Observation of a pseudogap in the optical conductivity of underdoped Ba$_{1-x}$K$_x$Fe$_2$As$_2$

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We report the observation of a pseudogap in the $ab$-plane optical conductivity of underdoped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($x = 0.2$ and 0.12) single crystals. Both samples show prominent gaps opened by a spin density wave (SDW) order and superconductivity at the transition temperatures $T_{SDW}$ and $T_c$, respectively. In addition, we observe a pseudogap below $T^* \sim 75$ K, a temperature much lower than $T_{SDW}$ but much higher than $T_c$. A spectral weight analysis shows that the pseudogap is closely connected to the superconducting gap, indicating the possibility of its being a precursor of superconductivity. The doping dependence of the gaps is also supportive of such a scenario.

Among all the families of iron-pnictide superconductors discovered to date, the BaFe$_2$As$_2$ (Ba122) family is one of the most studied. The parent BaFe$_2$As$_2$ composition is a poor Pauli-paramagnetic metal with a structural and magnetic phase transition at 140 K$^2$. Superconductivity arises with the suppression of magnetism which can be achieved by applying pressure or chemical substitution.$^{2,4}$ The substitution of Ba with K atoms yields hole-doping$^2$ with a maximum $T_c \approx 39$ K and the substitution of Fe atoms by Co or Ni results in electron-doping$^4$ with a maximum $T_c \approx 25$ K. Extensive studies have been carried out in the parent BaFe$_2$As$_2$ electron-doped BaFe$_{2-x}$As$_2$ ($A = Co$, Ni)$^{12-15}$ as well as optimally hole-doped Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ compounds.$^{16,18}$ However, the hole-underdoped regime of the phase diagram is relatively unexplored.

This hole-underdoped region is arguably the most important regime because of the following two reasons. First, the superconducting mechanism is deeply tied with magnetism. The interplay between magnetism and superconductivity is manifest in this regime. In a considerably large portion of the underdoped regime, the SDW phase and superconductivity coexist.$^{19-23}$ Second, in cuprates, the most exciting, yet puzzling, physics takes place in the hole-underdoped regime. This regime thus is pivotal to the comparison between iron-pnictides and cuprates.

Xu et al.$^{24}$ have performed the surface sensitive angle-resolved photoemission (ARPES) measurements on underdoped Ba$_{1-x}$K$_x$Fe$_2$As$_2$. Their data showed a distinct pseudogap coexisting with the superconducting gap and suggested that both the pseudogap and superconductivity are driven by antiferromagnetic fluctuations. However, one key issue in understanding the origin of the pseudogap and, in particular, its relation to superconductivity is the question of whether it shares electronic states with the superconducting condensation.$^{25}$ Infrared spectroscopy probes the charge dynamics of bulk materials and the spectral weight analysis is a powerful tool to address this issue.

We present broadband infrared spectroscopy measurements on two underdoped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($x = 0.2$ and 0.12) single crystals. In both samples, the opening of the SDW gap and the superconducting gap was clearly observed on the optical conductivity. In addition, another small gap opens below $T^* \sim 75$ K, closely resembling the famous pseudogap in the hole-underdoped cuprates. We find that the SDW gap depletes the spectral weight available for the superconducting condensate, which suggests that the SDW order competes with superconductivity. However, both doping and temperature dependence of the spectral weight inside the pseudogap indicate that it shares the same electronic origin with the superconducting gap.

High quality Ba$_{1-x}$K$_x$Fe$_2$As$_2$ single crystals were grown by the self-flux method using FeAs as the flux.$^{26}$ The left panel of Fig. 1 shows the temperature dependence of the DC resistivity [$\rho(T)$] for the Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($x = 0.2$) sample. The $\rho(T)$ curve is characterized by a steep superconducting transition at $T_c = 19$ K. The inset shows the derivative of the resistivity $d\rho/dT$ as a function of temperature. The SDW transition manifests itself as a sharp peak in $d\rho/dT$ at $T_{SDW} = 104$ K, which corresponds to a small kink on the $\rho(T)$ curve. The right panel displays the same curves for the $x = 0.12$ sample, which has $T_c = 11$ K, and $T_{SDW} = 121$ K.

The $ab$-plane reflectivity [$R(\omega)$] was measured at a near-normal angle of incidence on Bruker IFS113v and IFS66v/s spectrometers. An in situ gold overfilling technique$^{27}$ was used to obtain the absolute reflectivity of the samples. Data from 20 to 12000 cm$^{-1}$ were collected at 18 different temperatures from 5 to 300 K on freshly cleaved surfaces. In order to use Kramers-Kronig analysis, we extended the data to the visible and UV range (10000 to 55000 cm$^{-1}$) at room temperature with
Figure 1. (color online) Left panel: Temperature dependence of the resistivity of Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($x = 0.2$) single crystal (red solid line). A steep superconducting transition can be seen at $T_c = 19$ K. The blue solid squares are values from the zero frequency extrapolation of the optical conductivity. The inset shows the derivative of the resistivity $d\rho/dT$ as a function of temperature. The sharp peak at 104 K in $d\rho/dT$ is associated with the SDW transition. The right panel depicts the same curves for $x = 0.12$ sample with $T_c = 11$ K, and $T_{SDW} = 121$ K.

Figure 2 shows the infrared reflectivity at selected temperatures for both samples up to 1200 cm$^{-1}$. The inset in each panel displays the reflectivity for the full measured range at 300 K. For the $x = 0.2$ sample, shown in the top panel, the reflectivity exhibits a metallic response and approaches unity at zero frequency. Below $T_{SDW} = 104$ K, a substantial suppression of $R(\omega)$ at about 650 cm$^{-1}$ sets in and intensifies with the decreasing temperature. Simultaneously, the low frequency reflectivity continues increasing towards unity. This is a signature of a partial SDW gap on the Fermi surface. Below 75 K, defined as $T^*$ here, another suppression of $R(\omega)$ appears in a lower energy scale ($\sim 150$ cm$^{-1}$) signaling the opening of a second partial gap (pseudogap) with a smaller value. Upon crossing the superconducting transition, which occurs at $T_c = 19$ K, the reflectivity below $\sim 150$ cm$^{-1}$ increases indicating the opening of a superconducting gap. Similar features are observed on $R(\omega)$ for the $x = 0.12$ sample as shown in the bottom panel of Fig. 2.

The real part of the optical conductivity $\sigma_1(\omega)$ was determined by Kramers-Kronig analysis of the measured reflectivity. Figure 3 shows $\sigma_1(\omega)$ at different temperatures for the two samples. The zero frequency extrapolations of $\sigma_1(\omega)$ represent the inverse dc resistivity of the sample, shown as blue solid squares in Fig. 1 which are in good agreement with the transport measurement. The top panel of Fig. 3 shows $\sigma_1(\omega)$ for Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($x = 0.2$) below 1700 cm$^{-1}$. At 150 K and 125 K, hence above $T_{SDW}$, a Drude-like metallic response dominates the low frequency optical conductivity. Below $T_{SDW}$, $\sigma_1(\omega)$ below about 650 cm$^{-1}$ is severely suppressed. Meanwhile, it increases in a higher energy scale from 650 cm$^{-1}$ to 1700 cm$^{-1}$. The optical conductivity for the normal state and that for the SDW state just below $T_{SDW}$ show an intersection point at about 650 cm$^{-1}$. As the temperature decreases, both the low energy spectral suppression and the high energy bulge become stronger; and the intersection point moves to a higher energy scale. This spectral evolution manifests the behavior of the SDW gap in this material: transfer of low frequency spectral weight to high frequencies. If we take the intersection points as an estimative of the gap values, we can see that the gap increases with decreasing temperature. Below $T^* \sim 75$ K, a second suppression in the optical conductivity below roughly 110 cm$^{-1}$ with a bulge extending from about 110 cm$^{-1}$ to 250 cm$^{-1}$ sets in and develops with the temperature decrease, implying the opening of the pseudogap on the Fermi surface. The inset of Fig. 3 shows the enlarged view of the low temperature optical conductivity at low frequencies, where the pseudogap is seen more clearly. This pseudogap is unlikely due to the SDW transition as (i) it opens at 75 K, well below $T_{SDW}$ and (ii) it redistributes spectral weight at a different, smaller energy scale.

The superconducting transition at $T_c = 19$ K implies the opening of a superconducting gap. As shown in the inset of Fig. 3 this leads to the reduction of the optical conductivity at low frequencies between 20 K and 5
The imaginary part of the optical conductivity. Nevertheless, its weight can be calculated from Figure 3. (color online) Top panel: Optical conductivity gap and superconducting) with doping also suggests that 

\[ \sigma(\omega) = \frac{1}{\omega} \int_{\omega_a}^{\omega_b} \sigma_1(\omega) d\omega, \]

where \( \omega_a \) and \( \omega_b \) are lower and upper cut-off frequencies, respectively. By choosing appropriate values for \( \omega_a \) and \( \omega_b \), one can study the relations among different phase transitions. When replacing \( \omega_b \) by 0 and \( \omega_b \) by \( \infty \), we fall back to the standard \( f \)-sum rule and the spectral weight is conserved.

Figure 4 shows the temperature dependence of the \( x = 0.2 \) sample spectral weight, normalized by its value at 300 K, at different cut-off frequencies. The vertical dashed lines denote \( T_c \), \( T^* \) and \( T_{SDW} \).
below that energy by fitting the low frequency normal state optical conductivity to a Drude model. The upper cut-off frequency ($\omega_b = 12000$ cm$^{-1}$) is high enough to cover the whole spectrum responsible for the phase transitions in this material. Hence the blue solid circles form a flat line at about unity, indicating that the spectral weight is conserved.

The red solid circles in the top panel show the temperature dependence of the normalized spectral weight with cut-off frequencies $\omega_0 = 0^+$ and $\omega_b = 650$ cm$^{-1}$. Here $0^+$ means that the superfluid weight is not included. Above $T_{SDW}$, the continuous increase of the normalized $SW_{650}$ with decreasing $T$ is related to the narrowing of the Drude band. This is the typical optical response of a metallic material. A strong spectral weight suppression occurs at $T_{SDW}$, which is the consequence of the SDW gap opening. At $T_c$, another sharp drop of the spectral weight breaks in, indicating the superconducting gap opening.

The temperature dependence of the normalized $SW_{1700}$, shown as green solid circles in the bottom panel of Fig. 4 provides clues about the relation between the superconducting and the SDW gaps. Above $T_{SDW}$, the material shows a metallic response which can be described by a Drude peak centered at zero frequency. With the temperature decrease, the DC conductivity increases and the scattering rate reduces. The continuous narrowing of the Drude band induces a transfer of spectral weight from the mid-infrared to the far infrared, resulting in the continuous decrease of the spectral weight observed in the 650–1700 cm$^{-1}$ range. Below $T_{SDW}$, the opposite behavior dominates the optical conductivity. The SDW gap depletes the spectral weight below 650 cm$^{-1}$ and transfers it to the 650–1700 cm$^{-1}$ range, leading to the continuous increase of $SW_{1700}$ with decreasing $T$. This behavior continues into the superconducting state and does not show any feature at $T_c$. These observations indicate that the SDW and superconducting gaps are separate and even act as competitive orders in this material.

If a partial gap is due to a precursor order of superconductivity, for example preformed pairs without phase coherence, once the long range superconductivity is established, a significant part of the spectral weight transferred to high frequencies by the partial gap should be transferred back to low energies and join the superconducting condensate. However, a partial gap due to a competitive order to superconductivity depletes the low-energy spectral weight and holds it in a high energy scale without transferring it back to the superfluid weight below $T_c$. From the normalized $SW_{1700}$ vs $T$ curve (green solid circles) we note that no loss of spectral weight is observed at $T_c$. This means that the spectral weight transferred to high frequencies by the SDW gap remains in the high frequency scale and does not contribute to the superconducting condensate. Therefore, the SDW acts as a competitive order to superconductivity in this material.

Along these lines, the origin of the pseudogap and its relationship to superconductivity can be revealed by a close inspection of the temperature dependence of the normalized $SW_{110}$, shown as pink solid circles in the bottom panel. Above $T^*$, this curve shows the same feature as the normalized $SW_{650}^*$ vs $T$ curve, i.e., continuous increase upon cooling down followed by a suppression at $T_{SDW}$ due to the SDW gap opening. At $T^*$, the spectral weight in the 110–250 cm$^{-1}$ range reaches a minimum and starts to increase with decreasing temperature. This is due to the opening of the pseudogap. The pseudogap, opening at $T^*$, depletes the spectral weight below 110 cm$^{-1}$ and retrieves it in the 110–250 cm$^{-1}$ frequency range, leading to the increase of $SW_{110}$ below $T^*$. An interesting phenomenon happens to the pseudogap when the material undergoes the superconducting transition. In contrast to the case of the SDW gap, a significant loss of spectral weight in the 110–250 cm$^{-1}$ frequency range is observed below $T_c$. This observation indicates that the spectral weight transferred to the 110–250 cm$^{-1}$ range by the pseudogap joins the superconducting condensate when superconductivity is established. Hence, the pseudogap is likely a precursor order with respect to superconductivity.

In summary, we measured the optical conductivity of two underdoped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ($x = 0.2$ and 0.12) single crystals. In both samples, besides the SDW gap and superconducting gap, the optical conductivity reveals another small partial gap (pseudogap) opening below $T^*$ closing $75$ K an intermediate temperature between $T_{SDW}$ and $T_c$. A spectral weight analysis shows that the SDW gap diminishes the low energy spectral weight available for the superconducting condensate while the pseudogap shares the same electronic states with the superconducting gap. These observations, together with the doping dependence of these gaps, suggest the SDW as a competitive order and the pseudogap as a precursor to superconductivity.

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