Normal state charge dynamics of Fe$_{1.06}$Te$_{0.88}$S$_{0.14}$ superconductor probed with infrared spectroscopy

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We have used optical spectroscopy to probe the normal state electrodynamic response of Fe$_{1.06}$Te$_{0.88}$S$_{0.14}$, a member of the 11 family of iron-based superconductors with $T_c$ = 8 K. Measurements have been conducted over a wide frequency range (50 - 50000 cm$^{-1}$) at selected temperatures between 10 and 300 K. At low temperatures the material behaves as an "incoherent metal": a Drude-like peak is absent from the optical conductivity, and all optical functions reveal that quasiparticles are not well defined down to the lowest measured temperature. We introduce "generalized spectral weight" analysis and use it to track temperature induced redistribution of spectral weight. Our results, combined with previous reports, indicate that the 11 family of iron-based superconductors might be different from other families.

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I. INTRODUCTION

Iron-based superconductors are currently at the focus of condensed-matter research. Discovered less than two years ago, these novel materials have attracted attention not only because of their high critical temperatures, but also because of their similarities with cuprates. Their phase diagram resembles that of the cuprates, most notably, the superconducting state seems to develop from an unconventional normal state, after magnetic order is destroyed by doping. They are also layered materials, consisting of FeAs, FeTe, FeS or FeSe planes, separated by spacer layers. However, there are also some important differences. Most notably, the parent compounds of cuprates are antiferromagnetic insulators, whereas the parent compounds of iron-based superconductors are antiferromagnetic spin density wave (SDW) metals.

Optical spectroscopy is a powerful probe of electrodynamic response of high-$T_c$ superconductors$^{3-5}$. Optical constants provide insight into low-energy excitations and charge dynamics, critical for understanding physics of strongly correlated systems. The information obtained from optical constants can be used to test existing theories and/or stimulate development of new theoretical models. Optical spectroscopy is also a crucial experimental method for electronic band structure determination.

Several families of iron-based superconductors have been discovered and they are conveniently referred to as the "11", "1111" or "122" families$^{6-8}$. The 11 family is peculiar because the spacer layers are absent, and it is believed that this family will allow the intrinsic properties of iron-containing planes to be isolated. However, most of optical studies so far have focused on the 122 family$^{9-13}$ (and to a lesser extent 1111 family$^{14,15}$) for which large single crystals can be grown. In this work, we have investigated the electrodynamic response of a member of the 11 (FeTe) family. To the best of our knowledge this is the first optical study of a superconducting member of this family. The only previous IR study on 11 family was on a non-superconducting Fe$_{1.05}$Te (Ref. 16).

Structural analysis has shown that the exact chemical composition of the analyzed sample is Fe$_{1.06}$Te$_{0.88}$S$_{0.14}$ (Ref. 17). Note that in addition to being doped with S, this sample also has 6 % of excess iron, which might play important role in charge dynamics$^{16,18}$. Magnetization measurements have revealed that the studied system undergoes structural and magnetic transitions, with transition temperature around 23 K (Ref. 17). Transport measurements on the other hand do not display any signatures of these transitions, and DC resistivity monotonically increases as temperature decreases down to 8 K, when the system undergoes superconducting transition and the resistivity abruptly drops to zero.

Infrared (IR) reflectance measurements were performed at The University of Akron on a Bruker IFS 66v/s, whereas UV-visible experiments were conducted using Varian/Cary 300. An overfilling technique was used to obtain the absolute values of reflectance from the sample with surface area of approximately 1 mm $\times$ 1 mm (Ref. 19). Electrodynamic response was probed in the frequency range 50 - 50 000 cm$^{-1}$ (6 meV - 6.2 eV) and as a function of temperature in the range 10 K - 300 K, all in the normal state. The optical constants were extracted from reflectance data using Kramers-Kronig (KK) analysis. In addition, we have performed magneto-optical measurements at 4.2 K (in the superconducting state) with magnetic fields up to 18 Tesla.

II. EXPERIMENTAL RESULTS

Figure 1 displays raw reflectance data $R(\omega)$ and the real part of optical conductivity $\sigma_1(\omega)$. The absolute value of reflectance gradually decreases with frequency...
from about 0.9 at 50 cm\(^{-1}\) to about 0.25 at 50000 cm\(^{-1}\), which can be interpreted as metallic behavior. However the temperature dependence is opposite from expected\(^{25}\), as the reflectivity decreases with decreasing temperature. This anomalous behavior is even more obvious in \(\sigma_1(\omega)\): zero-energy (Drude-like) peak is absent from the data (except maybe at 300 K) and the conductivity decreases monotonically with decreasing temperature at the lowest measured frequencies. This result indicates that at low temperatures quasiparticles are not well defined, which will become even more obvious from the extended Drude analysis below. Fe\(_{1.06}\)Te\(_{0.88}\)S\(_{0.14}\) therefore can be considered to be an "incoherent metal"\(^{21}\).

Although the overall behavior of reflectance is metallic, the plasma edge cannot be clearly resolved in the spectra, as reflectance gradually decreases with frequency. At higher frequencies the response is dominated by interband transitions, but they also are not easily discerned in the spectra. A shoulder in optical conductivity around 14000 cm\(^{-1}\) (1.7 eV) may originate from transitions involving iron 3\(d\) states, similar to what was predicted by Haule et al. in a theoretical study of another iron-based superconductor\(^{22}\). We also do not observe any phonon peaks in the far-IR, similar to Fe\(_{1.05}\)Te (Ref.\(^{10}\)). Structural and magnetic transitions at 23 K do not seem to have any significant effect on optical spectra.

Theoretical calculations of FeSe, based on LDA+DMFT, predicted "incoherent metal" with a pseudogap at low frequencies\(^{21}\), in accord with our results. These calculations also predict a smooth crossover of optical conductivity to a power law behavior \(\sim \omega^{-\eta}\) at higher frequencies\(^{25}\). Dashed black line in Fig. 1(b) represents the best fit to the optical conductivity and the obtained power law \(\eta \simeq 0.4\).

To gain further insight into the electronic properties of Fe\(_{1.06}\)Te\(_{0.88}\)S\(_{0.14}\) we fit the data using a standard Drude-Lorentz (DL) model\(^{3,4,20}\). The minimal model to achieve a good fit consisted of a Drude and three Lorentzian modes, centered at around 300 cm\(^{-1}\), 3000 cm\(^{-1}\) and 14800 cm\(^{-1}\). The total fits at all temperatures, as well as the three individual contributions at 10 K, are shown in Fig. 1(b) with gray lines. The lowest lying oscillator displays most prominent temperature dependence. Its energy and intensity grow significantly as temperature decreases. The mid-IR peak at 3000 cm\(^{-1}\) (372 meV) might be a generic feature of iron-based superconductors; similar peaks have been observed in other families.\(^{5}\) The oscillator at 14800 cm\(^{-1}\) simulates the effect of interband transitions, presumably involving iron 3\(d\) states, as discussed above.

In the one-component approach one assumes that only a single type of carriers are present in the system, but their scattering rate acquires frequency dependence\(^{3,4,20}\). Within the so-called "extended" Drude model one calculates the optical scattering rate \(1/\tau(\omega)\) and effective mass \(m^*(\omega)/m_b\) from the complex optical conductivity \(\sigma(\omega)\) as:

\[
\frac{1}{\tau(\omega)} = \frac{\omega_p^2}{4\pi m_b} \left[ \frac{1}{\sigma(\omega)} \right]
\]

\[
m^*(\omega) = \frac{\omega_p^2}{4\pi m_b} \left[ \frac{1}{\sigma(\omega)} \right] \frac{1}{\omega}
\]

where the plasma frequency \(\omega_p^2 = 4\pi\epsilon^2 n/m_b\) (\(n\) is the carrier density and \(m_b\) their band mass) can be estimated from the integration of \(\sigma_1(\omega)\) up to the frequency of the onset of interband absorption. However, as pointed out above, the plasma edge is not very prominent in the spectra of Fe\(_{1.06}\)Te\(_{0.88}\)S\(_{0.14}\), rendering the value of plasma frequency ambiguous. Instead, we fit the value of \(\omega_p\) in Eq. 2 so that the effective mass at frequencies around 3500 cm\(^{-1}\) is equal to unity (dashed line in Fig. 2(b)). The best fit is achieved for \(\omega_p = 26,000\) cm\(^{-1}\) (3.2 eV) and the results of the analysis are shown in Fig. 2. The features of an "incoherent metal" are now obvious. The scattering rate is relatively flat and featureless at room
temperature, but at low temperatures it develops a peak below 180 cm$^{-1}$. The effective mass, on the other hand, becomes negative. These features of the spectra are indications that the quasiparticle concept, on which the extended Drude model is based, is not applicable at low temperatures.

III. GENERALIZED SPECTRAL WEIGHT ANALYSIS

Model-independent sum rules are important tools in condensed-matter physics$^{23}$. The so-called effective spectral weight function $N(\omega)$ is frequently used for the analysis of IR spectra. It is defined as$^{23,20}$:

$$N(\omega) = \int_0^\omega \sigma_1(x)dx,$$  

(3)

and for $\omega \to \infty$ it becomes the global oscillator strength sum rule:

$$N(\infty) = \int_0^\infty \sigma_1(x)dx = \frac{\pi ne^2}{2m_e},$$  

(4)

which is a statement on the conservation of electric charge$^{23}$. Eq. (3) is often used to quantify spectral weight redistribution between temperatures $T_1$ and $T_2$ in the form:

$$\Delta N(\omega) = N_{T_1}(\omega) - N_{T_2}(\omega).$$  

(5)

Fig. 2 shows the results of this analysis applied to Fe$_{1.06}$Te$_{0.88}$S$_{0.14}$ at $T_1 = 77$ K and $T_2 = 10$ K. $\Delta N(\omega)$ has a characteristic shape, which indicates that the spectral weight is removed from the low frequency region, below 1000 cm$^{-1}$ and is transferred to higher frequencies, in the region around 2000-4000 cm$^{-1}$. Within the error bars, the spectral weight is recovered by the mid-IR region. However, $\Delta N(\omega)$ spectrum cannot reveal the energy scale at which the transfer of spectral weight occurs.

In order to address this question we introduce ”generalized spectral weight” function $\mathcal{N}(\Omega)$:

$$\mathcal{N}(\Omega) = \int_0^\infty \sigma_1^T(x + \Omega)\sigma_1^T(x)dx$$  

(6)

The idea comes from the correlation functions frequently used in signal processing$^{24}$. More recently (auto)correlation function was used for the analysis of ARPES spectra, where it is directly related to the quasiparticle density of states$^{25}$. Note that $\mathcal{N}(\Omega)$ is a function of the frequency shift $\Omega$, not the upper integration limit $\omega$. Function $\mathcal{N}(\Omega)$ is expected to display characteristic features at the values of energy shifts $\hbar\Omega$ that connect regions between which a large amount of spectral weight is transferred. In practical applications, function $\mathcal{N}(\Omega)$ is usually dominated by the spectral weight which does not participate in redistribution, and these characteristic features might not be obvious. In those cases the first derivative of $\mathcal{N}(\Omega)$ is useful. Examples of these calculations will be presented in a separate publication$^{26}$.

In Fig. 3 we show the results of generalized spectral weight analysis (Eq. (6)) applied to Fe$_{1.06}$Te$_{0.88}$S$_{0.14}$ at $T_1 = 77$ K and $T_2 = 10$ K. The $\mathcal{N}(\Omega)$ is dominated by the spectral weight that does not participate in transfer, so instead we display the first derivative $\mathcal{N}'(\Omega)$. The $\mathcal{N}'(\Omega)$ spectrum reveals a broad peak centered around 2250 cm$^{-1}$ (280 meV). We take this as the characteristic energy scale over which the majority of spectral weight is transferred between 10 and 77 K.

IV. DISCUSSION

In a density functional study of FeS, FeSe and FeTe it was reported that the Fermi surface and electronic structure of these compounds is similar to those of iron pnictides (1111 and 122 families). Therefore, the 11 family was suppose to be a model system in which to study the intrinsic properties of iron-containing planes. However, our results, combined with previous IR studies, indicate that there might be some important differences between the electronic structure of 11 and the other families.
Testing. Recent ARPES study of a parent compound is absent. Optical spectra reveal that spectral weight is Fe contrasted with a coherent response which has been resistivity changes character from insulating to metallic on charge transport than in Fe-ized spectral weight (Eq. 6) analysis for Fe.

FIG. 3: (Color online). Spectral weight (Eq. 5) and general- families display well defined Drude-like modes. Similar to both undoped (parent) and doped phases of these 1

appears below structural and magnetic phase transition in Fe-10 K and T-2 = 77 K.

The reflectance of Fe1.06Te0.88S0.14 is similar to the reflectance of Fe1.05Te (Ref. 16). In the far-IR region the reflectance of Fe1.05Te decreases with decreasing temperature, resulting in a reduction of conductivity in far-IR region, similar to what we observe in Fe1.06Te0.88S0.14. Chen et al. speculate that this incoherent transport in Fe1.05Te is caused by strong scattering from excess iron. However, there are also some important differences between Fe1.06Te0.88S0.14 and Fe1.05Te. We observe no rapid increase in conductivity at low frequencies for 10 K measurements. On the other hand in Fe1.05Te a narrow Drude-like peak develops in the optical conductivity at low temperatures. This coherent behavior appears below structural and magnetic phase transition at 65 K, which implies that it is related to SDW order. In Fe1.05Te this phase transition has stronger influence on charge transport than in Fe1.06Te0.88S0.14: the DC resistivity changes character from insulating to metallic below the transition.

Incoherent charge transport in the 11 family should be contrasted with a coherent response which has been observed in 1111 and 122 families. Infrared spectra of both undoped (parent) and doped phases of these families display well defined Drude-like modes. Similar to Fe1.05Te SDW transition has a dramatic effects on their optical properties. The response of parent compounds BaFe2As2 and SrFe2As2 becomes even more coherent below the SDW transitions as the width of Drude mode is reduced by an order of magnitude. The infrared spectra of these parent compounds are dominated by the mid-IR peak, which may have the same origin as the peak we observe in Fe1.06Te0.88S0.14 around 3000 cm-1 (372 meV).

The absence of SDW gap from IR spectra of both Fe1.06Te0.88S0.14 and Fe1.05Te (Ref. 16) is also interesting. Recent ARPES study of a parent compound Fe1.05Te (Ref. 28) has also revealed that the SDW gap is absent. Optical spectra reveal that spectral weight is shifting with temperature, and the generalized spectral weight analysis we introduced indicates that a typical energy scale for the shift is about 280 meV. The spectral weight of Fe1.06Te0.88S0.14 is removed from the low-energy region, which can be interpreted as a pseudogap feature, however we point out that this behavior starts already at room temperature (Fig. 1) and therefore is unlikely to be related to SDW transition. All this indicates that the electronic structure of the 11 family might be different from the 1111 and 122 families, for which clear signatures of SDW gaps have been observed. IR studies on these two families have found a gap (or even several gaps) in the excitation spectra. On the other hand both the parent compound and a doped sample studied in this work did not reveal the presence of a gap in their excitation spectra.

Finally we address what happens below 8 K, when the system becomes superconducting. We have performed magneto-optical studies in 18 Tesla superconducting magnet at the National High Magnetic Field Lab. Fig. 4 displays the results of these measurements. The magneto-reflection ratio R(18 T)/R(0 T) is shown as a function of frequency. Apart from the vertical offset within the noise level the ratio is a straight line, which indicates the absence of field-induced effects in Fe1.06Te0.88S0.14. This is in contrast with BaFe2-xCo_xAs2 with T_c = 22 K (optimally doped member of the 122 family), where clear field-induced changes have been observed caused by the suppression of the superconducting gap. We speculate that field-induced changes in Fe1.06Te0.88S0.14 are not observed because either they are below the detection limit of our experiment, or because the superconducting gap is outside of our frequency window.
V. SUMMARY

In summary, we have presented the results of infrared and optical spectroscopy studies of novel iron-based superconductor Fe$_{1.06}$Te$_{0.88}$Sr$_{0.14}$. The results indicate incoherent normal state charge transport and absence of well defined quasiparticles at all temperatures down to $T_c$. We have introduced "generalized spectral weight analysis" and used it to track redistribution with temperature. The analysis reveals that the characteristic energy scale for the spectral weight shifts is approximately 280 meV. Our results, combined with previous reports, indicate that there are important differences between 11 and other families of iron-based superconductors.

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