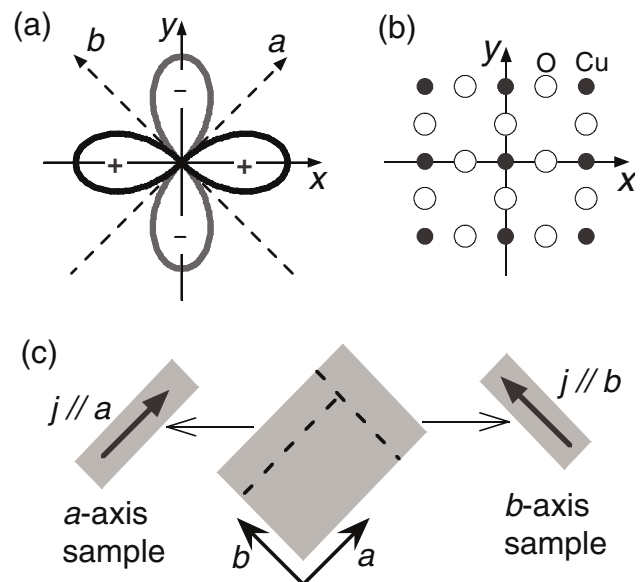

FIG. 1. Schematic pictures of the $d_{x^2-y^2}$-wave gap and the present experiment. (a) Shape of the gap, $\Delta(\mathbf{k}) = \Delta_0 \cos 2\phi$, is plotted together with the directions of the a and b crystallographic axes of BSCCO. (b) Structure of the two-dimensional CuO_2 plane. (c) Two samples used for the measurements of the a- and b-axis transports are essentially the same crystal.

result, are currently under intense debate (see Ref. [9], and references therein). In this Letter, we report a striking in-plane anisotropy of the QP heat transport, which not only sorts out the "plateau" issue but also suggests an unexpected anisotropy in the superconducting gap symmetry.

We measure the thermal conductivity κ of high-quality BSCCO single crystals using a conventional steady-state technique [9,10]. The crystals used here are grown by the floating-zone (FZ) technique [11] and are annealed to be slightly underdoped (zero-resistance T_c is 90.5 K); their crystallographic axes are determined by the x-ray Laue analysis within the accuracy of a few degrees. To obtain reliable data on the in-plane anisotropy, we cut a piece of high-quality single-domain crystal into three parts and measure the a and b axis transports separately [Fig. 1(c)]; both the resistivity (ρ_a and ρ_b) and the thermal conductivity (κ_a and κ_b) are measured on the same samples. Note that in BSCCO the a and b crystallographic axes are along the nodal directions [Fig. 1(a)]. The magnetic field is applied perpendicular to the ab plane and the H dependence of κ is measured with the field-cooled procedure [9] to avoid complications that are associated with the vortex-pinning-related hysteresis [12].

Figure 2 displays the temperature dependences of ρ and κ for the a and b directions; the resistivity shows only a small, if any, in-plane anisotropy, while the thermal conductivity demonstrates a nontrivial anisotropy that cannot be attributed to an error in the geometrical factors; namely, the peak height at $\sim$70 K is smaller along the a axis and yet κ_a becomes larger than κ_b below $\sim$30 K. This anisotropy in $\kappa(T)$ suggests that the number of thermally excited QPs is larger for the nodes along the a axis, because the QP population near the nodes is more directly reflected in κ at lower temperatures [13]. (The anisotropy in κ above T_c is most likely due to phonons; it has been estimated [9] that phonons are responsible for roughly 2/3 of the total κ at T_c.) We note that the uncertainties in the absolute values in Fig. 2 are less than 10% for both ρ and κ.

The magnetic-field dependence of κ reveals more striking in-plane anisotropy of the heat transport. Figure 3 shows the result of our precise H-dependence measurement (where the relative accuracy is $\sim$0.1% [9]) for κ_a and κ_b at 12.5 and 10 K up to 6 T. Interestingly, only κ_b shows the plateau-like feature, indicating that there cannot be a true "fully-gapped" state where all the nodes are lifted. (If a "full gap" causes the QPs to disappear, there should be no QP heat transport in *any* direction.)

To further investigate the magnetic-field dependence of κ down to lower temperatures and up to higher magnetic fields, we have measured κ_a and κ_b in a ^{3}He refrigerator equipped with a 16 T magnet (with a bit worse signal-to-noise ratio). Figures 4(a) and 4(b) show the H dependences of κ_a and κ_b, respectively, down to 0.36 K and up to 16 T; the trend is clear that κ_a tends to decrease with H while κ_b tends to flatten or increase with H. Note that, since the phonons contribute to the heat transport in paral-
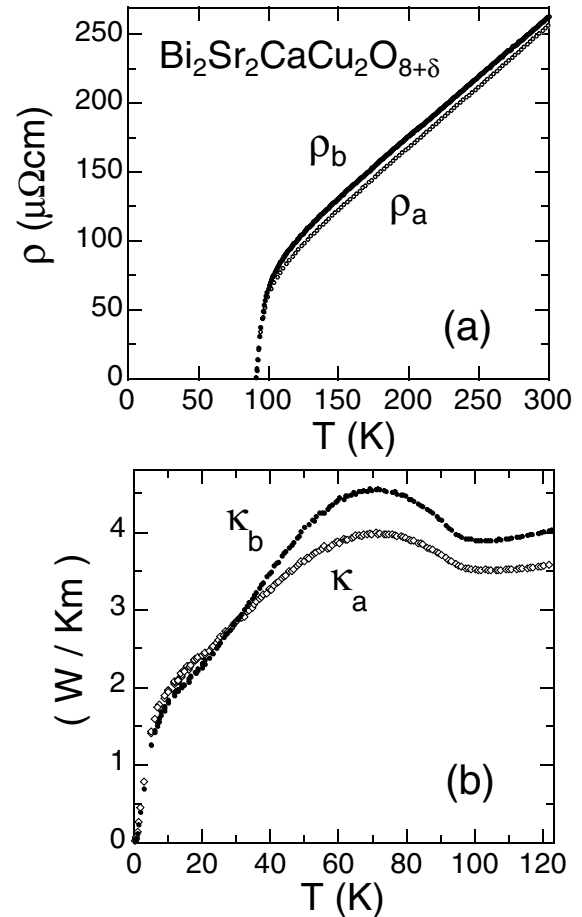

FIG. 2. Anisotropic transport in zero field. (a) Resistivity shows a very small, if any, in-plane anisotropy, indicating that the electronic properties are essentially isotropic in the normal state. (b) Thermal conductivity κ shows a peculiar in-plane anisotropy below T_c that suggests a difference in the QP populations along the two nodal directions.

lel to the QPs, the observed thermal conductivity is a sum of the QP term κ_{QP} and the phonon term κ_{ph}. For BSCCO, it has been discussed [8,9,12] that more than 90% of the total κ is due to κ_{ph} around 10 K and this κ_{ph} is independent of H; thus, although the H dependence of the total κ is rather weak in Figs. 4(a) and 4(b), κ_{QP} is actually changing a lot with H.

The physical origin of this anisotropic behavior can be inferred by plotting κ_a and κ_b at the same temperature together, taking the absolute values for the vertical axis [Figs. 4(c)–4(e)]. In the simple kinetic theory, the QP thermal conductivity κ_{QP} is proportional to both the number of QPs available for the heat transport, N_{QP}, and their mean free path l_{QP}; namely, $\kappa_{QP} \propto N_{QP} l_{QP}$. In d-wave superconductors, the quantized vortices produced by applied magnetic fields are expected to *both* induce QPs near the nodes *and* scatter them [14–16]. If there are already a large number of QPs along the a axis in zero field, the increase in N_{QP} due to the vortices may be inconsequential, while the vortex scattering will cause a noticeable reduction in l_{QP}; as a result, κ_{QP} along the a axis would tend to decrease with increasing H. On the other hand,
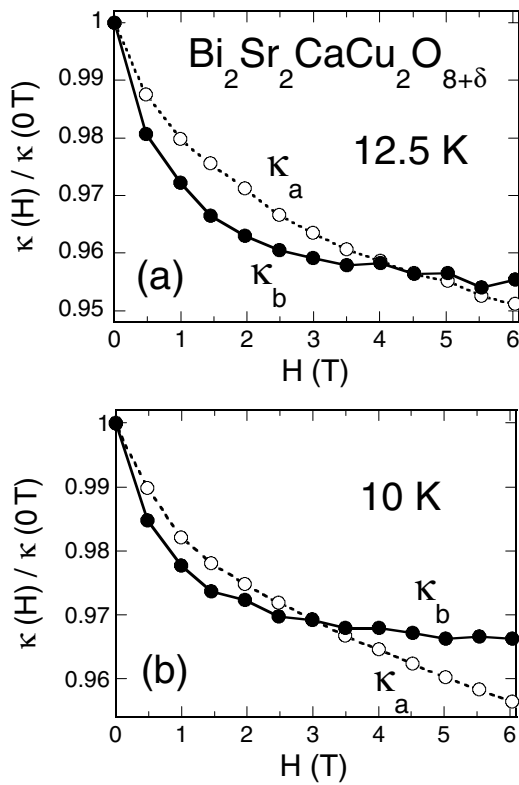
FIG. 3. Magnetic-field dependences of the anisotropic thermal conductivity at (a) 12.5 K and (b) 10 K. The "plateau" is observed only for the b direction. The data are normalized by the values in zero field. (At 12.5 K, κ_a and κ_b are 2.07 and 1.96 W/Km; at 10 K, κ_a and κ_b are 1.96 and 1.81 W/Km).

if there are only a small number of QPs along the b axis in zero field, both N_{QP} and l_{QP} along the b axis will be noticeably changed due to the vortices and the changes in N_{QP} and l_{QP} will tend to cancel each other, leading to a weaker H dependence along the b axis. [Actually, one of the probable explanations of the "plateau" in $\kappa(H)$ is that it comes from an exact cancellation of the two effects [15].] This phenomenology explains the behaviors of $\kappa_a(H)$ and $\kappa_b(H)$ in Figs. 4(c) and 4(d) (7.5 and 3.0 K) and is consistent with their relative magnitude. At lower temperature [Fig. 4(e)], it appears that the changes in N_{QP} dominate over the changes in l_{QP}, causing κ to increase with H. (Unfortunately, more quantitative analysis of the data is hindered by the lack of proper understanding of the vortex scattering of the QPs in the cuprates, which remains rather controversial [15,16].)

Thus, the novel anisotropies in both the temperature dependence and the magnetic-field dependence of the thermal conductivity consistently tell us that there are more QPs near the nodes along the a axis; this gives strong evidence that the gap structure notably deviates from the $d_{x^2-y^2}$ symmetry. Mixing of a small s-wave component, which the orthorhombic distortion may cause [17], would *not* produce the observed anisotropy. One simple possibility to make the gap consistent with the observed anisotropy is to introduce "higher harmonics"; for example, for a gap with an expression $\Delta(\mathbf{k}) = \Delta_0 \cos 2\phi - \Delta_1 \sin 4\phi$, the slope at
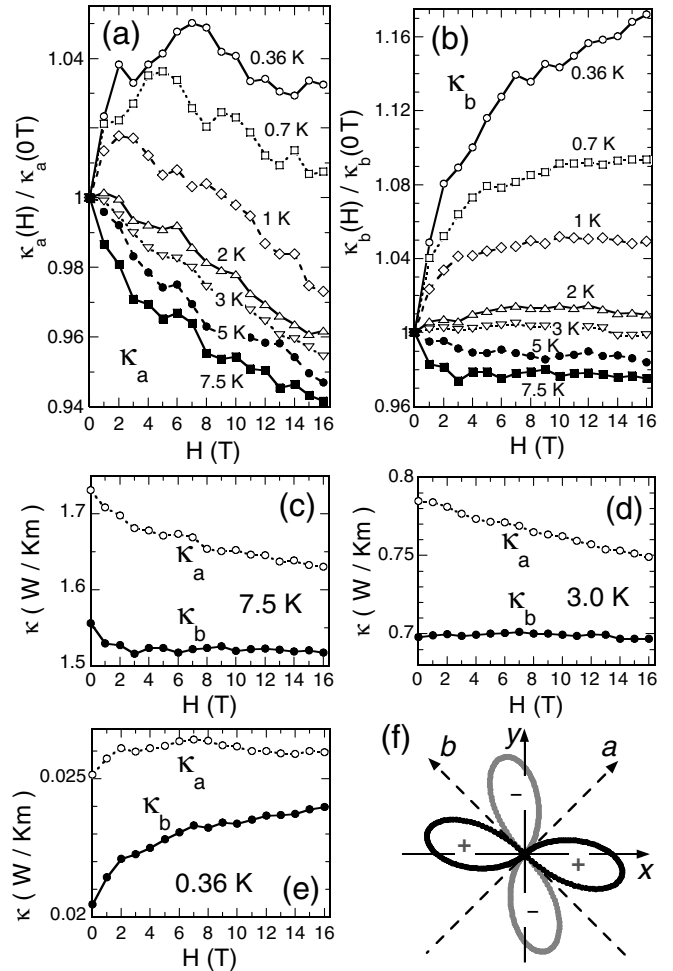

FIG. 4. Magnetic-field dependences of the anisotropic thermal conductivity at lower temperatures and in higher fields. (a),(b) H dependences of κ_a and κ_b at various temperatures down to 0.36 K. (c)–(e) Comparisons of κ_a and κ_b in absolute magnitude at representative temperatures. (f) Possible shape of the gap suggested by the data; the slope of the gap at the b-axis nodes is steeper than that at the a-axis nodes.

the nodes is gentle along the a axis and steeper along the b axis [Fig. 4(f)] [18]. Of course, some nonanalytic modulation of the $d_{x^2-y^2}$ symmetry may also be possible.

Since the reproducibility of the plateau in $\kappa(H)$ has been a matter of debate [9], we comment on the reproducibility issue. We reported previously that the plateau was not observed in the majority of samples [9]; recently, we found that this majority of samples was cut along the a axis, because our crystals usually grow along the a axis in the FZ furnace. In fact, the $\kappa(H)$ data published in Ref. [9] agree with the κ_a data reported here. To check whether our salient observation reported here is reproducible, we have measured another b-axis sample (sample B2) that was taken from a different batch where the crystal grew along the b axis. We have also measured a sample (sample M) which was cut along the direction 45° rotated from the a axis [Cu-O-Cu bond direction, see Fig. 1(b)]. As shown in Fig. 5, sample B2 (which is optimally doped with $T_c = 95$ K) reproduces the behavior of
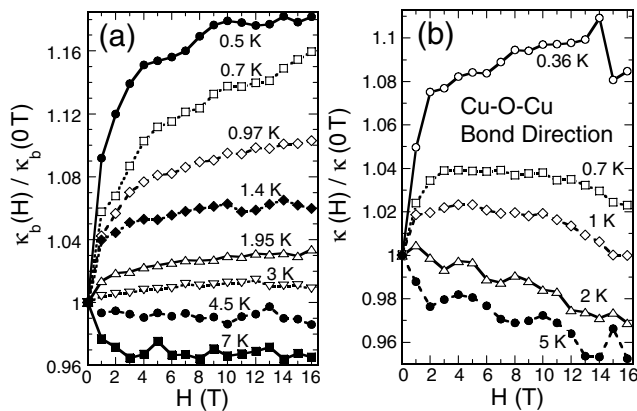
FIG. 5. *H* dependences of κ in (a) the second *b*-axis sample (sample B2) and (b) a sample cut along the Cu-O-Cu bond direction (sample M).

the first *b*-axis sample [Fig. 4(b)], and sample M shows the behavior which is essentially an average of $\kappa_a(H)$ and $\kappa_b(H)$. These results give confidence in the reproducibility as well as the intrinsic nature of our observation.

It is useful to note that the angle-resolved photoemission experiments have reported almost completely fourfold-symmetric intrinsic Fermi surface for BSCCO [19], and therefore the novel anisotropy of the gap found here is not likely to be due to a Fermi surface anisotropy [20]. (In fact, the normal-state resistivity shows negligibly small in-plane anisotropy [Fig. 2(a)], which is consistent with the almost-isotropic Fermi surface.) Therefore, the striking anisotropy in the gap structure suggests that the driving force of the gapping, or the "glue" to bind the electrons into Cooper pairs, is anisotropic itself. From this point of view, it would be difficult to imagine that the antiferromagnetic interaction on the nearly square CuO_2 planes is wholly responsible for the pairing, though it is often considered to be the most likely candidate. On the other hand, the phonons are expected to be anisotropic in BSCCO because of the pronounced modulation structure [21], and thus the role played by the phonons in pairing is an interesting possibility. Another possibility is a role of the charge inhomogeneity [22], which could be related to the isotropic or nematic phases of the electronic liquid crystal [23,24], though the impact of such inhomogeneous structure on the gapping and on the QP behavior is not understood yet. In any case, the novel anisotropy in the gap structure suggested by the present experiment should help in clarifying how the superconducting gap is formed in high-T_c superconductors.

In summary, we have measured the thermal conductivity κ of BSCCO along the *a* and *b* axes in essentially the same high-quality single crystal down to 0.36 K. It is found that the temperature dependence *and* the magnetic-field dependence of κ both show peculiar in-plane anisotropy, and the observed anisotropy is consistent with the interpretation that there are more QPs near the nodes along the *a* axis. This result gives evidence that the *d*-wave gap is uniquely distorted in BSCCO, which suggests that the electron pairing is contributed by the

interaction that develops a pronounced anisotropy between the *a* and *b* axes. In addition, our measurement clarifies that only κ_b shows the "plateau" in its magnetic-field dependence; this new information should help in elucidating the true origin of this intriguing plateau.

*Electronic address: ando@criepi.denken.or.jp
[1] C. C. Tsuei and J. R. Kirtley, Physica (Amsterdam) **282C–287C**, 4 (1997).
[2] J. Mesot *et al.,* Phys. Rev. Lett. **83**, 840 (1999).
[3] In principle, the angle-resolved photoemission should be able to resolve the **k** dependence of the gap around the Fermi surface; however, in BSCCO, measurements along the *a* direction are complicated due to the existence of the shadow bands.
[4] A. C. Durst and P. A. Lee, Phys. Rev. B **62**, 1270 (2000).
[5] M. J. Graf *et al.,* Phys. Rev. B **53**, 15 147 (1996).
[6] F. Yu *et al.,* Phys. Rev. Lett. **74**, 5136 (1995).
[7] H. Aubin *et al.,* Phys. Rev. Lett. **78**, 2624 (1997).
[8] K. Krishana, N. P. Ong, Q. Li, G. D. Gu, and N. Koshizuka, Science **277**, 83 (1997).
[9] Y. Ando, J. Takeya, Y. Abe, K. Nakamura, and A. Kapitulnik, Phys. Rev. B **62**, 626 (2000).
[10] J. Takeya, I. Tsukada, Y. Ando, T. Masuda, and K. Uchinokura, Phys. Rev. B **62**, R9260 (2000).
[11] Y. Ando and K. Nakamura, Phys. Rev. B **59**, R11 661 (1999).
[12] H. Aubin, K. Behnia, S. Ooi, and T. Tamegai, Science **280**, 9a (1998).
[13] P. J. Hirschfeld and W. O. Putikka, Phys. Rev. Lett. **77**, 3909 (1996).
[14] C. Kübert and P. J. Hirschfeld, Phys. Rev. Lett. **80**, 4963 (1998).
[15] M. Franz, Phys. Rev. Lett. **82**, 1760 (1999).
[16] I. Vekhter and A. Houghton, Phys. Rev. Lett. **83**, 4626 (1999).
[17] Q. P. Li, B. E. C. Koltenbah, and R. Joynt, Phys. Rev. B **48**, 437 (1993).
[18] The component "$\sin 4\phi$" would not occur in a simple expansion of the gap function in Fermi surface harmonics and thus is highly unusual; however, the most natural harmonic "$\cos 6\phi$" [2], which is consistent with the $d_{x^2-y^2}$ symmetry, does not produce the observed anisotropy.
[19] Z.-X. Shen and D. S. Dessau, Phys. Rep. **253**, 1 (1995).
[20] The "shadow" Fermi surfaces, which show up anisotropically, are caused by the extrinsic effect of the modulations in the Bi-O layers [21] and thus are not a sign of a true Fermi surface anisotropy.
[21] Y. Le Page, W. R. McKinnon, J.-M. Tarascon, and P. Barboux, Phys. Rev. B **40**, 6810 (1989).
[22] S. H. Pan *et al.,* Nature (London) **413**, 282 (2001).
[23] S. A. Kivelson, E. Fradkin, and V. J. Emery, Nature (London) **393**, 550 (1998).
[24] Y. Ando, K. Segawa, S. Komiya, and A. N. Lavrov, Phys. Rev. Lett. **88**, 137005 (2002).