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Thermal conductivity measurements of the energy-gap anisotropy of superconducting LaFePO at low temperatures

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The superconducting gap structure of LaFePO (T_c=7.4 K) is studied by thermal conductivity (κ) at low temperatures in fields H parallel and perpendicular to the c axis. A clear two-step field dependence of $\kappa(H)$ with a characteristic field H_s(~350 Oe) much lower than the upper critical field H_{c2} is observed. In spite of the large anisotropy of H_{c2}, $\kappa(H)$ in both H directions is nearly identical below H_s. Above H_s, $\kappa(H)$ grows gradually with H with a convex curvature, followed by a steep increase with strong upward curvature near H_{c2}. These results indicate multigap superconductivity with active two-dimensional (2D) and passive three-dimensional bands having contrasting gap values. Together with the recent penetration depth results, we suggest that the 2D bands consist of nodal and nodeless ones, consistent with extended s-wave symmetry.

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Recent discovery of a new class of Fe-based superconductors[1] has attracted much attention. Among them, FeAs-based compounds have aroused great interest because of the high transition temperature T_c. Undoped arsenide LaFeAsO is nonsuperconducting and has a spin-density-wave (SDW) ground state but becomes superconducting (T_c=25 K) when electron doped.[2] By changing the rare-earth ion, T_c reaches as high as 55 K in SmFeAs (O,F).[3] A key question is the origin of the pairing interaction. Since the symmetry of the superconducting order parameter is intimately related to the pairing interaction at the microscopic level, its identification is of primary importance.

Fully gapped superconducting states in FeAs-based superconductors have been reported by the penetration depth measurements of $PrFeAsO_{1-y}$,[4] $SmFeAsO_{1-x}F_y$,[5] and $Ba_{1-x}K_xFe_2As_2$,[6] angle-resolved photoemission,[7] thermal conductivity,[8] and NMR (Ref. 9) measurements of $Ba_{1-x}K_xFe_2As_2$. Some of them give evidence of multiband superconductivity with two distinct gaps. On the other hand, the NMR of $LaFeAsO_{1-x}F_x$ (Ref. 10) and $PrFeAsO_{0.89}F_{0.11}$ (Ref. 11) and the penetration depth measurements of $Ba(Fe_{1-x}Co_x)_2As_2$ (Ref. 12) suggest the presence of low-lying excitations, which could be indicative of nodes. Theoretically, it is proposed that a good nesting between hole and electron pockets prefers the "$s_{\pm}$" symmetry where the gap is finite at all Fermi surfaces but changes its sign on different bands.[13–15] Recent neutron resonant scattering[16] and the impurity effects on the penetration depth[6,17] are consistent with this symmetry.

The phosphide LaFePO (Ref. 18) has quite different magnetic and superconducting properties from LaFeAsO, e.g., LaFePO is nonmagnetic in the normal state,[19] while they are isomorphic and share a similar electronic structure.[20,21] Recently, a superconducting state of LaFePO has been suggested to possess line nodes in the gap function by a linear temperature dependence of the penetration depth at low temperatures.[22,23] However, on which Fermi surfaces the nodes locate in the multiband electronic structure is not yet clarified, and while several candidates have been theoretically proposed,[24,25] the superconducting symmetry in LaFePO remains elusive. Thus the clarification of the detailed gap structure of LaFePO is expected to provide important clues to the origins of magnetism and superconductivity of Fe-based compounds.

Here, to shed further light on the gap symmetry of LaFePO, we present the thermal conductivity measurements at low temperatures. The thermal conductivity probes delocalized low-energy quasiparticle excitations and is an extremely sensitive probe of the anisotropy of the gap amplitude. We provide strong evidence of the multigap superconductivity in a more dramatic fashion than FeAs-based superconductors, with two very different gap values. We show that there are passive three-dimensional (3D) bands and two kinds of active two-dimensional (2D) bands; one is fully gapped and the nodes inferred from the penetration depth measurements[22,23] are most likely on the other 2D bands. This is compatible with the extended s-wave (nodal $s_{\pm}$) symmetry for the gap structure of LaFePO.

Single crystals with dimensions of ~$0.8\times0.4\times0.05$ mm^3 were grown by a Sn-flux method.[26] We carefully removed the Sn flux at the surface of the crystals by rinsing in diluted hydrochloric acid. The resistivity and susceptibility measurements show the sharp superconducting transition at T_c=7.4 K (determined by the midpoint of the resistive transition), which is slightly higher than the values reported by other groups.[22,27] The thermal conductivity κ was measured by a standard four-wire steady method for a heat current $\boldsymbol{q}$ within the ab plane.

The temperature dependence of the in-plane resistivity ρ in the zero field (inset of Fig. 1) depends on T as $\rho=\rho_0+AT^2$, with ρ_0=4.9 $\mu\Omega$ cm and $A=3.3\times10^{-3}$ $\mu\Omega$ cm/K^2 below 50 K down to T_c. The residual resistivity ratio (RRR) is 28. Clear de Haas–van Alphen (dHvA) oscillations were observed in samples from the same batch with nearly the same RRR value.[26] The upper critical fields at $T\rightarrow0$ K estimated by the resistivity measurements are $\mu_0H_{c2}^c$=1.0 T for $\boldsymbol{H}\|c$ axis and $\mu_0H_{c2}^{ab}$=8.6 T for $\boldsymbol{H}\|ab$ plane. This large anisotropy of the upper critical field H_{c2} indicates that the

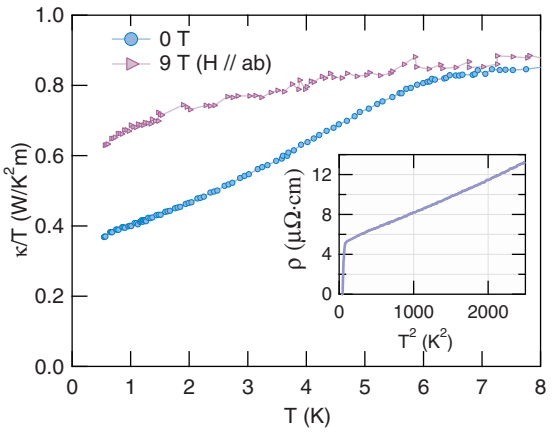

FIG. 1. (Color online) Temperature dependence of κ/T in zero field and at $\mu_0 H=9$ T [above H_{c2}^{ab} ($\boldsymbol{H}\|ab, \boldsymbol{H}\perp\boldsymbol{q}$)]. Inset: ρ plotted as a function of T^2.

bands active for the superconductivity have a very anisotropic 2D electronic structure.

Figure 1 depicts the temperature dependence of κ/T in zero field and in the normal state above H_{c2}^{ab}. As the temperature is lowered, κ/T decreases below T_c. It can be clearly seen that the electron contribution dominates well the phonon heat contribution because the electronic contribution in κ at 1 K estimated by the Wiedemann-Franz law, $\kappa=L_0T/\rho$ ($L_0=2.44\times10^{-8}$ Ω W/K is the Sommerfeld value), is 0.5 W/K^2 m which is close to the observed value 0.65 W/K^2 m. Further, the phonon contribution measured in a related compound $BaFe_2As_2$ (Ref. 28) is one order of magnitude smaller in low temperatures.

First we discuss the thermal conductivity in zero field. A residual term κ_{00}/T at $T\to0$ K in κ/T is clearly resolved. In the nodal superconductors, such a residual term appears as a result of the impurity scattering which induces quasiparticles even at $T=0$ K. In the presence of line nodes in a single band superconductor, κ_{00}/T is roughly estimated as $\sim2(\xi_{ab}/\ell)\cdot(\kappa_n/T)$, where ξ_{ab} is the in-plane coherence length, ℓ is the mean free path and κ_n is the thermal conductivity in the normal state. Using $\ell=94$ nm from the dHvA measurements[26] and $\xi_{ab}=\sqrt{\Phi_0/(2\pi H_{c2}^c)}=18$ nm, κ_{00}/T is estimated to be ~0.19 W/K^2 m. This value is comparable to the observed $\kappa_{00}/T\sim0.30$ W/K^2 m, but we note that this estimate includes large ambiguities due to the multiband effect which could alter effective ξ_{ab} and κ_n. So this comparison alone cannot be taken as the evidence for line nodes in the superconducting gap. It should be also noted that the residual term may arise from an extrinsic origin, such as nonsuperconducting metallic region[27] with high thermal conductivity although the sharp superconducting transition and the observation of the dHvA oscillation indicate a good quality of the crystal.

More vital information on the gap structure can be provided by the field dependence of κ at low temperatures. The field-dependent part of $\kappa(H)$ in a mixed state mainly stems from the superconducting part of the crystals even if a nonsuperconducting region was present in the crystal. Moreover, the phonon scattering at the low temperatures is governed by static defects and is therefore field independent. Further, it is

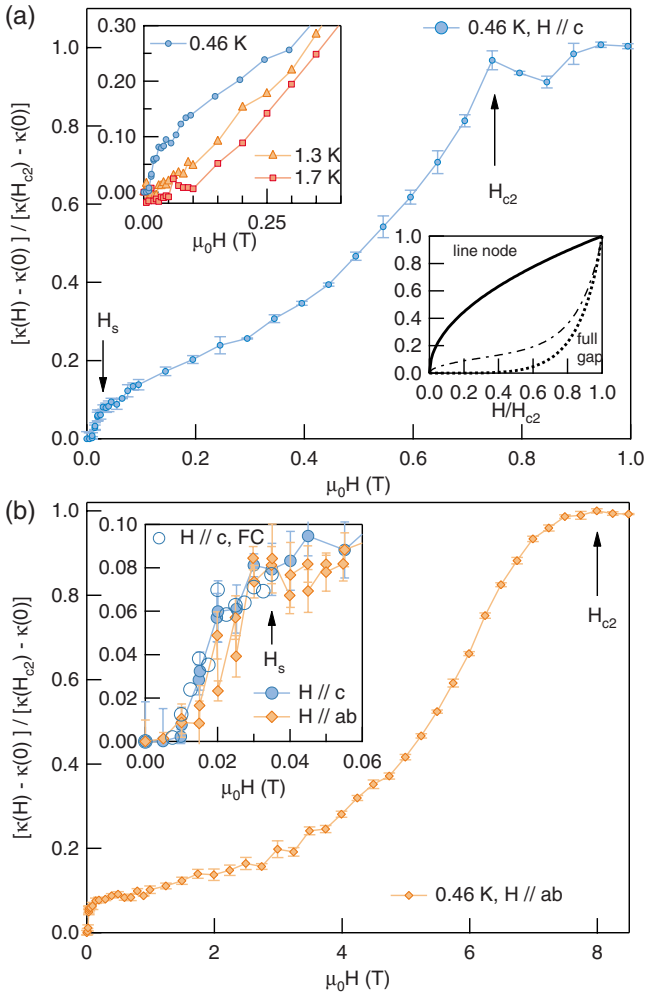

FIG. 2. (Color online) (a) Field dependence of $\kappa(H)-\kappa(0)$ normalized by $\kappa(H_{c2}^c)-\kappa(0)$ for $\boldsymbol{H}\|c$ at $T=0.46$ K. Upper inset: the same plot in low fields for $\boldsymbol{H}\|c$ at 0.46 K (circles), 1.3 K (triangles), and 1.7 K (squares). Lower inset: schematic field dependence of $\kappa(H)-\kappa(0)$ in superconductors with a full gap (dotted line), with line nodes (solid line), and with two kinds of gaps with and without nodes (dash-dotted line). (b) Field dependence of $[\kappa(H)-\kappa(0)]/[\kappa(H_{c2}^{ab})-\kappa(0)]$ for $\boldsymbol{H}\|ab$ plane at 0.46 K. Inset: a comparison of the low-field data for $\boldsymbol{H}\|ab$ with those for $\boldsymbol{H}\|c$ measured in the zero-field (filled circles) and field cooling (FC) conditions (open circles).

well known that fully gapped and nodal superconductors show a contrasting field dependence,[29] as illustrated in the lower inset of Fig. 2(a). In fully gapped superconductors, quasiparticles excited by vortices are localized and unable to transport heat until these vortices are overlapped each other. Consequently, $\kappa(H)$ shows a slow growth with H in low fields and a rapid increase near H_{c2} (the dotted line), as reported in Nb.[30] In sharp contrast, the heat transport in superconductors with nodes or with a large anisotropy in the gap is dominated by contributions from delocalized quasiparticles outside vortex cores.[31] In the presence of line nodes where the density of states has a linear energy dependence, $\kappa(H)$ increases in proportion to $\sqrt{H}$ (Ref. 32) (the solid line). If nodeless and nodal gaps are mixed in a multiband system without interband scatterings, an inflection point emerges in

the intermediate field regime (the dash-dotted line).[33]

Figures 2(a) and 2(b) depict $\kappa(H)-\kappa(0)$ normalized by $\kappa(H_{c2})-\kappa(0)$ for $\boldsymbol{H}\|c$ and $\boldsymbol{H}\|ab$, respectively, measured at $T=0.46$ K $(0.062T_c)$ by sweeping H after zero-field cooling. We note that little difference was observed between the data measured in zero-field and field cooling conditions, indicating that the field trapping effect is very small. For both field directions, the overall H dependence of κ is quite similar. At very low fields, κ exhibits a pronounced increase with increasing H. Remarkably, in spite of the large anisotropy of H_{c2}, $\kappa(H)$ is nearly identical for both H directions at low fields and almost saturates at around $H_s \sim 350$ Oe, as shown in the inset of Fig. 2(b). Above H_s, $\kappa(H)$ becomes anisotropic with respect to the field direction and is governed by the anisotropy of H_{c2}. For both $\boldsymbol{H}\|c$ and $\boldsymbol{H}\|ab$, $\kappa(H)$ grows gradually with H with a convex curvature followed by a rapid increase with a concave curvature up to H_{c2}; there is an inflection point at $\sim H_{c2}/4(\sim H_{c2}/8)$ for $\boldsymbol{H}\|c$ $(\boldsymbol{H}\|ab)$.

The steep increase and subsequent gradual increase in $\kappa(H)$ above H_s for both $\boldsymbol{H}\|c$ and $\boldsymbol{H}\|ab$ indicate that a substantial portion of the quasiparticles is already restored at H_s, much below H_{c2}. Such a two-step field dependence has been reported in MgB_2,[34] $PrOs_4Sb_{12}$,[35] and URu_2Si_2,[36] providing direct evidence for the multiband superconductivity. Here H_s is interpreted as a "virtual upper critical field" that controls the field dependence of the smaller gap of the "passive" band. Its superconductivity is most likely induced by the proximity effect of the "active" bands with primary gap. The ratio of the large and small gaps is roughly estimated to be $\Delta_L/\Delta_S \sim \sqrt{H_{c2}^c/H_s} \sim 6$. If we take $\Delta_L \sim 1.7k_BT_c \sim 13$ K, we obtain $\Delta_S \sim 2$ K.

We note that the steep increase in $\kappa(H)$ below H_s is not due to the influence of the first vortex penetration field, which is expected to be anisotropic and much smaller than $H_{c1} \sim 100$ Oe if the demagnetization is taken into account.[37] We can also rule out a possibility that the steep increase is caused by the remanent Sn flux because $\kappa(T)$, magnetization, and microwave surface impedance measurements[38] show no anomaly at T_c of Sn (=3.72 K). Moreover, the low-field steep increase disappears at $T \sim 1.5$ K well below T_c of Sn [the upper inset of Fig. 2(a)], which is rather in good agreement with the gap size estimation.

The observed multiband superconductivity is compatible with the band structure of LaFePO. The band structure calculations show that Fermi surface consists of two electronic cylinders centered at the M point and two hole cylinders centered at the Γ point, together with a single hole pocket with 3D-like dispersion at the Z point in the Brillouin zone (see the sketch in Fig. 3).[20,21,39] The 3D band is suggested to appear in LaFePO, not in LaFeAsO, and has a character of the $3d_{3z^2-r^2}$ orbital which is expected to have a weak coupling to other 2D bands.[21] Note that only the 2D cylindrical electron and hole bands have been reported by photoemission[40] and dHvA measurements.[26,39]

Nearly isotropic $\kappa(H)$ with respect to the field direction below H_s shown in the inset of Fig. 2(b) indicates that the smaller gap is present most likely in the 3D hole pocket. This is consistent with the expected weak coupling between the 3D and 2D bands. This passive 3D band is inferred to be fully gapped because of the following reasons. The field dependence of κ below H_s does not show strong $\sqrt{H}$ dependence expected for line nodes. Moreover, since the coherence length of the smaller gap, $\xi_s=\sqrt{\phi_0/(2\pi H_s)} \simeq 100$ nm, is comparable to the mean free path, it is unlikely that a nodal superconductivity can survive against such a "dirty" condition $(\xi_s \simeq \ell)$.

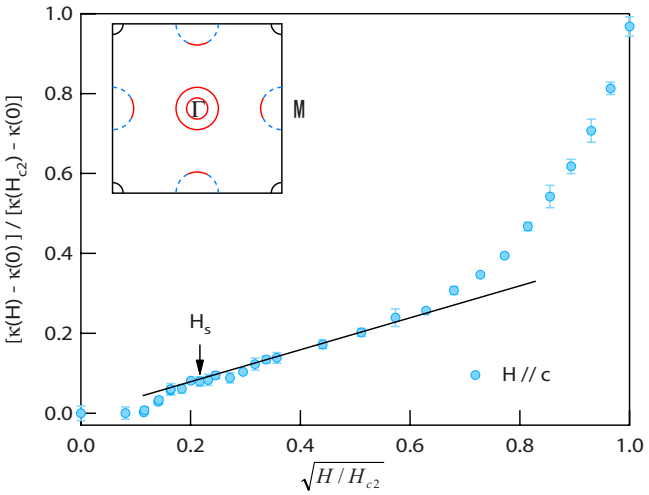

FIG. 3. (Color online) The same data in the main panel of Fig. 2(a) plotted against $\sqrt{H/H_{c2}}$. The solid line is a guide for the eyes. The inset illustrates the extended s-wave (nodal $s_{\pm}$) gap structure (Ref. 25) in the unfolded Brillouin zone. The sign of the gap changes between the solid and dotted lines. The small band at the zone corner represents the 3D band around the Z point.

Next we discuss the gap structure of the 2D bands from $\kappa(H)$ above H_s, where essentially all quasiparticles of the 3D band with smaller gap have already contributed to the heat transport. As shown in Fig. 3, $\kappa(H)$ for $\boldsymbol{H}\|c$ increases as $\sim\sqrt{H}$ just above H_s to $\sim 0.4H_{c2}$ (the $\sqrt{H}$ dependence is not clear below H_s in our resolution). This H dependence and the appearance of the inflection point from convex to concave H dependence are in sharp contrast to the H dependence of the simple fully gapped superconductors, in which H dependence is always concave well below T_c.[3] Such a convex (sublinear) H dependence at low fields appears when the gap is highly anisotropic with a large amplitude modulation. On the other hand, the concave H dependence just below H_{c2} has never been reported in superconductors with large anisotropic gap, such as $Tl_2Ba_2CuO_{6+\delta}$,[32] $CePt_3Si$,[41] and $LuNi_2B_2C$.[42] Therefore, it is likely that at least one of the active 2D bands is fully gapped without nodes. In fact, the H dependence of $\kappa(H)$ with two kinds of gaps with and without nodes[33] shows an inflection behavior [see the dash-dotted line in the lower inset of Fig. 2(a)], which qualitatively reproduces the data. This result, along with the finite κ_{00}/T observed in our dHvA-available clean crystal, supports the nodal superconductivity suggested by the linear temperature dependence of the superfluid density.[22,23] Thus the whole H dependence of κ above H_s implies that the 2D bands consist of two kinds; one has nodes and the other is fully gapped.

We note that this multi-gap feature we deduce is compatible with the penetration depth measurements;[22,23] the nodal gap dominates the linear temperature dependence in low temperatures where the effects of large nodeless gap and the

small gap are negligible because the former is already saturates far below T_c and the latter has a tiny density of states (less than 5% of total density of states).

Finally we discuss the position of the nodes. As candidates of the gap structure with line nodes, the "nodal $s_{\pm}$-wave" and "d-wave" symmetries have been proposed for LaFePO.[25] We infer that the d-wave can be excluded because it possesses line nodes in the 3D band (as well as the 2D hole bands), which is unlikely for the reasons discussed above. The nodal $s_{\pm}$-wave structure has a nodal gap on the 2D electron band around M point and the 2D and 3D hole bands are fully gapped (see the sketch in Fig. 3). The gap size of the 2D electron and hole bands can be comparable to each other.[25] Therefore, we suggest that the nodal $s_{\pm}$-wave structure can be the best candidate for the gap symmetry of LaFePO.

In summary, from the measurements of the thermal conductivity, LaFePO is found to be a multigap superconductor with 2D active and 3D passive bands. The peculiar field dependence of κ provides a stringent constraint on the superconducting gap structure in this system: there exist fully gapped 2D and 3D bands and the nodes locate most likely on the other 2D bands. These results are consistent with the nodal $s_{\pm}$-wave symmetry proposed for the superconducting state of LaFePO.

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