Probing superconducting energy gap from infrared spectroscopy on a Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ single crystal with $T_c$=37 K

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We performed optical spectroscopy measurement on a superconducting Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ single crystal with $T_c$=37 K. Formation of the superconducting energy gaps in the far-infrared reflectance spectra below $T_c$ is clearly observed. The gap amplitudes match well with the two distinct superconducting gaps observed in angle-resolved photoemission spectroscopy experiments on different Fermi surfaces. We determined absolute value of the penetration depth at 10 K as $\lambda \simeq 2000\AA$. A spectral weight analysis shows that the Ferrell-Glover-Tinkham sum rule is satisfied at low energy scale, less than 6$\Delta$.

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The energy gap created by the pairing of electrons is the most important parameter of a superconductor. Probing the pairing energy gap is crucial for elucidating the mechanism of superconductivity. For conventional superconductors, infrared spectroscopy is a standard technique to probe the superconducting energy gap, as the electromagnetic radiation below the gap energy $2\Delta$ could not be absorbed. However, detecting superconducting energy gap by infrared spectroscopy is not always straightforward. For example, in the case of high-$T_c$ cuprates it has been a long standing controversial issue whether the superconducting gap could be detected from ab-plane infrared spectra, as it was argued that the ab-plane of the cuprates is in the clean limit, and as a consequence the pairing gap could not be seen.

The recent discovery of superconductivity in FeAs-based RFeAsO$_{1-x}$F$_x$ (R=rare earth elements like La, Ce, Pr, Nd, Sm and etc.) and (A,K)Fe$_2$As$_2$ (A=Ba, Sr) has generated new excitement in superconductivity community because they represent a new class of high temperature superconductors. It raises the question whether the pairing mechanism in the new systems is conventional, or related to that in cuprates. With the success of the growth of single crystals in the FeAs-based superconductors, it is important to investigate the fundamental properties of the new systems. Such studies are expected to shed new light on the high temperature superconductivity in cuprates.

In this letter we present an infrared study on a superconducting Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ single crystal with $T_c$=37 K. We provide clear evidence that the superconducting gap is present in the optical reflectance spectra with a s-wave-like pairing lineshape. From the onset absorption in optical conductivity, the gap is found to be close to $2\Delta=150$ cm$^{-1}$, however, from the peak position in the ratio of the $R_c(10 \text{ K})/R_n(45 \text{ K})$ (where the subscript $s$ stands for superconducting state, $n$ for normal state) which reflects a more steep drop above this frequency in optical reflectance relative to the normal state, a different gap amplitude of 200 cm$^{-1}$ is seen. Those two different values match well with the two distinct superconducting gaps observed in angle-resolved photoemission spectroscopy (AREPS) experiments on different Fermi surfaces. The ability to observe clearly pairing gaps in infrared spectra indicates that the material is in the dirty limit. The penetration depth for $T\ll T_c$ is estimated to be $\lambda \simeq 2000\AA$.

High-quality single crystals of Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ were grown by a FeAs flux method. Figure 1 shows the temperature dependence of the dc resistivity and ac susceptibility. A sharp superconducting transition is seen at $T_c=37$ K. The optical measurements were performed on a combination of Bruker IFS 66v/s and 113v spectrometers on newly cleaved surfaces. An in-situ gold and aluminium overcoating technique was used for the experiment. Optical conductivity was derived from Kramers-Kronig transformation of the reflectance.

Figure 2 shows the reflectance spectra $R(\omega)$ for the crystal at different temperatures. The inset shows the spectra over broad energy range up to 25000 cm$^{-1}$, while

![Graph](image-url)
FIG. 2: (Color online) T-dependent $R(\omega)$ curves in the far-infrared region. The inset shows $R(\omega)$ over broad frequencies up to 25000 cm$^{-1}$.

the main panel is the expanded plot below 800 cm$^{-1}$. $R(\omega)$ exhibits a metallic response in both frequency and temperature. At 10, 27 and 45 K, the reflectance curves almost overlap with each other above 300 cm$^{-1}$. However, a sudden upturn $R(\omega)$ develops below $T_c$ at low frequencies. This is a strong indication for the formation of an superconducting energy gap due to the pairing of electrons.

It is well known that for a s-wave BCS superconductor with an isotropic superconducting energy gap, the reflectivity approaches unity below $2\Delta$. The $R(\omega)$ curve at 10 K is almost flat below 150 cm$^{-1}$ (18 meV), thus being very similar to the lineshape for a s-wave superconductor. However, above this frequency $R(\omega)$ develops a downward curvature, and its magnitude becomes slightly lower than unity, suggesting that week absorption already exists. The decrease of $R(\omega)$ becomes steep above 200 cm$^{-1}$.

Figure 3 shows the optical conductivity $\sigma_1(\omega)$ derived from the Kramers-Kronig transformation of reflectance spectra. The conductivity values at very low frequencies continue to increase with decreasing temperature in the normal state, being consistent with dc resistivity measurement. Below $T_c$, the curves at 27 K and 10 K decreases steeply near 300 cm$^{-1}$. The conductivity is almost zero below roughly 150 cm$^{-1}$, yielding optical evidence for a s-wave like superconducting energy gap. The onset of the absorption marks the superconducting energy $2\Delta \approx 150$ cm$^{-1}$. In recent ARPES experiments on the same batch of single crystals, two distinct superconducting gaps were observed: a large gap ($\Delta \approx 12$ meV) on the two small hole-like (centered at $\Gamma$) and electron-like (centered at $M$) Fermi surface (FS) sheets, and a small gap ($\Delta \approx 6-8$ meV) on the large hole-like FS (centered at $\Gamma$). Both gaps, closing simultaneously at the bulk $T_c$, are nodeless and nearly isotropic around their respective FS sheets. In comparison with those work, the conductivity onset should correspond to the small gap observed in ARPES, since optical absorption should exist when the radiation energy is higher than this small superconducting pairing gap. Within experimental uncertainties, the measurement results from the two different techniques match quite well.

Figure 4 shows the ratio of the $R_s(10 \text{ K})/R_n(45 \text{ K})$. The total variation exceeds 2%. A peak can be clearly seen at 200 cm$^{-1}$ (25 meV). Within BCS framework in the dirty limit, the peak frequency roughly corresponds to the superconducting energy gap $2\Delta$. We noticed that this gap value is different from the absorption onset in the optical conductivity, which gives smaller value of 150 cm$^{-1}$. In fact, the frequency at 200 cm$^{-1}$ below $T_c$ represents a more steep drop beyond this energy in optical reflectance relative to the normal state. It is most likely this peak frequency is related to the large superconducting gap observed on the two small hole-like and electron-like FS in ARPES experiment.

Note that, the $2\Delta \approx 200$ cm$^{-1}$ (25 meV) seen in optics also matches excellently with the gap $\Delta \approx 12$ meV measured relative to Fermi level in ARPES.

Well below $T_c$, there is a substantial suppression in the low-frequency conductivity due to the formation of superconducting energy. According to the Ferrell-Glover-Tinkham (FGT) sum rule, the difference between the conductivity at $T\approx T_c$ and $T\ll T_c$ (the so-called missing area, see the inset of Fig. 1) is related to the formation of a superconducting condensate,

$$\omega_{ps}^2 = 8 \int_{0^+}^{\omega_c} [\sigma_1(\omega, T \approx T_c) - \sigma_1(\omega, T \ll T_c)]. \quad (1)$$

where $\omega_{ps}^2 = 4\pi n_s e^2/m^*$ is the square of the supercon-
ductivity at $T_c$.

The determination of $\lambda$ from this equation at low frequency limit relies only on the imaginary part of conductivity at $T < T_c$. When the FGT sum rule is fulfilled, the $\lambda$ value determined from the missing area in the real part of conductivity should equal to the value obtained by Eq. (2) in the low-frequency limit.[20] Figure 5 shows the $\lambda(\omega)$ obtained from above formula, at low frequency limit, $\lambda \approx 1950 \text{ Å}$. We find that the value of $\lambda$ obtained directly from Eq. (1) is close to that obtained from the imaginary part of conductivity through Eq. (2). The good agreement (within an accuracy of 5-8%) between the values obtained from the two different approaches suggests that the FGT sum rule is satisfied.

It would be interesting to compare the optical response of FeAs-based superconductor with those found for the high-$T_c$ cuprate superconductors. Several eminent differences exist: first, the observation of clear superconducting gap in the far-infrared spectra suggests that the in-plane superconductivity is in the dirty limit, i.e. the carrier scattering rate $1/\tau \geq 2\Delta$. In this case, the spectral weight of itinerant carriers in optical conductivity distributes in a broad frequency region, however, a large part of the condensate has been captured by the energy of $2\Delta$, as can be seen in the inset of Fig. 1. While in the clean limit case, nearly all spectral weight associated with the condensate lies below $2\Delta$, so that no discernable change appears at $2\Delta$ across the superconducting transition. It is still a controversial issue whether the high-$T_c$ cuprates is in the dirty or clean limit and whether the superconducting gap is visible in infrared reflectance spectra. It is expected that studies on the new FeAs-based superconductors may shed light on the pairing gap feature in the infrared spectra in cuprates.

Secondly, our analysis based on a comparison of the penetration depth values determined from the missing area in the real part of conductivity and imaginary part of conductivity at low frequency limit indicates that the FGT sum rule is satisfied. An inspection of the inset of Fig. 3 reveals that the missing area extends to the frequency roughly below 600 cm$^{-1}$, about 3 times larger than the higher superconducting energy gap $2\Delta$. This indicates that the superconducting condensate forms rapidly or the FGT sum rule is rapidly recovered. This is very different from underdoped high-$T_c$ cuprates where recovery of the FGT sum rule goes to very high energy, or the FGT sum rule is even violated.[21]

Thirdly, the determination of the penetration depth or equivalently the condensed carrier density enables us to check whether the well-known scaling behaviors between the condensed carrier density and $T_c$ still work for the present system. One of such scaling behaviors is called Uemura relation,[22] which states that the superfluid density scales linearly with the transition temperature, $\rho_s = \omega_{ps}^2 = c^2/\lambda^2 \propto T_c$. Uemura relation works well for the hole-doped cuprates in the underdoped region. The relatively low value of $T_c$ and low penetration depth in the present system may fall off the Uemura plot. Chairs proposed another scaling relation, $\rho_s \approx 65\sigma_{dc} T_c$ for BCS weak coupling case, where $\sigma_{dc}$ is the value just above

\[ \lambda(\omega) = c/\omega_{ps} = c/\sqrt{4\pi\omega\sigma_2(\omega)}. \]
measurement on a superconducting Ba$_2$K$_3$Fe$_2$As$_2$ single crystal with $T_c$=37 K. We observe clearly that the superconducting gap is present in the optical reflectance spectra with a $s$-wave pairing lineshape. The onset absorption in optical conductivity appears close to 150 cm$^{-1}$, however, a more steep reflectance decrease relative to the normal state appears at frequency near 200 cm$^{-1}$, leading to a peak position in the ratio of the $R(10 K)/R_n$ (45 K). Those two values match well with the two distinct superconducting gaps observed in AREPS experiments on different Fermi surfaces. The ability to observe clearly pairing gaps in infrared spectra indicates that the material is in the dirty limit. The penetration depth for $T_c$ is estimated to be $\lambda \simeq 2000\AA$.

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