Evidence of a pseudogap for superconducting iron-pnictide $\text{Ba}_{0.6+\delta}\text{K}_{0.4-\delta}\text{Fe}_2\text{As}_2$ single crystals from optical conductivity measurements

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Abstract. We report the observation of pseudogap (PG) behavior in optical conductivity in an underdoped $\text{Ba}_{0.6+\delta}\text{K}_{0.4-\delta}\text{Fe}_2\text{As}_2$ ($T_{\text{onset}} \approx 36$ K) single crystal far above $T_c$, up to 100 K ($\approx 3T_c$). Wide separation of the energy scales of the magnetic and superconducting (SC) correlations in the $\text{Ba}_{0.6+\delta}\text{K}_{0.4-\delta}\text{Fe}_2\text{As}_2$ sample enabled us to establish that the PG structures observed in optical conductivity in the range of 50–150 cm$^{-1}$ are uniquely driven by the SC correlation, and the magnetic correlation coexists in the far separated range of about 700 cm$^{-1}$. Theoretical calculations, based on the preformed Cooper pair model, provided an excellent description of the temperature evolution of the SC correlation in optical conductivity data from below to above $T_c$.

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1. Introduction

The pseudogap (PG), a phenomenon of the opening of gap-like features above the superconducting (SC) transition temperature $T_c$, has been universally observed in the so-called unconventional superconductors (UCSs) such as the high-$T_c$ cuprates (HTC) [1, 2], some heavy fermion superconductors [3] and the recently discovered iron-pnictide superconductors [4–7]. As such, the origin of the PG is believed to hold the key to understanding the pairing mechanism of the UCSs, in particular the HTC superconductors.

Although there are several theoretical proposals for the PG origin, for example the resonating valence bond [8], magnetic correlation [2], SC [9–11] correlation, Mott gap [12], nematic order [13], etc, clear consensus has not yet been reached among researchers. One of the main obstacles to the experimental study of the PG phenomena is that the competing correlations—as listed above—in the UCSs often have similar energy scales so that it is naturally difficult to distinguish the responsible correlation from others. For example, after 25 years of research on HTC, it became known that there is more than one kind of PG in HTC: the low-energy PG immediately above $T_c$ and the high-energy PG far above $T_c$, and possibly more differentiation of the high-energy PG. The low-energy PG has been almost proven to originate from the SC fluctuations [9–11]. As to the high-energy PG, there is still a lack of a complete consensus [1, 2, 8, 12, 13], and it is also possible that different UCSs may have different origins for their own PG.

In this paper, we measured the optical conductivity of Ba$_{0.64+\delta}$K$_{0.4-\delta}$Fe$_2$As$_2$ ($T_{\text{onset}} \approx 36$ K) single crystal, and observed the PG behavior in the range of 50–150 cm$^{-1}$ far above $T_c$, up to 100 K ($\approx 3T_c$). Our sample is unique in that the SC and magnetic correlations simultaneously appear, yet with widely separated energy scales. This unique scale separation of two correlations in the Ba$_{0.64+\delta}$K$_{0.4-\delta}$Fe$_2$As$_2$ compound enabled us to establish that the PG structures observed in the range of 50–150 cm$^{-1}$ in optical conductivity are driven by the SC correlation and the magnetic correlation is ruled out as an origin. This is the first observation of the PG behavior in optical conductivity of the UCS where the magnetic correlation is clearly separated from the SC correlation. We showed with the theoretical calculations that the preformed Cooper pair model [14] provided an excellent description of the temperature evolution of the optical...
Figure 1. Temperature dependence of the dc electrical resistivity of a Ba$_{0.64+\delta}$K$_{0.4-\delta}$Fe$_2$As$_2$ single crystal. Inset (a) is the enlarged dc resistivity between 10 and 60 K. A sharp SC transition is observed at $T^\text{onset}=36$ K. Inset (b) shows $d^2\rho/d^2T$ as a function of temperature. The inflection point is at 100 K.

conductivity data from below to above $T_c$. Our observation will put a strong constraint on the pairing mechanism of the iron-based superconductors as well as other UCSs.

2. The sample and measurement

High-quality single crystals of Ba$_{0.64+\delta}$K$_{0.4-\delta}$Fe$_2$As$_2$ were grown by the Bridgeman method with a sealed tungsten crucible [15] at 1550 °C. The ratio of room temperature to residual resistivity, $\rho(300 \text{ K})/\rho(T_c)$, is 7.5 and the sharp SC transition is observed at $T^\text{onset}=36$ K in the dc-resistivity data in figure 1. Compared to $T_c \approx 38.5$ K of Ba$_{0.68}$K$_{0.32}$Fe$_2$As$_2$ [16], the doping level of our sample is less than 0.32. Also compared to the undoped BaFe$_2$As$_2$ data [17], our sample shows no evidence of the spin density wave (SDW) transition, such as a discontinuity in the resistivity (see the inset of figure 1) and magnetic susceptibility.

We measured the optical spectroscopy of this sample for a broad range of temperatures from 4 to 300 K and frequencies from 15 to 28 000 cm$^{-1}$. The key advance of our spectroscopic measurement is the achievement of high-resolution THz spectroscopy down to 15 cm$^{-1}$ and combined infrared (IR) spectroscopy to cover the wide frequency range. With this technique the common limitation of IR spectroscopy—which does not provide reliable data below $\approx 100 \text{ cm}^{-1}$ that is often the most interesting frequency regime for the study of the SC correlation—has been overcome. We used a Michelson-type rapid-scan Fourier spectrometer (Jasco FTIR610) in the frequency range of 40–12 000 cm$^{-1}$ and the Martin–Puplett-type rapid scan Fourier spectrometer (Jasco Faris) for the THz frequency range of 15–200 cm$^{-1}$ for temperatures of 4–300 K. We have built a specially designed feedback positioning system to maintain the overall uncertainty level. The uncertainties of our data in frequency ranges below and above 100 cm$^{-1}$ were maintained as less than 0.6 and 0.3%, respectively. For more details of experimental methods of reference setting, control of data uncertainty, etc, see the appendix.

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Figure 2. Reflectivity spectra, $R(\omega)$, at several temperatures between 4 and 300 K up to 1000 cm$^{-1}$. At $T < T_c$, $R(\omega)$ approaches unity at low frequencies due to the SC gap formation. The inset shows wide range spectra up to 28 000 cm$^{-1}$. Metallic behavior is observed for the whole measured frequency range.

3. Results and discussion

Figure 2 shows the raw data on the reflectivity $R(\omega, T)$ for different temperatures from 4 to 300 K. It clearly shows a systematic evolution of the temperature-dependent features below 150 cm$^{-1}$, the region where the previous IR spectroscopy studies [17, 18] did not have sufficient resolution. The reflectivity data below $T_c$ of 4, 8, 20 and 30 K become flat at low frequencies approaching unity ($R \rightarrow 1$)—within the uncertainty level of our measurement which is 0.6% below 100 cm$^{-1}$—reflecting the presence of an SC gap, and it shows a systematic drop starting from $\omega \approx 50$ cm$^{-1}$, indicating the beginning of absorptions. These features of the low-frequency data below 100 cm$^{-1}$ are beyond the experimental uncertainty mentioned above and are intrinsic properties of the sample; the quality of our data should be compared with the data of [17, 18].

This liability and control of the uncertainty level of the data at low frequencies is of ultimate importance for studying the SC and PG features [19].

The broad depletion in the region of 200–800 cm$^{-1}$ for $T \leq 100$ K is qualitatively similar to the data of the undoped BaFe$_2$As$_2$ [17] below its magnetic transition temperature in shape as well as for its temperature evolution. While the undoped BaFe$_2$As$_2$ showed a sharp drop and kink feature in resistivity [17] at the magnetic transition temperature $T_{SDW} = 138$ K, our sample does not show any hint of a transition in resistivity in figure 1 around 100–150 K. Therefore, we conclude that our sample develops only a short-range magnetic correlation for $T \leq 100$ K. Finally, the fact that our reflectivity data go flat at low frequencies approaching $R \rightarrow 1$ below $T_c$ proves that our sample is microscopically homogeneous and has no phase separation of the magnetic and SC domains. All these put together, it is clear that our sample simultaneously exhibits both the SC correlation and the short-range SDW correlation. However, the most unique and interesting feature of our sample is that these two correlations appear in widely separated energy (frequency) regimes: the SC correlation in the region below 150 cm$^{-1}$ and the SDW correlation in the region of 200–800 cm$^{-1}$.
Figure 3. Real part of the optical conductivity, $\sigma_1(\omega, T)$, at several temperatures between 4 and 300 K. Dashed lines represent the Drude fitting for 300 K (gray dashes) and 50 K (dark gray dashes). At $T < T_c$, $\sigma_1(\omega, T)$ rapidly decreases and becomes gapped at low frequencies below about 50 cm$^{-1}$. The inset is a plot of plasma frequencies versus temperatures $\omega_p(T)$ obtained from the optical $f$-sum rule, $\omega_p^2 \approx 8 \int_0^{\omega_c} \sigma_1(\omega, T) \, d\omega$. A cutoff frequency $\omega_c = 2500$ cm$^{-1}$ is used instead of infinity to avoid the variation caused by the high-frequency interband transitions. The $\omega_p^2$ is nearly constant in the PG temperature region from 50 to 100 K and then abruptly decreases below $T_c$, signaling the long-range phase coherence of Cooper pairs as expected. From the missing area of the plasma frequency $\omega_p$, the penetration depth can be extracted as $\lambda = c/\omega_p$. We obtained 251 nm, which is comparable to the value 208 nm in [18].

3.1. The Kramers–Kronig transformation: optical conductivity $\sigma_1(\omega, T)$

For a more convenient analysis, we convert $R(\omega, T)$ into the real part of conductivity $\sigma_1(\omega, T)$ using the Kramers–Kronig transformation [19, 20] in figure 3. For the extrapolations of the data, the high-frequency part beyond 28 000 cm$^{-1}$ is extrapolated with the standard $1/\omega^4$ form. For the low-frequency region below 15 cm$^{-1}$, the Hagen–Rubens formula is used in the normal state and $R(\omega, T) = 1$ is taken in the SC state. Figure 3 shows the results of the real part of optical conductivity, $\sigma_1(\omega, T)$, at various temperatures. The data of 200 and 300 K display a very broad Drude form centered at $\omega = 0$ and monotonically decrease until the interband transitions begin around 2000 cm$^{-1}$. Decreasing the temperature to 100 and 50 K (still above $T_c = 36$ K), three noticeable changes occur: (i) the spectral density around 700 cm$^{-1}$ builds up to form a broad hump structure; (ii) concomitantly, the Drude peak rapidly sharpens; (iii) another new hump structure around 100 cm$^{-1}$, widely separated from the 700 cm$^{-1}$ structure, appears.

The first feature is the development of the short-range SDW correlation discussed with figure 2. Instead of two distinct peaks resolved at 360 and 890 cm$^{-1}$ in the undoped BaFe$_2$As$_2$ sample [17], a single broad peak centered around 700 cm$^{-1}$ is formed for $T < 100$ K and it became almost temperature independent from 50 K to lower temperatures. The second feature
is the evolution of the coherent quasiparticle formation, which interestingly coincides with the development of the SDW correlation in the mid-IR frequency range around 700 cm\(^{-1}\). However, the most striking feature of our data is the third one, i.e. the appearance of the gap-like hump structure far above \(T_c\) (50 and 100 K data) on top of the Drude spectra in the range of 50–200 cm\(^{-1}\). Below \(T_c\), the data of 30, 20, 8 and 4 K display a clean opening of the SC energy gap with the absorption edge at about 50 cm\(^{-1}\) (~3.12 meV) [21] and peaked around 100 cm\(^{-1}\). Comparing with this SC gap structure around 100 cm\(^{-1}\) below \(T_c\), the hump structure around 100 cm\(^{-1}\) in the 50 and 100 K data appears to be a continuous evolution from the SC gap structure below \(T_c\) and we believe that it is the unmistakable observation of PG behavior solely driven by the SC correlation persisting up to temperatures of \(\approx 3T_c\).

There might be other possibilities for the hump structure around 100 cm\(^{-1}\) above \(T_c\). Recent experiments on Ba(K)Fe\(_2\)As\(_2\) [18] and BaFe(Co)\(_2\)As\(_2\) [23–25] have also measured the low-frequency absorptions around 100 cm\(^{-1}\) in \(\sigma_1(\omega, T)\) below \(T_c\), with some differences in shape and frequency scale. But all agreed that they have observed an SC gap opening in their spectra. What was not clear was: do the whole of these spectra represent the free carriers that form the SC gap(s)? While Li [18] took this interpretation, Heumen \textit{et al} [23] and Lobo \textit{et al} [24] argued that part of these low-frequency absorption spectra below \(T_c\) is contributed from a localized Lorentz oscillator peaking at about 100 cm\(^{-1}\) with a width of about 50 cm\(^{-1}\) because their spectra also displayed a hump-like feature at this frequency range even above \(T_c\)—although it was not as clearly distinct as in our data. Then the obvious question is the possible origin of such a narrow absorption peak at such a low frequency which also exactly coincides with the SC \(2\Delta\) energy scale. Heumen \textit{et al} [23] proposed that it is due to the interband transition, arguing that there are several bands passing close to the Fermi level in these compounds. We disagree with this interpretation because the interband transition scenario should satisfy several conditions: (i) a band passing as close as 100 cm\(^{-1}\) (~12.5 meV) to the Fermi level, (ii) its band width as narrow as 50 cm\(^{-1}\) (~6.25 meV) or the allowed \(k\)-space for transition should be extremely limited and (iii) none of these bands should cross the Fermi level because then the interband transition hump structure should show a systematic shift with the SC gap opening below \(T_c\), which was not observed in our data. Therefore, we think that it is very unlikely to satisfy all these conditions for the narrow interband transition. On the other hand, Lobo \textit{et al} [24] suspected the presence of the localized carriers induced by disorder for its origin. There is no possible mechanism to produce a bound state induced by disorder on the over the SC energy scale. Therefore, we suspect that these authors have also measured a PG feature in their samples.

The high-frequency features in figure 3 are easy to understand. The SC gap structure goes through a kind of dip around 200 cm\(^{-1}\) and is then connected to the low end of the SDW correlation structure. The hump around 4000 cm\(^{-1}\) is the standard interband transitions that were observed in the previous papers [17, 18], too. Finally, the shape of the PG structure seen in 50 and 100 K data looks as if it consists of two peaks of two \(\Delta\) gaps around 50 and 100 cm\(^{-1}\) (\(\Delta_{\text{small}} \approx 3.5\) meV and \(\Delta_{\text{large}} \approx 7\) meV). So we will assume that there exist two gaps \(\Delta_{\text{small, large}}\) in our theoretical fitting in the next section. This choice of the gap sizes is somewhat smaller than, for example, the ARPES data of the \(x = 0.4\) sample [21]. However, it is now known there is a substantial variation of the gap size along the \(z\)-direction [26] and hence it is expected that the optical and thermodynamic measurements should sensitively pick the smallest gap size in the entire Brillouin zone.

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3.2. Subtracted optical conductivity $\sigma_{\text{subt}}(\omega, T)$

Since we have identified the SC feature and its evolution above $T_c$, we would like to focus on these low-energy spectra and their temperature evolution. Therefore, we subtract out the magnetic structure and the interband transition structure from the total $\sigma_1(\omega, T)$. The magnetic structure is weakly temperature dependent below 50 K and the interband transition around 4000 cm$^{-1}$ is almost temperature independent. Therefore, we used the 4 K data for subtraction. Figure 4 shows the subtracted optical conductivity, $\sigma_{\text{subt}}(\omega, T)$, and the inset shows the Lorentz oscillators used to fit the subtracted structures. The subtracted optical conductivity below $T_c$, $\sigma_{\text{subt}}(\omega, T < T_c)$, exhibits an absorption edge about 50 cm$^{-1}$ and a peak at around 100 cm$^{-1}$ and then monotonically decreases afterwards. Also it is now much better seen, in $\sigma_{\text{subt}}(\omega, T > T_c)$ above $T_c$ up to 100 K, that the PG structure around 100 cm$^{-1}$ exists on top of the Drude spectra. As mentioned before, the PG structure of 50 and 100 K data in figure 4 looks as if it actually consists of two peaks with two adjacent frequencies. This two-peak structure becomes unresolved below $T_c$ because of their close adjacency [21, 22].

4. Preformed Cooper pairs model

We now discuss a theoretical model that can explain the temperature evolution of $\sigma_{\text{subt}}(\omega, T)$. The subtracted conductivity $\sigma_{\text{subt}}(\omega, T)$ is the contribution only from the free carriers that enter the SC condensate below $T_c$ and is released to the Drude spectra of the conductivity above $T_c$. The optical conductivity below $T_c$ was first fitted by the standard Kubo formula with the SC gap. For above $T_c$, we adopted the phase incoherent preformed Cooper pair model [14] and

\[ \sigma_{\text{subt}}(\omega) = \sigma_{\text{Drude}}(\omega) + \sigma_{\text{PG}}(\omega) \]

The inset shows the Lorentzian oscillators used for the subtracted spectra.
followed the recipe of Franz and Millis [27] for the phase fluctuation average. The theoretical results are overlaid on the experimental data $\sigma_{\text{subt}}(\omega, T)$ in figure 4. The theoretical conductivity $\sigma_{\text{theory}}(\omega, T > T_c)$ indeed captures the main features of $\sigma_{\text{subt}}(\omega, T > T_c)$ qualitatively well, namely the PG structure as well as the Drude spectra.

First, to fit $\sigma_{\text{subt}}(\omega, T > T_c)$ below $T_c$, we consider an isotropic s-wave pairing state and assumed that the SC condensate consists of two SC gaps $\Delta_{S1}$ and $\Delta_{S2}$; the two-gap assumption is motivated by the shape of the PG structure above $T_c$, which looks as if it has two adjacent humps, and it is not an essential assumption for our calculations since one-gap fitting would also be possible using a larger damping rate. Below $T_c$, the real part of the optical conductivity is calculated by a standard Kubo formula with the sum of two bands ($a = S1, S2$) as follows:

$$\sigma_1(\omega, T) = -\sum_a \frac{\text{Im} \Lambda_{a,xx}(\omega)}{\omega}$$

with

$$\text{Im} \Lambda_{a,xx}(\omega) = \pi e^2 \int d^2k \omega v_{a,x}^2(k) \text{Tr}[\tilde{A}_a(k, \omega + \omega') \tilde{A}_a(k, \omega')][f(\omega + \omega') - f(\omega)],$$

where $v_{a,x}$ is the Fermi velocity along the x-direction of the electrons of the band $a$, $f(\omega)$ is the Fermi–Dirac distribution function and $\tilde{A}_a(k, \omega)$ is the $2 \times 2$ spectral density matrix of Nambu Green’s function of the band $a$ in the SC state defined as $A_a(k, \omega) = -\text{Im} \tilde{G}_a(k, \omega)/\pi$ and

$$\tilde{G}_a(k, \omega) = \frac{\omega \tau_0 + \tilde{\xi}_a(k) \tau_3 + \Delta_a \tau_1}{\omega^2 - \tilde{\xi}_a^2(k) - \Delta_a^2},$$

where $\tau_i$ are the Pauli matrices and $\Delta_a$ is the SC gap of the band $a$. The gap value $\Delta_a$ was straightforwardly determined without much ambiguity. We chose $50 \text{ cm}^{-1}$ for the smaller absorption gap $2\Delta_{S1}$ and $100 \text{ cm}^{-1}$ for the larger absorption gap $2\Delta_{S2}$.

When we calculate $\sigma_1(\omega, T)$ using the above equations (1)–(3), we note that it is well known that a clean homogeneous superconductor does not allow any absorption signal in $\sigma_1(\omega, T < T_c)$ [28]. Theoretically established cases to allow absorption for $\omega > 2\Delta_S$ are: (1) extreme anomalous skin effect limit ($\lambda \ll \xi$; $\lambda = \text{penetration depth}, \xi = \text{SC coherence length}$) [29]; (2) extreme dirty limit with $1/\tau \gg 2\Delta_S$. The first condition simply does not apply for the iron superconductors where $\lambda > \xi$ generally holds. We ruled out the dirty limit condition, too, which was commonly employed by previous works [18, 23–25] to justify the use of the Mattis–Bardeen formula [29], usually assuming $1/\tau \approx 4–8\Delta_S$. The problem with this assumption is that such a large number of disorders in the sign-changing s-wave superconductors—which is the widely accepted SC gap symmetry for Ba(K)Fe$_2$As$_2$ and BaFe(Co)$_2$As$_2$ compounds—would completely fill in the gap with impurity band if not completely kill the superconductivity itself. The clean opening of the absorption gap in our data (see figures 3 and 4) as well as Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ data [18] clearly shows that there is almost no in-gap state formed by impurity scattering, indicating a clean limit superconductor ($1/\tau \ll 2\Delta_S$).

The remaining possibility to allow absorption in $\sigma_1(\omega, T)$ is the inelastic scattering which dynamically damps the quasiparticles and was used to explain the absorption feature in the optical conductivity of the HTS below $T_c$ [30]. To this end, we introduce a phenomenological damping to the quasiparticles as $\tilde{\omega} = \omega + i\Gamma + i\text{Im} \Sigma_{\text{inela}}(\omega)$ to fit the overall line shape, where $\Gamma$ is a constant damping due to disorder and $\text{Im} \Sigma_{\text{inela}}(\omega)$ represents a dynamic scattering rate due to a coupling with various bosonic modes. Since we argued above for a clean limit superconductor, we chose a small value of $\Gamma = 0.1\Delta_{S1}$. The form of $\text{Im} \Sigma_{\text{inela}}(\omega)$ was
experimentally extracted for BaFe_{1.85}Co_{0.15}As_2 [25] and Ba_{0.55}K_{0.45}Fe_2As_2 [31] and shown to be roughly proportional to \sim \omega for the relevant frequency range of \omega < 400 cm^{-1}; it should become \sim \omega^2 at very low frequencies of the Fermi liquid regime that is, however, likely less than the 2\Delta_c scale in these compounds [25, 31] and will be cut out anyway in the SC and PG phases. Hence, we phenomenologically chose Im \Sigma_{1,2}(\omega) = 0.8\omega for [\omega] > \Delta_{1,2} in our calculations for the overall best fitting, where the dynamic scattering rate is cut out below the threshold energy \Delta_{1,2} for assuming the isotropic s-wave gaps in each band. The authors of [30] proposed the threshold frequency of Im \Sigma_2(\omega) as 3\Delta_c for the HTC assuming that the coupled boson mode itself is affected by the SC gap opening. If this strong feedback effect occurs in iron-based superconductors, too, we just need to interpret the absorption edge in our optical conductivity data as 4\Delta_{1,2} instead 2\Delta_{1,2}. Without knowing \nu_{a,b} and the density of states N_a(0), the relative weight of the contribution from each band is taken as a fitting parameter. We chose the ratio of the spectral weights of the S1 and S2 bands as 2 to 1. The results for T < T_c are shown in figure 4 by open square symbols.

Above T_c, we adopted the phase incoherent preformed Cooper pair model [14]. In order to simulate the phase fluctuations, we followed the recipe of Franz and Millis [27] and averaged the Nambu Green’s function with Doppler shift energy \eta in the quasiparticle excitations as

\[ \tilde{G}(k, \omega) = \int d\eta P(\eta) \tilde{G}(k, \omega - \eta), \]

where the probability distribution of \eta is given by \[ P(\eta) = e^{-\eta^2/2W} / \sqrt{2\pi W} \] with W \approx 3.48\alpha_e(T/T_c)\Delta^2. \alpha_e is a parameter derived from the XY model and was estimated as \approx 0.009 in the HTC by Franz and Millis [27], for example. Here, we take the whole W as a fitting parameter and chose W = 0.144\Delta_{S1}^2 with T = 1.2\Delta_{S1} for best fitting. The results are plotted in figure 4 by open circle symbols.

5. Conclusion

We measured the optical reflectivity R(\omega, T) of hole doped Ba_{0.6+x}K_{0.4-x}Fe_2As_2 (T_c^{onset} \approx 36 K) for temperatures from 4 to 300 K and frequencies from 15 to 28 000 cm^{-1}. We combined the THz and IR spectroscopies with a specially designed feedback positioning system to maintain the overall uncertainty level as less than 0.6%. We identified the SC gap structure in 50–150 cm^{-1} below T_c and its continuous evolution to the PG structure far above T_c, up to 100 K (\approx 3T_c). Below 100 K, our data simultaneously displayed a development of the magnetic correlation structure in the mid-IR frequency range around 700 cm^{-1}, together with the SC correlation structure around 100 cm^{-1}. This coexistence and yet wide separation of the energy scales of the two correlation structures led us to the conclusion that the PG structure in 50–150 cm^{-1} is solely driven by the SC correlation. Our observation, therefore, made Ba_{0.6+x}K_{0.4-x}Fe_2As_2 the first example of an unconventional high-T_c superconductor where the SC and magnetic correlations are clearly separated and the SC correlation-driven PG structure is observed for a wide temperature range up to three times T_c. Theoretical calculations based on the phase incoherent preformed Cooper pair model [14, 27] provided an excellent description of the PG evolution of our optical conductivity data above T_c. Our findings put a strong constraint on the pairing mechanism of the iron-based superconductors and will shed new light on the origin of the PG in UCSs including the HTC.
Acknowledgments

After the submission of this paper, we became aware of the work by Dai et al [32], where they have obtained similar optical data and drawn a similar conclusion for the PG observation as ours with the more underdoped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ samples with $x = 0.12$ and 0.2 with corresponding $T_c = 11$ K and 19 K, respectively. YSK was supported by the Basic Science Research Program (2010-0007487 and 2010-0006484) and the Mid-career Researcher Program (no. R01-2008-000-20586-0). YB was supported by grant numbers NRF-2010-0009523 and NRF-2011-0017079 funded by the National Research Foundation of Korea. AVB was supported by the US Department of Energy and by UCOP-TR01.

Appendix. The experimental method used to set up the absolute reflectivity and the feedback device used to control the uncertainty

For the reference of the absolute reflectivity in the THz and IR regions, an in-situ gold over-coating technique, which is a common method, was used in our experiment. In this setup, a Au mirror as well as a sample was vertically mounted on the cryostat as shown in figure A.1. The sample was mounted on the sample holder, a mountain-shaped Cu block, using silver paste. Moving the sample holder vertically, the intensity of the reflected beam from the sample and Au mirror was sequentially measured using a Si bolometer cooled down to 4.2 K. After these measurements, Au was evaporated on the sample surface in situ and the intensity of the reflected beam was measured with the same method. The absolute reflectivity was calculated from the ratio of these two measurements. Here, the fresh Au mirror was taken as the perfect reflectance calibration point whose reflectance value was larger than 99.6% below 100 cm$^{-1}$ derived from the Hagen–Rubens formula.

In the reflectivity measurement of most pnictide superconductors including ours, the sample size is much smaller than the beam size ($\phi \sim 8$ mm). In this case, the interference from the sample edge and on-passing optical filters/windows is the main source of uncertainty of the measurement. In order to reduce this uncertainty, it is crucial to locate the same vertical
positions of the targets (the sample, Au mirror and Au-coated sample) through measurements. The usual method of finding the vertical position by searching for the maximum intensity of reflection from the targets does not work because the intensity is nearly the same wherever the targets are inside the beam area.

In order to resolve this problem we used the feedback method. As shown in figure A.1, we mounted a small piece of reference mirror on the opposite side of the sample and searched for the maximum laser intensity reflected from the mirror with exactly the same size of laser beam. This was performed by vertically moving the sample holder using a stepping motor with the resolution of 0.1 µm per step. Once the distances between the reference mirror and the sample and between the sample and Au mirror were measured, we can find the same vertical position of the targets through our measurements and thus we could reduce the uncertainty level to 0.6 and 0.3% below and above 100 cm⁻¹, respectively.

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