Probing the Superconducting Energy Gap from Infrared Spectroscopy on a $Ba_{0.6}K_{0.4}Fe_2As_2$ Single Crystal with $T_c = 37$ K

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We performed optical spectroscopy measurement on a superconducting $Ba_{0.6}K_{0.4}Fe_2As_2$ 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. A flat and close to unity reflectance is observed roughly below 150 cm^{-1} for $T \ll T_c$, following an s-wave pairing line shape. A more rapid decrease occurs near 200 cm^{-1}, leading to a peak in the ratio of the reflectance at $T \ll T_c$ over that for $T \geq T_c$. We determined the absolute value of the penetration depth at 10 K as $\lambda \simeq 2000 \pm 80$ Å. A spectral weight analysis shows that the Ferrell-Glover-Tinkham sum rule is satisfied at low energy scale, less than 6Δ.

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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 2Δ could not be absorbed [1]. However, detecting the superconducing 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 [2].

The recent discovery of superconductivity in FeAs-based RFeAsO$_{1-x}$F$_x$ (R denotes rare-earth elements such as La, Ce, Pr, Nd, Sm, etc.) [3–6] and $(A, K)Fe_2As_2$ (A = Ba, Sr) [7–9] has generated new excitement in the superconductivity community because they represent a new class of high-temperature superconductors. It raises the question whether the paring 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 [10,11], 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 $Ba_{0.6}K_{0.4}Fe_2As_2$ single crystal with $T_c =$ 37 K. We provide clear evidence that the superconducting gap is present in the optical reflectance spectra $R(\omega)$ with an s-wave-like pairing line shape. The onset absorption occurs near 150 cm^{-1} in $R(\omega)$ and optical conductivity, however, from the peak position in the ratio of the $R_s(10\text{ K})/R_n(45\text{ K})$ (where the subscript s stands for the 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 seem to match with the two distinct superconducting gaps observed in angle-resolved photoemission spectroscopy (ARPES) experiments on different Fermi surfaces [12,13]. The ability to observe a clearly pairing gap in infrared spectra indicates that the material is in the dirty limit. The penetration depth for $T \ll T_c$ is estimated to be $\lambda \simeq 2000 \pm$ 80 Å.

High-quality single crystals of $Ba_{0.6}K_{0.4}Fe_2As_2$ were grown by a FeAs flux method [11]. 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 aluminum overcoating technique was used for the experiment, which enables us to get enough signal at the far-infrared region

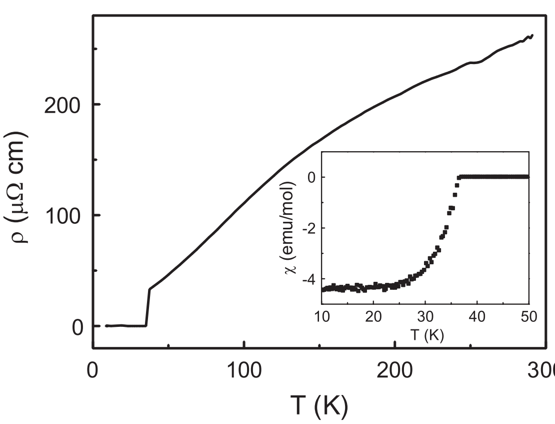

FIG. 1. The dc resistivity as a function of temperature for a $Ba_{0.6}K_{0.4}Fe_2As_2$ single crystal. Inset shows the ac susceptibility of the sample. Sharp superconducting transition occurs at $T_c =$ 37 K.

[14]. Optical conductivity was derived from Kramers-Kronig transformation of the reflectance.

Figure 2 shows the reflectance spectra for the crystal at different temperatures. The inset shows the spectra over a broad energy range up to 25 000 cm^{-1}, while 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 a superconducting energy gap due to the pairing of electrons.

It is well known that for an *s*-wave BCS superconductor with an isotropic superconducting energy gap, the reflectivity approaches unity below 2Δ [15,16]. The $R(\omega)$ curve at 10 K is almost flat below 150 cm^{-1} (18 meV), thus being very similar to the line shape for an *s*-wave superconductor. However, above this frequency $R(\omega)$ develops a downward curvature, and its magnitude becomes slightly lower than unity, suggesting that weak absorption already exists.

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 and 10 K decreases steeply near 300 cm^{-1}. The conductivity is almost zero below roughly 150 cm^{-1} due to the flat and close to unity $R(\omega)$, yielding optical evidence for an *s*-wave-like superconducting energy gap. The onset of the absorption marks the superconducting energy $2\Delta \simeq 150$ cm^{-1}. In recent ARPES experiments on the same batch of single crystals, two distinct superconducting gaps were observed: a large gap ($\Delta \simeq 12$ meV) on the two small holelike (centered at Γ) and electronlike (centered at M) Fermi surface (FS) sheets, and a small gap ($\Delta \simeq 6$–8 meV) on the large holelike FS (centered at Γ). Both gaps, closing simultaneously at the bulk T_c, are nodeless and nearly isotropic around their respective FS sheets [12,13]. In comparison with those works, 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.

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 the BCS framework in the dirty limit, the peak frequency roughly corresponds to the superconducting energy gap 2Δ [16,17]. We noticed that this gap value is different from the absorption onset in $R(\omega)$ or optical conductivity, which gives smaller value close to 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. We find that this peak frequency matches well with the large superconducting gaps observed on the two small holelike and electronlike FS sheets with $\Delta \simeq 12$ meV measured relative to the Fermi level in ARPES [12].

Well below T_c, there is a substantial suppresssion in the low-frequency conductivity due to the formation of superconducting energy gap. According to the Ferrell-Glover-Tinkham (FGT) sum rule [18,19], the difference between the conductivity at $T \simeq T_c$ and $T \ll T_c$ (the so-called missing area; see the inset of Fig. 3) is related to the formation of a superconducting condensate,

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

where $\omega_{ps}^2 = 4\pi n_s e^2/m^*$ is the square of the superconducting plasma frequency, n_s is the condensed carrier density, and ω_c is the high-frequency cutoff frequency which should be chosen such that the ω_{ps}^2 converges smoothly. The penetration depth is related to the superconducting plasma frequency by $\lambda = c/\omega_{ps}$. Equation (1)

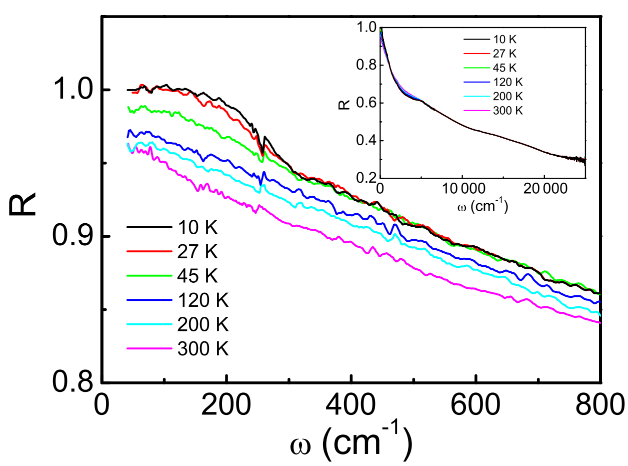

FIG. 2 (color online). *T*-dependent $R(\omega)$ curves in the far-infrared region. The inset shows $R(\omega)$ over broad frequencies up to 25 000 cm^{-1}.

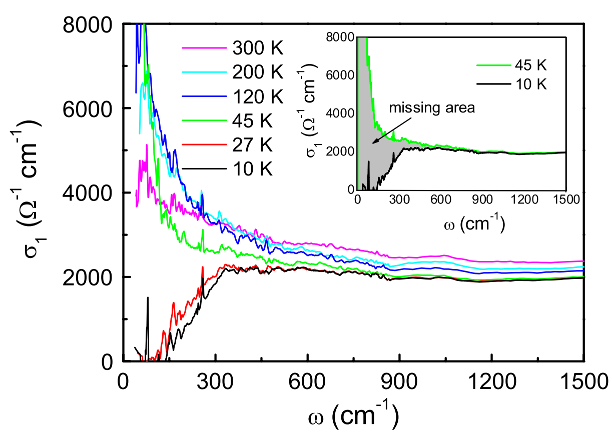

FIG. 3 (color online). *T*-dependent $\sigma_1(\omega)$ curves. The inset shows $\sigma_1(\omega)$ at 10 and 45 K. The shaded area represents the missing area due to the opening of superconducting energy gap.

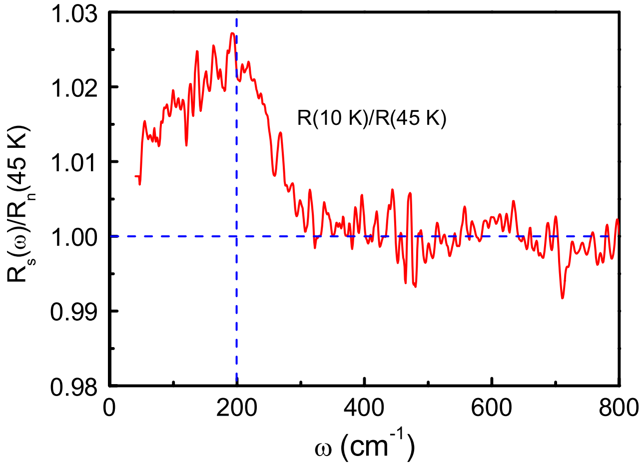

FIG. 4 (color online). The reflectance $R(\omega)$ at 10 K normalized to the values at 45 K in the normal state. A peak near 200 cm^{-1} is seen.

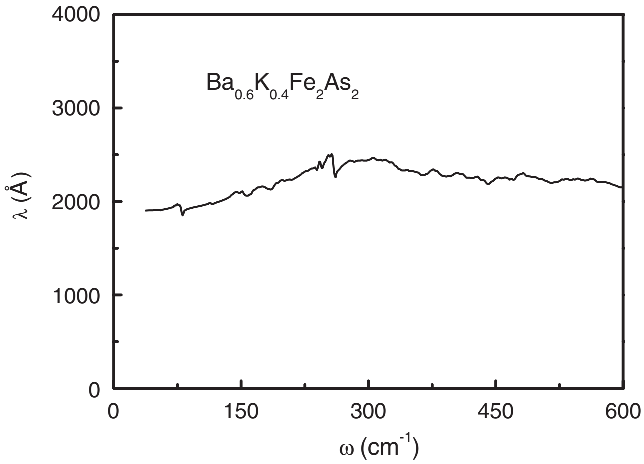

FIG. 5. Frequency dependent London penetration depth $\lambda(\omega) = c/\omega_{ps} = c/\sqrt{4\pi\omega\sigma_2(\omega)}$ for $Ba_{0.6}K_{0.4}Fe_2As_2$ at 10 K.

states that the spectral weight lost in $\sigma_1(\omega)$ in the superconducting state has been transferred to the zero frequency delta function response of the superconducting condensate. A direct estimation from the missing area gives $\lambda =$ 2080 Å.

The superconducting penetration depth can also be estimated from the imaginary part of the complex conductivity in the low-frequency limit via [20]

$$\lambda(\omega) = c/\omega_{ps} = c/\sqrt{4\pi\omega\sigma_2(\omega)}. \quad (2)$$

Equation (2) is related to the supercondcuting condensate in the real part of conductivity described in Eq. (1) by Kramers-Kronig transformation. The determination of λ from this equation at the low-frequency limit relies only on the imaginary part of conductivity at $T \ll T_c$. When the FGT sum rule is fulfilled, the λ 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 [21]. Figure 5 shows the $\lambda(\omega)$ obtained from above formula, at the low-frequency limit, $\lambda \simeq 1950$ Å. We find that the value of λ 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 supercondutor 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 2Δ, as can be seen in the inset of Fig. 3. While in the clean limit case, nearly all spectral weight associated with the condensate lies below 2Δ, so that no discernable change appears at 2Δ across the superconducting transition. It is still a controversial issue whether the high-T_c cuprates are in the dirty or clean limit and whether the superconducting gap is visible in infrared reflectance spectra [2,22–24]. 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.

Second, 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 the 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 2Δ. 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 [23,25,26].

Third, 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 the Uemura relation [27,28], which states that the superfluid density scales linearly with the transition temperature, $\rho_s = \omega_{ps}^2 = c^2/\lambda^2 \propto T_c$. The 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. Homes proposed another scaling relation, $\rho_s \simeq 65\sigma_{dc}T_c$ for BCS weak coupling case, where σ_{dc} is the value just above T_c

[23]. The present system seems to fit better to Homes's scaling relation.

To summarize, we performed infrared spectroscopy measurement on a superconducting $Ba_{0.6}K_{0.4}Fe_2As_2$ single crystal with $T_c = 37$ K. We observe clearly that the superconducting gap is present in the optical reflectance spectra with an 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 $R_s(10\ K)/R_n(45\ K)$. Those two values seem to match with the two distinct superconducting gaps observed in ARPES 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 $\lambda \simeq 2000 \pm 80$ Å.

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