# In-plane optical spectral weight redistribution in the optimally doped $Ba_{0.6}K_{0.4}Fe_2As_2$ superconductor*

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We performed detailed temperature-dependent optical measurements on optimally doped $Ba_{0.6}K_{0.4}Fe_2As_2$ single crystal. We examine the changes of the in-plane optical conductivity spectral weight in the normal state and the evolution of the superconducting condensate in the superconducting state. In the normal state, the low-frequency spectral weight shows a metallic response with an arctan ($T$) dependence, indicating a $T$-linear scattering rate behavior for the carriers. A high energy spectral weight transfer associated with the Hund's coupling occurs from the low frequencies below 4000 $cm^{-1}$ $\sim$ 5000 $cm^{-1}$ to higher frequencies up to at least $10^4$ $cm^{-1}$. Its temperature dependence analysis suggests that the Hund's coupling strength is continuously enhanced as the temperature is reduced. In the superconducting state, the FGT sum rule is conserved according to the spectral weight estimation within the conduction bands, only about 40% of the conduction bands participates in the superconducting condensate indicating that $Ba_{0.6}K_{0.4}Fe_2As_2$ is in dirty limit.

**Keywords:** iron-based superconductor, infrared spectroscopy, optical properties

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## 1. Introduction

The integral spectral weight of the real part of the optical conductivity with respect to frequency up to infinity is known as the $f$-sum rule. It is proportional to $n/m$ and is preserved by charge conservation.[1] This is particularly useful for the study and characterization of electronic properties in solids. In metallic systems, the conductivity sum rule is usually projected onto the lowest energy band and yields the classical plasma frequency.[2] In superconductors, it has been known that there is a noticeable loss of spectral weight in the optical conductivity below the critical temperature $T_c$, due to the presence of the superconducting gap and suppression of the single quasiparticles states.[3] The spectral weight loss below the superconducting energy gap reappears as a zero-frequency delta function associated with superfluid condensate. This is known as the Ferrell–Glover–Tinkham (FGT) sum rule.[4,5]

The newly discovered iron-based superconductors (FeSCs) have many novel properties, especially they possess multi-band electronic structure that may lead to a rich set of unconventional properties in both the normal and superconducting states.[6–8] The electronic structures of the multi-band systems should also be reflected in the charge excitation spectra which could be probed by the optical spectroscopy. Indeed, many previous optical investigations on the $BaFe_2As_2$ (Ba122) family FeSCs have also found two electronic subsystems, one is described by a narrow coherent Drude component, the other is referred to a broad incoherent background component.[9–12] The broad incoherent component is partially gapped due to the structural and spin-density-wave (SDW) transition in the parent compounds while s-wave-like superconducting gaps open in both components for superconducting compounds.[9,10] Moreover, the issues on the energy scale over which the spectral weight redistribution occurs due to the formation of superconducting gaps,[13] partial SDW gaps,[14,15] as well as the high frequency spectral weight transfer feature via the Hund's coupling have also been discussed.[15,16] However, due to the absence of fine temperature measurement, no detailed temperature dependence of the optical spectral weight redistribution has been investigated.

In this paper, we performed optical measurements on optimally doped $Ba_{0.6}K_{0.4}Fe_2As_2$ single crystal at 19 different temperatures from 5 K to 300 K. Our fine temperature measurement allows us to examine the changes of the spectral weight and the evolution of the superconducting condensate in $Ba_{0.6}K_{0.4}Fe_2As_2$. We found that the low frequency spectral weight increases with decreasing temperature, following an arctan ($T$) dependence, indicating a $T$-linear scattering rate behavior for the carriers. Moreover, upon cooling, the spectral weight below $4\times10^3$ $cm^{-1}$ $\sim$ $5\times10^3$ $cm^{-1}$ is transferred to very high frequency (up to at least $10^4$ $cm^{-1}$) due to strong Hund's coupling in iron-pnictides. The spectral weight redis-

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tribution associated with Hund's coupling exhibits a continuous evolution with temperature, even in the superconducting state. Furthermore, the spectral weight analysis in the superconducting state shows that the FGT sum rule is fulfilled, and only about 40% of the carriers participates in the superconducting condensate suggesting $Ba_{0.6}K_{0.4}Fe_2As_2$ is a dirty limit superconductor.

## 2. Experiments

High quality $Ba_{0.6}K_{0.4}Fe_2As_2$ single crystals with $T_c \simeq 39$ K were grown by the self-flux method using FeAs as the flux.[17] The *ab*-plane reflectivity $R(\omega)$ was measured at a near-normal angle of incidence on Bruker IFS113v and IFS66v spectrometers. An *in situ* gold overfilling technique[18] was used to obtain the absolute reflectivity of the samples. Data from 30 $cm^{-1}$ to $1.5 \times 10^4$ $cm^{-1}$ were collected at different temperatures from 5 K to 300 K on freshly cleaved surfaces on an ARS-Helitran crysostat. We extended the reflectivity to $4 \times 10^4$ $cm^{-1}$ at room temperature with an AvaSpec-2048 $\times$ 14 optical fiber spectrometer. The optical conductivity was obtained by Kramers–Kronig analysis of the reflectivity. At low frequencies, we employed a Hagen–Rubens ($R = 1 - A\sqrt{\omega}$) extrapolation for the normal state and a superconducting ($R = 1 - A\omega^4$) extrapolation below $T_c$. Above $1.5 \times 10^4$ $cm^{-1}$, for all temperatures, we utilized the room temperature data, followed by a constant reflectivity up to 12.5 eV, and terminated by a free-electron ($\omega^{-4}$) response.

## 3. Results and discussion

Figure 1(a) shows the temperature dependence of the real part of optical conductivity $\sigma_1(\omega)$ for $Ba_{0.6}K_{0.4}Fe_2As_2$. Above $T_c$, $\sigma_1(\omega)$ can be reasonably well described using a Drude–Lorentz model.[12] The Drude-like low-frequency $\sigma_1(\omega)$ narrows as the temperature decreases from room temperature to just above $T_c$. Below $T_c$, a dramatic suppression of the low-frequency $\sigma_1(\omega)$ sets in and the missing spectral weight has collapsed into the superfluid condensate. At 5 K, $\sigma_1(\omega)$ vanishes below $\sim 160$ $cm^{-1}$ ($\simeq 20$ meV) indicating a fully open gap.[13] We determined two superconducting gap values in our previous work:[19] $\Delta_1 \sim 4$ meV and $\Delta_2 \sim 10$ meV. Nevertheless, the gap opening at low energy is not the sole spectral feature observed with decreasing temperature. As reported by previous studies,[15,16] there exists another remarkable temperature-dependent spectral feature at much higher energy scale. As shown by the grey region in Fig. 1(b), the spectral weight below 4000 $cm^{-1}$ $\sim$ 5000 $cm^{-1}$ is gradually suppressed with decreasing temperature and transfer to higher frequencies up to at least $10^4$ $cm^{-1}$.

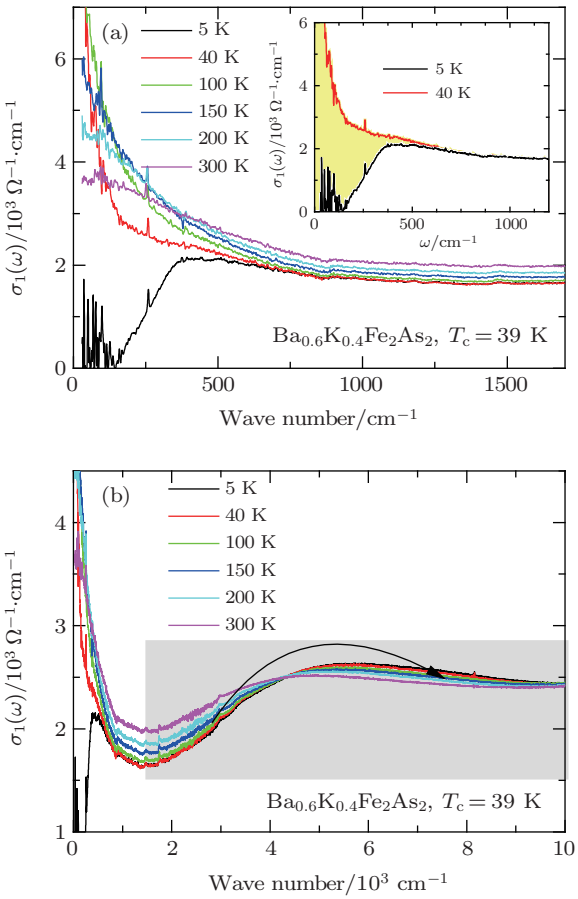



**Fig. 1.** (color online) (a) The optical conductivity of optimally doped $Ba_{0.6}K_{0.4}Fe_2As_2$ at several temperatures above and below $T_c$ in the far infrared region. Inset: The yellow area illustrates the spectral weight that has collapsed into the superconducting $\delta(\omega)$ function at the origin. (b) The optical conductivity of $Ba_{0.6}K_{0.4}Fe_2As_2$ up to $10^4$ $cm^{-1}$ at selected temperatures. The gray rectangle shows the spectral weight transfer in the high frequency region.

In experiments, the conductivity is measured up to a certain frequency cut-off. We quantify the changes in $\sigma_1(\omega)$ by introducing the restricted spectral weight:

$$N(\omega_c) = \int_{0^+}^{\omega_c} \sigma_1(\omega)\,d\omega, \tag{1}$$

where $\omega_c$ is the integral cut-off frequency. When $\omega_c \to \infty$, one gets the standard $f$-sum rule: $N(\infty) \propto \Omega_p^2 = 4\pi ne^2/m$, where $\Omega_p^2$ is the plasma frequency, $n$ refers to the total number of electrons (including the deep core electrons) and $m$ to the bare electron mass. When conduction electrons are limited to one band, and $\omega_c$ is representative of the bandwidth, then $n$ and $m$ refer to the conduction electrons density and their effective mass. Therefore, the spectral weight should be conserved for charge conservation.

Figure 2 shows the spectral weight as a function of temperature for $Ba_{0.6}K_{0.4}Fe_2As_2$ at different cut-off frequencies. First we discuss the result in the normal state. Figure 2(a) shows the temperature dependence of the spectral weight at $\omega_c = 200$ $cm^{-1}$, as the temperature decreases, the spectral weight below 200 $cm^{-1}$ increases all the way down to $T_c$. This

is due to the narrowing of the Drude absorption band. As discussed in another paper of our work,[12] the low-frequency spectral weight obeys an arctan($T$) temperature dependence indicated by the dashed line in Fig. 2(a), which is a result of the $T$-linear scattering rate behavior for the carriers in $Ba_{0.6}K_{0.4}Fe_2As_2$ that may arise out of spin-fluctuation scattering due to the possible existence of an antiferromagnetic quantum critical point in the superconducting dome. When integrating to a higher cut-off frequency, for instance $\omega_c = 4000$ cm$^{-1}$, an interesting phenomenon happens, as displayed in Fig. 2(b), the spectral weight below 4000 cm$^{-1}$ decreases upon cooling down. This phenomenon suggests that a large part of spectral weight below 4000 cm$^{-1}$ is transferred to a higher energy scale. Figure 2(c) shows that most of the spectral weight is roughly recovered when $\omega_c = 10^4$ cm$^{-1}$ is taken as the upper integration limit. In summary, the spectral weight analysis in the normal state implies that a free carrier metallic response at low frequencies coexists with a strange spectral weight transfer from below 4000 cm$^{-1}$ $\sim$ 5000 cm$^{-1}$ to very high, broad energy scale (up to at least $10^4$ cm$^{-1}$), this is not expected for a normal metal.

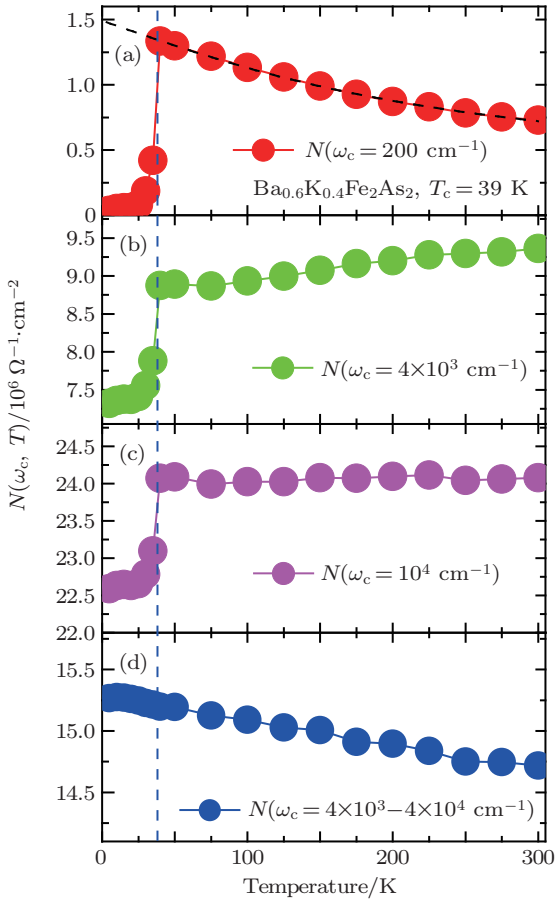



**Fig. 2.** (color online) (a) Spectral weight as a function of temperatures at different cut-off frequencies: (a) $\omega_c = 200$ cm$^{-1}$, (b) $\omega_c = 4000$ cm$^{-1}$, (c) $\omega_c = 10^4$ cm$^{-1}$, and (d) $\omega_c = 4\times10^3$ cm$^{-1}$–$10\times10^4$ cm$^{-1}$ for $Ba_{0.6}K_{0.4}Fe_2As_2$. The blue dashed line indicates the temperature at $T_c$, and the black dashed line is an arctan($T$) function fit to data.

Indeed, the high energy spectral weight above 5000 cm$^{-1}$ reflects the information of the incoherent and localized electrons. The spectral weight transfer from low energy to high energy suggests that a fraction of itinerant electrons with energy less than 5000 cm$^{-1}$ ($\sim$ 0.6 eV) transfers to the more localized part. By studying the energy scale (close to the Hund's coupling energy: 0.6 eV$\sim$ 0.9 eV in Ba122 FeSCs[15,20]) and comparing several possible physics origins of this high energy spectral redistribution, Wang *et al.*[16] elaborated that this spectral weight transfer feature is mainly caused by the strong Hund's coupling effect between the itinerant electrons and localized electron moment arising from the multiple Fe 3d orbitals. At low temperature, the thermal excitations become weak, then a fraction of relatively itinerant electrons tend to become more localized by Hund's coupling to local moments. Thus, we would expect to observe the spectral weight transfer from low energy to an energy scale higher than the Hund's coupling energy. Here, we might be able to estimate the strength of the Hund's coupling by the amount of the redistributed spectral weight in the high energy region. Figure 2(d) shows the temperature dependence of the spectral weight between 4000 cm$^{-1}$ and $10^4$ cm$^{-1}$ in the high energy region. The spectral weight in this region increases almost linearly as the temperature decreases from room temperature to the temperature well below $T_c$, implies that the Hund's coupling strength is continuously enhanced cross the normal state and superconducting state. Moreover, this unconventional spectral weight transfer from low to high energy is also observed in P-doped and Co-doped Ba122,[15,21] suggesting that the Hund's coupling could be a general phenomenon in Ba122 FeSCs.

Next we concentrate on the superconducting state. In all the three panels of Figs. 2(a)–2(c), a suppression of the spectral weight appears at $T_c$, indicated by the blue dashed line. From the optical conductivity, we can also see that a large part of the low-frequency optical spectral weight disappeared with the opening of the superconducting gaps, indicated by the yellow area in the inset of Fig. 1(a). This 'missing area' is referred to as the spectral weight of the superfluid condensate $\rho_s$ and may be estimated from the FGT sum rule,[4,5] $\rho_s \propto N(\omega_c, T \gtrsim T_c) - N(\omega_c, T \ll T_c)$. When the cut-off frequency $\omega_c$ covers the excitation spectrum responsible for the superfluid condensate, the FGT sum rule will be fulfilled. In the conventional superconductors, this happens at an energy corresponding roughly to superconducting energy gap $2\Delta$. However, in the underdoped high-$T_c$ cuprates, the recovery of the FGT sum rule goes to very high energy ($\sim$ 1 eV–2 eV), or the sum rule is even violated.[22–24] What should we expect for the FeSCs?

To confirm the FGT sum rule, we need to determine the superfluid weight, which is related to a London plasma

frequency $\Omega_L$, $\rho_s \equiv \Omega_L^2$. $\Omega_L$ can be determined from the imaginary part of the optical conductivity $\sigma_2(\omega)$ in the low-frequency limit where $\Omega_L^2 \propto \omega\sigma_2(\omega)$.[25,26] Next, we make an examination of the spectral weight transfer within the conduction bands and check whether the FGT sum rule is fulfilled. In the Ba122 FeSCs, a cut-off frequency of $\omega_c \simeq 1500$ cm$^{-1}$ is used for the estimation of the conduction bands spectral weight,[15,21] since most of the intraband transition processes are dominated by the optical conductivity responses below 1500 cm$^{-1}$, although such an estimation has a very small part of contribution from the low-frequency tail of the mid-infrared absorption. Indeed, as shown in Fig. 3, the blue solid circles show the spectral weight at $\omega_c = 1500$ cm$^{-1}$ as a function of temperature for $Ba_{0.6}K_{0.4}Fe_2As_2$, which is roughly conserved at the temperatures above $T_c$. This result implies that the band structure and the area of the Fermi surface almost do not change upon cooling. In the superconducting state, the spectral weight is suppressed by 40% indicating that about 40% of the conduction band electrons participate in the superconducting condensate. Actually, our earlier work[19] shows that a broad Drude component with scattering rate $\Gamma \sim 1000$ cm$^{-1}$ ($\simeq 10\Delta$) and a narrow Drude component with $\Gamma \sim 20$ cm$^{-1}$ ($\ll \Delta$) yield an excellent description of the low-frequency optical conductivity at $T \gtrsim T_c$, indicating the existence of two groups of carriers with different quasiparticle lifetimes. Therefore, when $T \ll T_c$, the spectral weight of the narrow Drude band almost completely condensates into superfluid, while most of carriers in the broad Drude band are not condensed. Hence, at least in some bands, $Ba_{0.6}K_{0.4}Fe_2As_2$ is in dirty limit. The red open circles in Fig. 3 are obtained by adding the superfluid weight $\rho_s$ calculated from $\sigma_2(\omega)$ to the spectral weight $N(\omega_c = 1500$ cm$^{-1})$ in the superconducting state. They follow the normal state very well demonstrating that the spectral weight lost within the conduction bands due to the superconducting condensate is recovered by the $\delta(\omega)$ function at zero frequency. Thus the FGT sum rule is satisfied within the accuracy of our measurements.

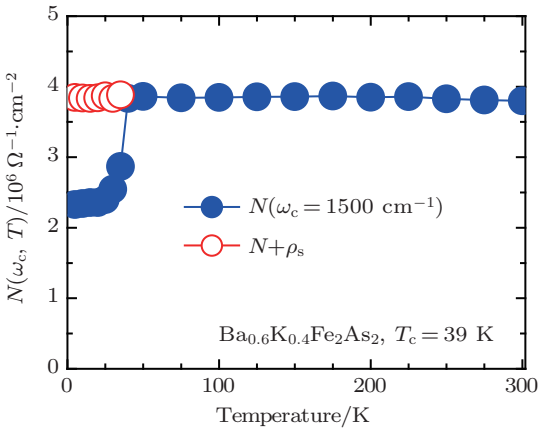



**Fig. 3.** (color online) The solid circles represent the spectral weight $N(\omega_c = 1500\ \mathrm{cm}^{-1})$ from Eq. (1) for $Ba_{0.6}K_{0.4}Fe_2As_2$. The open circles are obtained by adding the superfluid weight calculated from $\sigma_2(\omega)$ to $N(\omega_c)$.

## 4. Conclusion

We examine the changes of the in-plane optical conductivity spectral weight and the evolution of the superconducting condensate in the optimally doped $Ba_{0.6}K_{0.4}Fe_2As_2$. The low-frequency spectral weight in the normal state shows an arctan($T$) dependence, indicating a $T$-linear scattering rate behavior for the carriers. An unconventional spectral weight transfer from the low frequencies below 4000 cm$^{-1}$ $\sim$ 5000 cm$^{-1}$ to very high frequencies up to at least $10^4$ cm$^{-1}$ is found, which associates with the Hund's coupling. Its temperature dependence analysis suggests that the Hund's coupling strength is continuously enhanced as the temperature is reduced, even well below $T_c$. Moreover, this unconventional spectral weight transfer at the high energy scale seems to be universal in Ba122 FeSCs. In the superconducting state, the FGT sum rule is fulfilled corresponding to the spectral weight estimation within the conduction bands, and only about 40% of the conduction bands participates in the superconducting condensate indicating $Ba_{0.6}K_{0.4}Fe_2As_2$ is in dirty limit.

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