

Three-dimensionality of band structure and a large residual quasiparticle population in $\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2$ as revealed by c -axis polarized optical measurements

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We report on a c -axis polarized optical measurement on a $\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2$ single crystal. We find that the c -axis optical response is significantly different from that of high- T_c cuprates. The experiments reveal an anisotropic three-dimensional (3D) optical response with the absence of the Josephson plasma edge in $R(\omega)$ in the superconducting state. Furthermore, different from the ab -plane optical response, a large residual quasiparticle population down to $T \sim \frac{1}{5}T_c$ was observed in the c -axis polarized reflectance measurement. We elaborate that there exist horizontal nodes for the superconducting gap in regions of the 3D Fermi surface that contribute dominantly to the c -axis optical conductivity.

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I. INTRODUCTION

For quasi-two-dimensional (2D) superconducting materials, a striking difference could exist between the in-plane and out-of-plane (the c -axis) optical responses. In high- T_c cuprates, for example, the conducting CuO_2 planes are usually separated by insulating blocking layers. In the normal state, those CuO_2 planes appear to be almost decoupled, leading to nonmetallic charge transport and dynamics along the c axis. However, as soon as the systems enter into the superconducting state, the CuO_2 planes are coupled via the Josephson tunneling effect, then a sharp reflectance edge arising from the condensed superconducting carriers immediately shows up in the c -axis polarization.¹⁻⁵ The plasma edge is referred to as the Josephson plasma edge, which is linked with the c -axis penetration depth. It offers a direct measure to the strength of the interlayer coupling. This behavior is also connected with the mechanism of high-temperature superconductivity, since the absence of this edge above T_c is considered as indicative of the confinement mechanism.

Fe-pnictide superconducting materials also crystalize in the layered structure with FeAs layers separated by alkaline-earth metal ions (Ba^{2+} , Sr^{2+}) or other Re -O ($Re = \text{La}$, or rare-earth elements with trivalent Re^{3+} and negative divalent O^{2-}) layers which should be insulatorlike. It is important to see whether or not the Fe-pnictides share similar anisotropic charge dynamical properties with cuprates. Although there are a number of optical studies on the ab -plane properties of Fe-based superconducting materials, little is known about the properties along the perpendicular direction due to the lack of thick enough single-crystal samples. The c -axis polarized optical data are available only for parent compounds where the spin-density-wave gap structure was found to be substantially different from that of $\mathbf{E} \parallel ab$ plane.⁶

In this work we present the c -axis optical spectroscopy measurements on superconducting $(\text{Ba,K})\text{Fe}_2\text{As}_2$ crystals. We show that, in contrast to the high- T_c cuprates, the c -axis Josephson plasmon is completely absent in the superconducting state. Furthermore, different from the ab -plane optical

response where the full superconducting energy gaps could be seen clearly,⁷ the c -axis polarized measurement reveals a small difference between $T > T_c$ and $T < T_c$. The low-frequency optical conductivity below T_c still exhibits a Drude response. The experiment reveals a large residual quasiparticle population down to the lowest measurement temperature $T \sim \frac{1}{5}T_c$. Since the c -axis polarized measurement mainly probes the three-dimensional (3D) Fermi surface (FS) in the band structure, the data strongly suggest the presence of nodes for the superconducting gap in regions of the 3D FS that contribute dominantly to the c -axis optical conductivity.

II. EXPERIMENT

Thick $\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2$ single crystals were grown from the FeAs self-flux in Al_2O_3 crucibles sealed in Ta tubes filled with argon. The same batch of crystals was also used for a neutron scattering experiment, which shows an absence of the static antiferromagnetic order coexisting with superconductivity down to 2 K.⁸ The shiny cleaved ab plane could be easily obtained after cutting the Ta tube and breaking the crucible. The layered stacking could actually be seen along the edge for thick single crystals, making it rather easy to identify the c axis. Then the crystals were cut in a direction perpendicular to the cleaved ab plane. The cutting surfaces with dimensions up to $5 \text{ mm} \times 2 \text{ mm}$ were finely polished for the c -axis polarized measurement. The polishing was carried out initially on sandpapers with different surface roughnesses. The final step was done on a very fine polishing pad with a spray of a $0.5\text{-}\mu\text{m}$ alumina polishing lubricant from the Shenyang Kejing Corporation. The polished surface is mirrorlike and very shiny. To prevent possible surface degradation, the polishing process was done in a glove box filled with Ar.

The optical reflectance measurements with an $\mathbf{E} \parallel c$ axis were performed on Bruker IFS 80v and 113v spectrometers in the frequency range from 30 to $25\,000 \text{ cm}^{-1}$. An *in situ* gold and aluminum overcoating technique was used to obtain

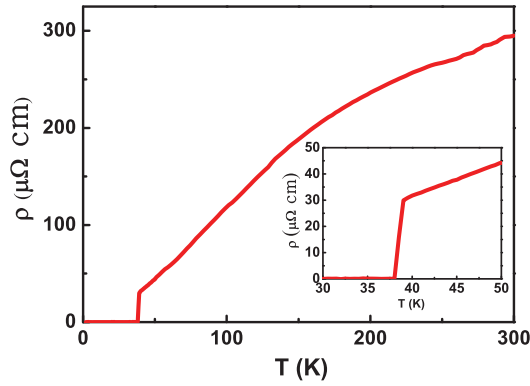


FIG. 1. (Color online) Temperature-dependent resistivity $\rho(T)$ for the $\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2$ single crystal. The superconducting transition temperature is 38 K.

the reflectivity $R(\omega)$. The real part of conductivity $\sigma_1(\omega)$ is obtained by the Kramers-Kronig transformation of $R(\omega)$.

III. RESULTS AND DISCUSSIONS

Figure 1 shows the temperature dependence of the in-plane dc resistivity measured by the four-contact technique for a $\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2$ crystal. The measurement indicates a superconducting transition temperature $T_c = 38$ K. The resistivity data are very similar to that reported for a $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ crystal where the in-plane optical data were presented.⁷

Figure 2 shows the c -axis $R(\omega)$ and $\sigma_1(\omega)$ at room T up to $15\,000\text{ cm}^{-1}$. For a comparison, we also plot the optical spectra with an $E \parallel ab$ plane for the same crystal. The in-plane optical data are rather close to those of our earlier measurement on a $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ crystal.⁷ Similar to the undoped compound, we find that the overall $R(\omega)$ along the c axis is quite similar to that in the ab plane except for relatively lower values. This is dramatically different from the optical spectra of high- T_c cuprates³ and other layered compounds, for example, layered ruthenates,⁹ where the c -axis $R(\omega)$ shows much lower values and quite different frequency-dependent behavior from the ab plane. This observation suggests that the band structure of Fe-pnictide should be quite 3D, in contrast to the expectation

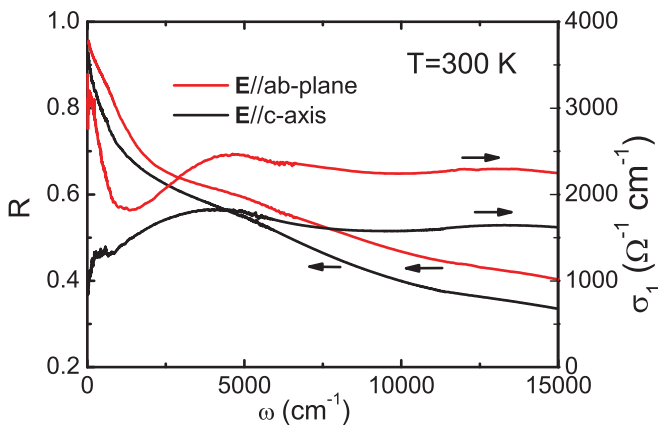


FIG. 2. (Color online) The c -axis $R(\omega)$ and $\sigma_1(\omega)$ at 300 K for the $\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2$ over a broad frequency up to $15\,000\text{ cm}^{-1}$. The ab -plane spectra were also included for comparison.

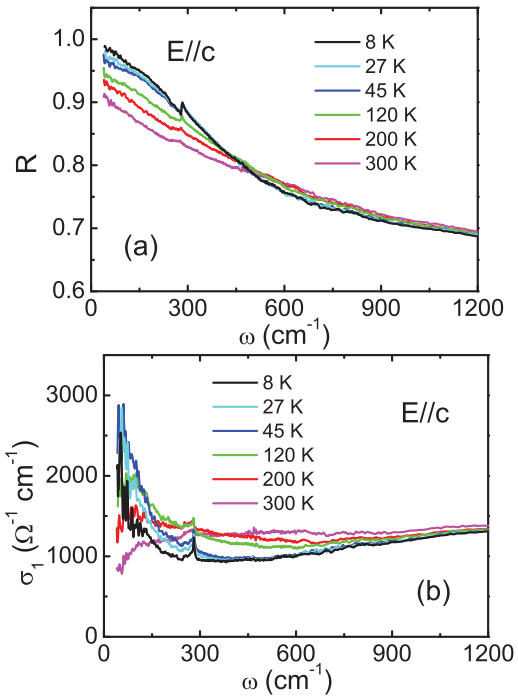


FIG. 3. (Color online) (a) Optical reflectance $R(\omega)$ spectra at different temperatures below 1200 cm^{-1} . (b) The conductivity $\sigma_1(\omega)$ spectra of the sample at different temperatures.

based on its layered crystal structure. An anisotropy ratio of optical conductivities at the low-frequency limit is less than 4, which is close to the value of the undoped compound,⁶ suggesting that the anisotropy does not show a significant change upon K doping.

We are mainly interested in the evolution of optical spectra at low frequencies across the superconducting transition. Figure 3 shows the temperature dependence of the low- ω reflectance and conductivity spectra. The reflectance $R(\omega)$ spectra below 1200 cm^{-1} are displayed in Fig. 3(a). The spectra indicate a typical metallic temperature dependence: The low- $\omega R(\omega)$ increases with decreasing temperature. Most noticeably, no sharp plasma edge develops below T_c . This is in sharp contrast to the high- T_c cuprates where a steep reflectance edge is seen in the superconducting state, which was ascribed to the Josephson coupling of superconducting CuO_2 planes.¹⁻⁵ The result unambiguously illustrates that the 122-type Fe-pnictides are significantly different from the cuprates: The condensed superconducting carriers are not coupled though the Josephson tunneling effect. Instead, the metallic optical response indicates that the compound behaves in a way similar to an anisotropic 3D system where dispersive bands along the c axis would exist.

The low-frequency conductivity $\sigma_1(\omega)$ spectra are shown in Fig. 3(b). Although the sample shows a metallic temperature dependence of the low-frequency conductivity, the frequency dependence at high temperature ($T > 45\text{ K}$) is not simply Drude-like. Instead, a broad peak at finite frequency exists in the $\sigma_1(\omega)$ spectra. The peak, which is located near 300 cm^{-1} at 300 K, shifts to lower frequency and gradually disappears with decreasing temperature. A Drude component centered at zero frequency is seen at a low temperature. It is worth noting

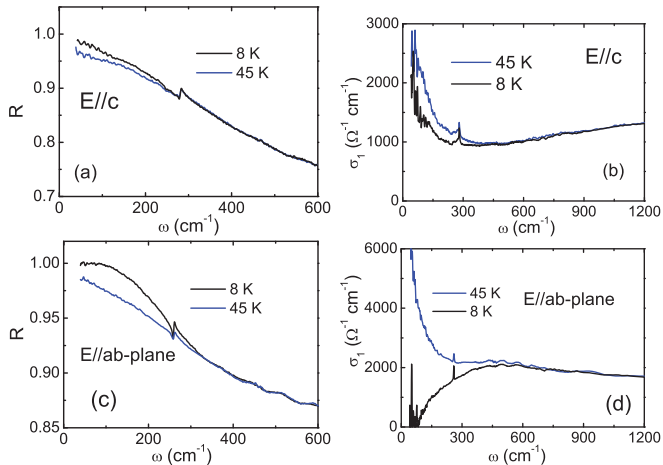


FIG. 4. (Color online) (a) The reflectance $R(\omega)$ spectra at 8 and 45 K below 600 cm^{-1} . (b) The low-frequency conductivity spectra at 8 and 45 K. For comparison, the in-plane $R(\omega)$ and $\sigma_1(\omega)$ spectra at 8 and 45 K on a cleaved ab -plane surface are shown in (c) and (d), respectively. A sharp c -axis A_{2u} mode related to the Fe-Ba atomic displacements (Ref. 14) is seen at 280 cm^{-1} , which is $\sim 30 \text{ cm}^{-1}$ higher than the ab -plane Eu mode related to the Fe-As vibrations. The in-plane reflectance and conductivity spectra show an s -wave superconducting energy gap. By contrast, a significant residual Drude response is still observed for the $\mathbf{E} \parallel c$ axis.

that the presence of the conductivity peak at finite frequency does not necessarily mean the absence of the dispersive band. Nevertheless, it is often seen in poor metallic materials at high temperatures where the quasiparticle peaks along the dispersive band are rather broad.^{10–13} It is a kind of dynamical localization effect and its origin remains to be explored.

Figures 4(a) and 4(b) show $R(\omega)$ and $\sigma_1(\omega)$ at 45 and 8 K in the expanded region. A sharp c -axis A_{2u} mode related to the Fe-Ba atomic displacements¹⁴ is seen at 280 cm^{-1} , which is $\sim 30 \text{ cm}^{-1}$ higher than the ab -plane Eu mode (250 cm^{-1}) related to the Fe-As vibrations. The clear difference in the phonon frequencies indicates that we are measuring the c -axis responses from the polished surface. The $R(\omega)$ spectrum at 8 K deviates upward from that at 45 K below $\sim 250 \text{ cm}^{-1}$, which could be attributed to the superconducting condensate. However, the difference between the two spectra is rather small. This is significantly different from the in-plane optical data at the two different temperatures measured on the cleaved surface of the same sample [Figs. 4(c) and 4(d)]. The spectrum at 8 K shows a clear upward deviation from the spectrum at 45 K below $\sim 300 \text{ cm}^{-1}$, and approaches unity at $\sim 100 \text{ cm}^{-1}$, a behavior already analyzed in detail for a $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ crystal.⁷ The upward deviation is a characteristic feature of the energy gap opening in the superconducting state. The feature has also been observed in the in-plane spectra of a number of other Fe-based materials.^{15–20} The small difference of the spectra between 45 and 8 K for $\mathbf{E} \parallel c$ indicates the presence of a significant amount of quasiparticles which have a dominant contribution to the c -axis conductivity.

Similar to the reflectance spectra, we can see from Fig. 4(b) that there exists only a small difference for the conductivity spectra at 8 and 45 K for $\mathbf{E} \parallel c$, while a dramatic difference

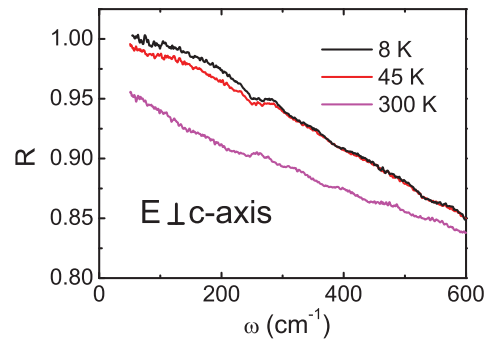


FIG. 5. (Color online) The low- ω reflectance spectra with the $\mathbf{E} \perp c$ axis measured on the cleaved surface.

is seen for $\mathbf{E} \parallel ab$ plane [Fig. 4(d)], which is attributed to the opening of a superconducting energy gap. The data show that an s -wave superconducting gap structure could be clearly observed only from the in-plane optical measurement. The low-frequency optical conductivity still exhibits a Drude response for the $\mathbf{E} \parallel c$ axis. The measurement reveals a large residual quasiparticle population down to the lowest measurement temperature at 8 K ($T \sim \frac{1}{5}T_c$).

There is concern about whether or not the polishing would damage the surface and destroy the superconductivity of the optically probed surface layer, so that the rather small difference could be an artifact of this kind of problem. To address the issue, we also measured the $R(\omega)$ spectra on the polished surface with an $\mathbf{E} \perp c$ -axis configuration, which should also reflect the in-plane response. Figure 5 shows the $R(\omega)$ spectra at 8 and 45 K. Regardless of the fact that the spectra have higher values than $R(\omega)$ for the $\mathbf{E} \parallel c$ axis, and the differences between 45 and 8 K are apparently larger than the c -axis data, the spectra for the $\mathbf{E} \perp c$ axis are not identical to the in-plane spectra measured on the cleaved surface as shown in Fig. 4(c). We remark that, although the polishing would inevitably cause some deterioration of the surface, it is not the major reason for the difference. We verified by performing the point-contact tunneling spectroscopy measurement that the polished surface on which we carried out a polarized optical measurement is still superconducting.²¹ On the other hand, the leakage of the polarizer and a small misalignment for such thick single crystals exist, which could lead to a small degree of mixing between the in-plane and the c -axis spectra. Indeed, the mixing could be easily identified from the presence of weak phonon features in respective frequencies. But this kind of mixing would reduce the difference between the two polarized configurations. This is essentially what we observed in experiment. Because the ab plane has a higher reflectance than the c axis, then the leakage and misalignment would result in an increase of the reflectance for the $\mathbf{E} \parallel c$ axis below the superconducting energy gap and a reduction of the reflectance for the $\mathbf{E} \perp c$ axis. Those could lead to overestimation and underestimation of the superconducting condensate for the $\mathbf{E} \parallel c$ and $\mathbf{E} \perp c$ axes, respectively. But even in this case we still found the presence of a large concentration of uncondensed carrier density for the $\mathbf{E} \parallel c$ axis. So, the conclusion of the presence of a large fraction of residual quasiparticles which contribute specifically to the c -axis transport was not affected by the small mixing.

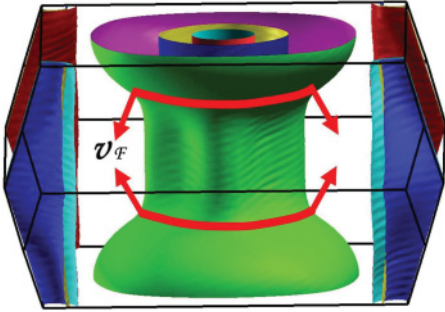


FIG. 6. (Color online) The Fermi surfaces of $(\text{Ba,K})\text{Fe}_2\text{As}_2$ from LDA+Gutzwiller calculations (Ref. 22). Besides the 2D cylinderlike FSs, there is an additional 3D FS. Possible horizontal nodes may exist near the highly flared region of the 3D FS, which has maximum contribution to the c -axis conductivity.

The absence of a Josephson plasma edge could be taken as an indication that the 122 iron-pnictides are quasi-3D systems with the presence of a dispersive band along the c axis. The presence of 3D FSs was also suggested from band-structure calculations²² and a number of other experimental probes.^{23–27} In particular, recent *ab initio* local-density approximation(LDA)+Gutzwiller calculations, where electron correlations are taken into account beyond LDA, indicated that the large-size 3D ellipsoidlike FS has a dominant $\text{Fe-}3d_{3z^2-r^2}$ orbital characteristic.²² Then the key issue is to understand why an s -wave superconducting energy gap is clearly seen from the in-plane optical measurement, whereas a large residual quasiparticle population is indicated in the c -axis polarized optical measurement. It has an important implication for unraveling the superconducting pairing in iron-pnictide systems.

In our previous study on undoped parent compounds, we found that the spin-density-wave gap structure in the c -axis optical spectrum is significantly different from that in the ab plane. The difference was explained naturally by assuming the existence of two types of FSs in the system: 2D cylinderlike FSs and a large-size 3D ellipsoidlike FS.⁶ Here the data could be again well understood from the same electronic structures, as schematically shown in Fig. 5.²² Because the Fermi velocities of the 2D cylinderlike FSs are essentially within the ab plane, the electrons on those FSs would contribute dominantly to the ab -plane conductivity, while the c -axis conductivity is mainly contributed by the 3D ellipsoidlike FS. On this basis, the superconducting energy gaps opened on the 2D cylinderlike FSs below T_c should be clearly seen by the ab -plane polarized measurement, but not by the c -axis polarization. Then the c -axis data could be easily understood if we assume that the superconducting gap formed on the 3D FS is strongly k -space dependent, in particular, if the horizontal node exists in the 3D FS in the region where the electrons contribute dominantly to the c -axis optical conductivity.

It is noted that recent functional renormalization group calculations on LaFePO superconductors²⁸ indicate that the 3D holelike FS centered around (π, π, π) with a dominant $\text{Fe-}3d_{3z^2-r^2}$ orbital characteristic is gapless, although in reality this FS may be absent for LaFePO . Since the 3D FS in K-doped 122 has the same $\text{Fe-}3d_{3z^2-r^2}$ characteristic²² and, furthermore,

the FS is not nested with any other FSs, it is possible that this FS has weak superconducting strength being induced by the proximity effect or by the weak mixing of different orbitals.

Our experimental results are consistent with recent c -axis penetration depth and heat transport measurements,^{25,26,29,30} which also suggested that the nodes contribute specifically to the c -axis transport. A recent neutron scattering experiment on the same batch of $\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2$ crystals revealed that the low-energy spin excitations below T_c display a sinusoidal modulation, indicating a clear gap at momentum transfer $\mathbf{q}_z = 0$ but gapless behavior at $\mathbf{q}_z = \pi$.⁸ The presence of nodes in different regions of FSs was also suggested in theoretical studies, including nodes on 2D electron-type FSs near the zone corner,³¹ vertical and small segment nodal lines on the 3D hole FS, and horizontal nodes on the 3D FS.³² As we elaborated above, the c -axis optical measurement mainly probes the 3D FS, while the nodes on the 2D electron FSs would have a strong effect on the in-plane conductivity. Since the in-plane optical data did not reveal substantial residual conductivity in the superconducting state,⁷ this possibility should be ruled out. Our data suggest that the horizontal node is most likely to be present, since the vertical node on the 3D FS should also contribute visibly to the ab -plane conductivity. If the horizontal nodes exist in the region that contribute dominantly to the c -axis optical conductivity, for example, in the highly flared region of the 3D FS as shown in Fig. 4, and meanwhile the gap amplitude changes in the 3D FS, i.e., being small near the node and relatively large in the region of $k_z \sim \pm\pi$ where the Fermi velocity has a vanishing c -axis component, then the subtle change in the optical reflectance spectrum across the T_c is expected.

IV. CONCLUSIONS

To conclude, our c -axis polarized optical measurement on $\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2$ single crystals revealed that (1) in contrast to the high- T_c cuprates, no Josephson plasma edge in $R(\omega)$ develops below T_c ; (2) different from the ab -plane optical response where an s -wave superconducting gap is clearly observed, the c -axis data only exhibit a small difference across T_c , with the indication of a large residual quasiparticle population. Our study indicates that the 122 iron-pnictides are quasi-3D systems with the presence of a dispersive band along the c axis. Furthermore, there may exist a node in the superconducting gap in the 3D FS in the region which dominates the c -axis optical conductivity. We suggest that a horizontal node at the highly flared region of the 3D FS is more consistent with our experimental observation.

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