Origin of the Spin density wave instability in AFe$_2$As$_2$ (A=Ba, Sr) as revealed by optical spectroscopy

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We performed optical spectroscopy measurement on single crystals of BaFe$_2$As$_2$ and SrFe$_2$As$_2$, the parent compounds of FeAs based superconductors. Both are found to be quite metallic with fairly large plasma frequencies at high temperature. Upon entering the spin-density-wave (SDW) state, formation of partial energy gaps was clearly observed with the presence of surprisingly two different energy scales. A large part of the Drude component was removed by the gapping of Fermi surfaces (FS). Meanwhile, the carrier scattering rate was even more dramatically reduced. We elaborate that the SDW instability is more likely to be driven by the FS nesting of itinerant electrons rather than a local-exchange mechanism.

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The recent discovery of superconductivity with transition temperature $T_c$ above 50 K in RFeAsO$_{1-x}$F$_x$ (R=La, Ce, Sm, Pr, Nd, etc) has created tremendous interests in the scientific community. Those compounds crystallize in a tetragonal ZrCuSiAs-type structure, which consists of alternate stacking of edge-sharing Fe$_2$As$_2$ tetrahedral layers and R$_2$O$_2$ tetrahedral layers along c-axis. The parent compound LaFeAsO itself is not superconducting but shows strong anomalies near 150 K in resistivity, magnetic susceptibility, specific heat, etc. Based on experimental observations and first principle calculations, it is suggested that the ground state is a spin-density-wave (SDW) ordered state with a stripe-type (or collinear) spin configuration. The predicted magnetic structure was confirmed by subsequent neutron diffraction experiment, although the neutron data indicated that a subtle structural distortion occurs first near 150 K, and the SDW long range order establishes at a slightly lower temperature. With fluorine doping, the SDW order is suppressed and superconductivity emerges. The very closeness of the superconducting phase to the SDW instability suggests that the magnetic fluctuations play a key role in the superconducting pairing mechanism.

Investigating the origin of the antiferromagnetic (AFM) SDW instability in the parent compound is an essential step towards understanding the mechanism of superconductivity in doped systems. The stripe-type AFM order was first suggested to result from the nesting between the hole and electron Fermi surfaces (FS) of itinerant electrons. Alternatively it was proposed that the superexchange interaction mediated through the off-plane As atom plays a key role in the spin configuration formation. A stripe-type AFM would arise when the next nearest neighbor exchange becomes larger than half of the nearest neighbor exchange interaction. Whether an itinerant picture or a local superexchange mechanism should be taken as a starting approach becomes one of the most important issues for those systems.

Very recently, it is found that the ThCr$_2$Si$_2$-type ternary iron arsenide BaFe$_2$As$_2$, which contains identical edge-sharing Fe$_2$As$_2$ tetrahedral layers as in LaFeAsO, exhibits a similar SDW instability at 140 K. It is therefore suggested that BaFe$_2$As$_2$ could serve as a new parent compound for ternary iron arsenide superconductors. Shortly after that, the superconductivity with $T_c$=38 K was found in K-doped BaFe$_2$As$_2$, which was suggested to be a hole-doped iron arsenide superconductor. The SDW instability was also found in SrFe$_2$As$_2$ with a higher transition temperature near 200 K while K-doping again introduces the superconductivity with $T_c$=38 K. For both parent compounds, the same stripe-type AFM order was confirmed by neutron experiments. A great advantage of those ternary iron arsenide compounds is that it is much easier to grow large size single crystals.

In this letter we present optical study on both BaFe$_2$As$_2$ and SrFe$_2$As$_2$ single crystals. We find the undoped compounds of Fe-pnictides are quite metallic with rather high plasma frequencies, $\omega_p \geq 1.5$ eV. Upon entering into the SDW state, formation of energy gaps was clearly observed. Surprisingly, the optical measurement revealed two distinct energy scales in the gapped state. Associated with the gapping of the FS, a large part of Drude component is removed, meanwhile the carrier scattering rate shows even steeper reduction. Beyond the character energy for SDW gap, another spectral suppression in R(\omega) which covers a much higher energy scale is present even above SDW transition temperature. The physical implications of those results were discussed. Our study favors an itinerant electron approach for the driving mechanism of SDW instability.

Single crystals of BaFe$_2$As$_2$ and SrFe$_2$As$_2$ (space group I4/mmm) were grown from the FeAs flux method. The plate-like crystals could be easily cleaved, resulting in very shiny surface. The T-dependent dc resistivity was measured by the four contact technique in a Quan-
immediately suggests a dramatic reduction of the carrier edge upon cooling the sample into SDW ordered state. One can see a rather sharp low-ω partially gapped and the compounds are still metallic being clearly that the F Fermi surfaces are only par-

The optical reflectance measurements were performed on a combination of Bruker IFS 66v/s, 113v and a grating-type spectrometers on newly cleaved surfaces (ab-plane) for AFe₂As₂ (A=Ba, Sr) single crystals in the frequency range from 40 to 50000 cm⁻¹. An in situ gold and aluminum overcoating technique was used to get the reflectivity R(ω). The real part of conductivity σ₁(ω) is obtained by the Kramers-Kronig transformation of R(ω).

The main panels of Fig. 1 (a) and (b) focus on the low frequency R(ω) up to 1800 cm⁻¹. For both compounds, R(ω) exhibits a metallic response, and approaches to unity at zero frequency. The most prominent feature is a substantial suppression in R(ω) for \( T < T_{SDW} \), which is a strong optical evidence for the formation of energy gaps. The low-ω reflectance increases faster towards unity at zero frequency than those at high T. As a consequence, one can see a rather sharp low-ω reflectance edge. This indicates clearly that the Fermi surfaces are only partially gapped and the compounds are still metallic below \( T_{SDW} \). The change of R(ω) from an overdamped linear-ω dependent behavior to a well-defined reflectance edge upon cooling the sample into SDW ordered state immediately suggests a dramatic reduction of the carrier scattering rate, while its low-energy location implies a considerable reduction of carrier density. A quantitative analysis will be given below. It is noted that the low-T R(ω) displays an almost linear-ω dependence over a certain frequency range below the suppression. This special shape of the suppression leads to the two-peak structure in optical conductivity.

The middle panels of Fig. 1 show the conductivity spectra \( \sigma_1(\omega) \) below 2500 cm⁻¹. The Drude-like conductivity can be observed for all spectra at low frequencies. For BaFe₂As₂ \( (T_{SDW}=138 \text{ K}) \), a weak feature around 890 cm⁻¹ develops for \( T=130 \text{ K} \) in \( \sigma_1(\omega) \), then the spectra are severely suppressed at low frequencies for 60 K and 10 K, resulting in a pronounced double-peak character at 360 and 890 cm⁻¹. Associated with the low-ω reflectance edge, a very sharp and narrow Drude component emerges below the double peaks. Very similar features can be seen for SrFe₂As₂ crystal, but the double peak features appear at higher energies, i.e. 500 and 1360 cm⁻¹, being consistent with the higher \( T_{SDW} \) for SrFe₂As₂. The electrodynamics of broken symmetry ground states, such as the superconducting and density wave states, have been well explored and understood. Due to different coherence factors, a density wave state behaves different from an s-wave superconductor at the gap frequencies in optical conductivity. In an s-wave superconducting state at \( T=0 \), the absorption smoothly rises at the gap frequency, while for a density wave state, a sharp maximum appears in conductivity at the gap frequency. Based on those studies, we can identify the double peak energies as the two SDW gaps. The observation of two distinct SDW gaps should be associated
with different Fermi surfaces, and reflect the multi-band property in FeAs based compound. From the gap values and SDW transition temperatures, we obtained the ratio of $2\Delta/k_B T_{SDW} \approx 3.5-3.6$ for the smaller gap, and 9-9.6 for the larger gap for the two compounds. The smaller gap coincides roughly with the gap value expected by the conventional BCS relation, while the large one is very different. Similar two-gap behaviors were also found in optical measurement for the itinerant SDW metal Cr.\textsuperscript{24}.

Due to the presence of two different gap values in the SDW ordered state, direct information on where the FS is is available \textsuperscript{25} for the larger gap for the two compounds. The smaller gap opening is also absent in this work. Apparently the local picture is less favored, and the itinerant scenario provides more reasonable explanation for the driving mechanism.

The above observations have important implication for the driving mechanism of SDW instability. As mentioned above, the key issue here is whether an itinerant picture based on FS nesting or a local superexchange mechanism is a proper approach. Our optical studies clearly demonstrate that the parent compound has a high itinerant carrier density with the plasma frequency a bit higher than 1.5 eV before SDW transition, and is rather metallic both above and below SDW ordering temperatures. Furthermore, the partial gap openings below SDW ordering temperatures are consistent with the expectation of nesting scenario where the temperature dependence of the gap should resemble that of BCS theory. On this basis, we think that the local picture is less favored, and the itinerant scenario provides more reasonable explanation for the driving mechanism.

Besides the dramatic spectral change at low frequencies, both compounds display very similar and pronounced spectral feature at the mid-infrared region. Take BaFe\textsubscript{2}As\textsubscript{2} as an example, the mid-infrared component takes up a large spectral weight as shown in the inset of Fig. 2 (b). The peak at such a high energy is usually ascribed to the interband transition. However, a puzzling problem is that the spectra above the peak energy exhibit an apparent T-dependence that $R(\omega)$ is obviously suppressed with decreasing T below 5000 cm\textsuperscript{-1} (Fig. 1 (a)). Such a gap-like feature is present at all temperatures. Indeed, an analysis for the spectral weight in $\sigma_1(\omega)$ revealed that the suppressed spectral weight below 5000 cm\textsuperscript{-1} is transferred to higher energies. Figure 2 (c) plots the spectral weight for $\sigma_1(\omega)$ at 10 and 300 K. Below 200 cm\textsuperscript{-1}, the narrowing Drude peak with decreasing T (see the extrapolation data in Fig. 2 (b)) yields a growing dc conductivity thus a larger low frequency spectral weight for T=10 K. Above 200 cm\textsuperscript{-1}, the SDW double-gap develops which strongly reduces the low T Drude weight, leading to the first suppression below 1000 cm\textsuperscript{-1} in Fig. 2 (c). The lost Drude weight fills into the SDW double-peak, and the total spectral weight is almost recovered.
around 2000 cm$^{-1}$ for 10 K. Then the T-dependent suppression before the mid-infrared peak results in the second spectral weight suppression at 10 K near 3000 cm$^{-1}$. The lost weight finally recovers at about 8000 cm$^{-1}$. We would like to emphasize that this gap-like feature is not directly related to the SDW order. This is because (1) the mid-infrared suppression feature is present above $T_{SDW}$, (2) similar features exist for K- or Co-doped superconducting samples where the SDW order and the associated low energy gap structures are completely absent [28], and (3) the energy scale is much larger than the SDW gaps. Usually a gap formation is associated with a broken symmetry state. As the AFe$_2$As$_2$ compounds are in their paramagnetic phase with a tetragonal crystal structure above $T_{SDW}$, both magnetic and crystal structures are in very high symmetry state, one would hardly expect an even higher magnetic or crystal structural symmetry at higher temperatures. Definitely, further experimental and theoretical works are necessary to understand this high energy gap-like behavior.

To summarize, the ab-plane optical measurements of AFe$_2$As$_2$ (A=Ba, Sr) single crystals were performed. For the SDW state, our findings indicate the Fermi surface are completely absent, [28] and would like to emphasize that this gap-like feature is not the SDW state, our findings indicate the Fermi surface above $T_{SDW}$. Definitely, further experimental and theoretical works are necessary to understand this high energy gap-like behavior.

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