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Incoherent Interplane Optical Response of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ Superconductor

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We report on the interplane c axis electronic response of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ investigated by infrared spectroscopy. We find that the normal-state c axis electronic response of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ is incoherent and bears significant similarities to those of mildly underdoped cuprates. The c axis optical conductivity $\sigma_c(\omega)$ of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ does not display well-defined Drude response at all temperatures. As temperature decreases, $\sigma_c(\omega)$ is continuously suppressed. The incoherent c axis response is found to be related with the strong dissipation in the ab plane transport: a pattern that holds true for various correlated materials as well as $\text{FeTe}_{0.55}\text{Se}_{0.45}$.

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Materials with layered structure commonly show a pronounced anisotropy of the electronic response. One of the most prominent examples is offered by the high- T_c cuprates revealing metal-like response along the CuO_2 planes and nearly insulating behavior along the c axis. Infrared spectroscopy is well suited for studying the electronic anisotropy of layered compounds. Extensive optical studies on the cuprates have revealed the incoherent nature of the c axis charge dynamics in the underdoped regime and the emergence of the c axis coherence in the optimally-doped or overdoped regimes [1].

Iron-based superconductors also have layered crystal structure composed of square planar sheets of Fe atoms [2]. Band calculations predicted that all the families of Fe-based superconductors share common cylinderlike Fermi surfaces, implying pronounced anisotropy of the electronic properties [3–6]. In order to achieve complete understanding of the electronic properties of Fe-based superconductors, it is imperative to experimentally investigate the anisotropy. The layered cuprates as well as other layered selenides may be regarded as particularly relevant reference materials to compare and contrast various aspects of the interlayer response. Up to now, optical studies on the Fe-based superconductors have been mostly focused on the ab plane responses [7–16] and much less is known about the c axis properties. Existing studies on the c axis optical properties are limited to undoped AFe_2As_2 ($A=\text{Ba}$, Sr , and Eu) [17, 18] and one single superconductor $\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2$ [19].

In this Letter, we analyze anisotropic electronic properties of $\text{FeTe}_{0.55}\text{Se}_{0.45}$. We find that the normal-state electronic response along the c axis is incoherent at all temperatures. The c axis optical conductivity $\sigma_c(\omega)$ does not show a well-defined Drude peak; the featureless incoherent conductivity is further suppressed at low temperature. We find that the incoherent c axis response of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ is related to the strong dissipation in the ab plane transport: a pattern that appears to be generic for a broad variety of correlated electron systems. Our results demonstrate the close similarity between the normal-state electronic responses of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ and mildly underdoped cuprates.

High-quality single crystals were grown by a unidirectional solidification method with a nominal composition of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ [20]. The bulk superconductivity at $T_c=14$ K in this sample was confirmed by the diamagnetic response in the magnetic susceptibility and the clear gap opening in $\sigma_{ab}(\omega)$ [16]. The ac surface with the size of $1\text{ mm}\times 4.5\text{ mm}$ was mechanically polished. The reflectance was measured between 25 and 30000 cm^{-1} for light polarized along the c axis using *in-situ* overcoating technique [21]. The reflectance is a complex quantity consisting of an amplitude and a phase, $\hat{r} = \sqrt{R}e^{i\theta}$. Only the amplitude R was measured in the experiment and the complex optical conductivity was determined using Kramers-Kronig analysis [22]. We found that the optical conductivity above 25 cm^{-1} was insensitive to the choice of low-frequency extrapolations of the reflectance.

Figure 1 displays c axis reflectance $R_c(\omega)$ at different temperatures. We also plot the ab plane reflectance $R_{ab}(\omega)$ for comparison [16]. At 295 K, the spectral shape of $R_c(\omega)$ is similar to that of $R_{ab}(\omega)$. $R_c(\omega)$ rises toward lower frequency implying the existence of an electronic contribution. However, the level of $R_c(\omega)$ is lower than that of $R_{ab}(\omega)$ by about 15%. In addition, the temperature dependence of $R_c(\omega)$ in the normal state is quite different from that of $R_{ab}(\omega)$. As the temperature decreases, $R_{ab}(\omega)$ at low and high frequencies are observed to increase and decrease, respectively. On the contrary, $R_c(\omega)$ decreases with decreasing temperature at all frequencies. The disparate temperature dependence as well as the suppressed value of $R_c(\omega)$ suggest that the normal-state charge dynamics along the c axis could be qualitatively different from that within the ab plane.

More insights on the anisotropic electronic response can be gained from the analysis of the conductivity spectra. Figures 2(a) and 2(b) show the real parts of the c axis and ab plane optical conductivities ($\sigma_c(\omega)$ and $\sigma_{ab}(\omega)$), respectively. The ab plane response is metal-like as implied in the behavior of $\sigma_{ab}(\omega)$. At 295 K, $\sigma_{ab}(\omega)$ is rather flat and shows weak metallic up-turn below 100 cm^{-1} . As the temperature decreases, the Drude-like behavior becomes evident in our data [16]. In contrast, $\sigma_c(\omega)$ shows no Drude peak at any temperature. Moreover, $\sigma_c(\omega)$ becomes suppressed as temperature

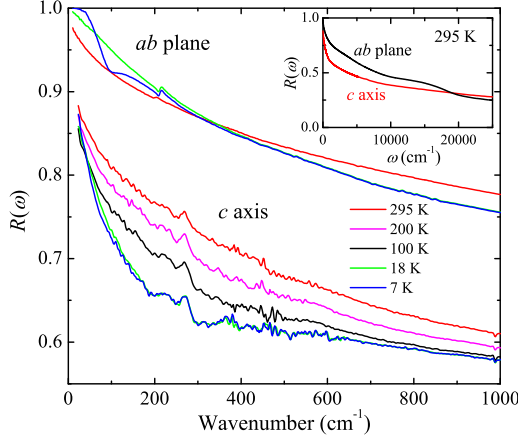


FIG. 1. (color online). Temperature-dependent c axis reflectance $R_c(\omega)$ of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ with $T_c=14$ K. The ab plane reflectance $R_{ab}(\omega)$ at 295 K, 18 K, and 6 K are also plotted for comparison [16]. The inset shows $R_{ab}(\omega)$ and $R_c(\omega)$ over broad frequency up to 25000 cm^{-1} .

is lowered and the spectral weight is transferred to higher energy. We will discuss the energy scale of the spectral weight transfer below. At 18 K, $\sigma_c(\omega)$ becomes almost frequency independent. The value of $\sigma_c(\omega)$ in the zero-frequency limit $\sigma_c(\omega \rightarrow 0)$ is about $160 \Omega^{-1}\text{cm}^{-1}$ at 18 K, which is smaller than $\sigma_{ab}(\omega \rightarrow 0)$ by the factor of about 25. All these observations indicate that the normal-state c axis electronic response of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ is incoherent.

The normal-state $\sigma_c(\omega)$ of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ bears significant similarities to that of the underdoped cuprates. In the underdoped cuprates, $\sigma_c(\omega)$ in the far-infrared region is also frequency independent and shows continuous depression with decreasing temperature, which has been attributed to the formation of pseudogap [1, 24, 25]. In addition, the absolute value of $\sigma_c(\omega \rightarrow 0)$ just above T_c is about $160 \Omega^{-1}\text{cm}^{-1}$ for $\text{FeTe}_{0.55}\text{Se}_{0.45}$ and this value lies between those of $\text{YBa}_2\text{Cu}_3\text{O}_{6.90}$ ($\sim 90 \Omega^{-1}\text{cm}^{-1}$) and $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ ($\sim 200 \Omega^{-1}\text{cm}^{-1}$) [25].

Apart from similarities in the form of $\sigma_c(\omega)$, the temperature dependence of interlayer spectral weight in $\text{FeTe}_{0.55}\text{Se}_{0.45}$ parallels the behavior registered in the underdoped cuprates [1]. In both classes of materials, the spectral weight is removed from low frequencies and is shifted to higher energy. In the cuprate literature this effect is usually referred as the c axis pseudogap reflecting strongly suppressed but finite conductivity at low temperatures [1, 25]. An immediate consequence of the pseudogap formation (or the spectral weight transfer to high frequency) for the c axis transport is the semiconducting temperature dependence of resistivity. Figure 2(a) shows that it is the spectral weight transfer that is responsible for the semiconducting trend of the interlayer resistivity of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ mirroring the behavior of the underdoped cuprates.

We stress that the spectral weight removed from far-infrared

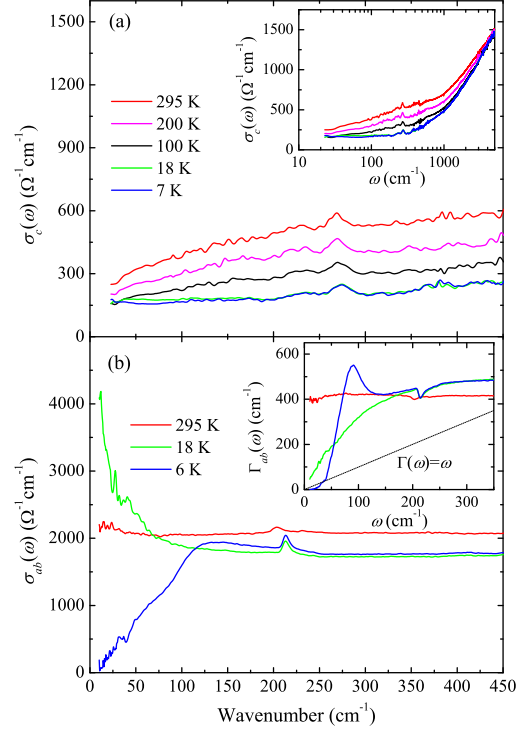


FIG. 2. (color online) (a) Temperature-dependent c axis optical conductivity $\sigma_c(\omega)$. The peak at about 268 cm^{-1} is due to the infrared-active phonon mode [23]. $\sigma_c(\omega)$ in the far-infrared region becomes clearly depressed with decreasing temperature and the suppressed spectral weight seems not to be recovered by 5000 cm^{-1} as shown in the inset. (b) Temperature-dependent ab plane optical conductivity $\sigma_{ab}(\omega)$. The inset shows the frequency-dependent ab plane scattering rate $\Gamma_{ab}(\omega)$ (Ref. 16). The dotted line corresponds $\Gamma(\omega)=\omega$

conductivity in $\text{FeTe}_{0.55}\text{Se}_{0.45}$ (as well as in the underdoped cuprates) is transferred to *higher* energies and not to the region below the low-frequency cutoff of our data. The latter effect would have resulted in drastic increases of $R_c(\omega)$ and the c axis dc conductivity, which is not supported either by our own reflectance or resistivity data (of $\text{FeTe}_{0.6}\text{Se}_{0.4}$ compound [26]). The oscillator strength sum rule implies a conservation of the global spectral weight. However, data in the inset of Fig. 2(a) show that the weight removed from far-infrared region is most likely not recovered even at 5000 cm^{-1} . The large energy scale of the spectral weight redistribution in $\sigma_c(\omega)$ of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ is in stark contrast to the expectations of the Fermi-liquid theory of metals and is generally regarded as a spectroscopic hallmark of strong electronic correlation [27].

It is worth noting that the behavior of $\sigma_c(\omega)$ in other layered selenides 2H-NbSe_2 and 2H-TaSe_2 are quite different from those in $\text{FeTe}_{0.55}\text{Se}_{0.45}$. In 2H-NbSe_2 , $\sigma_c(\omega)$ shows a Drude-like response at all temperatures [28]. In $\sigma_c(\omega)$ of 2H-TaSe_2 , there occurs a spectral weight transfer from high to low frequencies due to a charge-density wave (CDW) transition [29]. Above the CDW transition, $\sigma_c(\omega)$ of 2H-TaSe_2 is flat. At low

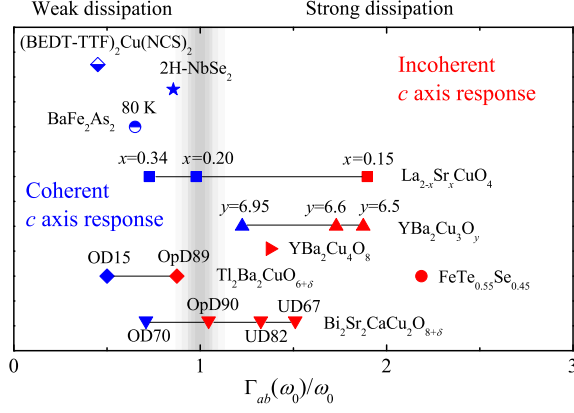


FIG. 3. (color online) ab plane scattering rate $\Gamma_{ab}(\omega_0)$ divided by ω_0 . ω_0 is the frequency that accounts for the 30% of the intraband spectral weight. The grey bar denotes the region close to $\Gamma_{ab}(\omega_0) \sim \omega_0$. The left and right sides of this region represent the weak and strong dissipation regimes, respectively. The majority of the materials with strong ab plane dissipation exhibit incoherent c axis response (red symbols). The latter is defined by the suppression of the low-energy spectral weight in $\sigma_c(\omega)$ at low temperature concomitant with $d\rho_c/dT < 0$. The majority of the materials with weak ab plane dissipation reveal coherent c axis behavior (blue symbols): the enhancement of the low-energy spectral weight in $\sigma_c(\omega)$ and $d\rho_c/dT > 0$. Data points: $\text{FeTe}_{0.55}\text{Se}_{0.45}$ (Ref. 16), BaFe_2As_2 (Ref. 39 and 40), 2H-NbSe_2 (Ref. 28), $(\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$ (Refs. 30 and 38), cuprates (Refs. 34–37, 42–45). (UD: underdoped, OpD: optimally-doped, OD: overdoped. The number denotes T_c of each compound.)

temperature, the spectral weight lost by the CDW gap opening is shifted to a narrow Drude-like peak indicating coherent c axis response [29].

The incoherent interlayer electrodynamics of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ should be contrasted to the coherent interlayer responses of SrFe_2As_2 and $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ compounds. In their $\sigma_c(\omega)$, the Drude response was clearly observed at low temperature [18, 19]. In addition, the conductivity anisotropy $\sigma_{ab}(\omega \rightarrow 0)/\sigma_c(\omega \rightarrow 0)$ of these compounds is only about 3–4 [18, 19], which is much smaller than that of $\text{FeTe}_{0.55}\text{Se}_{0.45}$. The coherent c axis transport was also observed in various layered materials with even larger conductivity anisotropy, such as $(\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$, Sr_2RuO_4 , and the overdoped cuprates [1, 30, 31]. These examples indicate that the layered structure or even strong conductivity anisotropy does not necessarily result in the incoherent c axis transport.

The degree of coherence in the c axis transport may be related to the strength of the scattering within conducting ab plane [32, 33]. The strength of ab plane scattering $\Gamma_{ab}(\omega)$ can be obtained from extended Drude analysis on the ab plane optical spectra [1]. The inset of Fig. 2(b) shows $\Gamma_{ab}(\omega)$ of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ [16]. The dashed line represents $\Gamma_{ab}(\omega) = \omega$. The region below this line corresponds to a Landau-Fermi-liquid regime, where the quasiparticles are well defined, i.e., $\Gamma_{ab}(\omega) < \omega$. As can be seen clearly, the magnitude of $\Gamma_{ab}(\omega)$

of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ is larger than the frequency ($\Gamma_{ab}(\omega) > \omega$) implying that the ab plane conduction is strongly dissipative. Data presented in Fig. 3 that we will discuss next show that the connection between strong dissipation in the ab plane response and incoherence in the interlayer transport is not unique to $\text{FeTe}_{0.55}\text{Se}_{0.45}$ but is in fact observed in a large variety of materials.

In order to elaborate on interdependence between trends in the ab plane conductivity and the degree of interlayer coherence, we plot in Fig. 3 $\Gamma_{ab}(\omega_0)$ of various layered compounds divided by ω_0 . Thus the shaded box represents the region close to $\Gamma_{ab}(\omega_0) \sim \omega_0$. We stress that an ambiguity with the choice of ω_0 does not make an impact on the inferences made out of Fig. 3. Especially for the cuprates, the value of ω_0 falls into the regime where $\Gamma_{ab}(\omega)$ shows linear frequency dependence [34–37]. The idea behind the horizontal axis is that the left side of the diagram represents the region of weak dissipation ($\Gamma_{ab}(\omega) < \omega$) whereas on the right of the grey bar dissipation is becoming progressively stronger. The materials showing coherent c axis response (blue symbols) including $(\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$, 2H-NbSe_2 , and BaFe_2As_2 all occupy the weak dissipation territory [28, 38–41]. In the same territory one also finds a number of moderately and heavily overdoped cuprates that are notorious for their coherent interlayer electrodynamics [1, 34–36, 42, 43]. As the doping level of the cuprates is decreased toward optimally and underdoped phases, the magnitude of $\Gamma_{ab}(\omega)$ increases continuously [34, 35, 37]. As soon as $\Gamma_{ab}(\omega)$ crosses over to strong dissipation regime on the right of the grey bar, the character of the interlayer transport is modified as well revealing marked incoherent trends [1, 44, 45]. These latter materials are labeled with red symbols and with one exception they all are located in the strong dissipation region of Fig. 3 [34–37, 46].

We emphasize that the strong dissipation in the ab plane transport has little to do with disorder. At least in several systems in the strong dissipation zone, disorder can be marginalized to enable quantum oscillations which requires exceptionally long mean free paths [47–49]. Instead, strong dissipation in infrared frequencies is a well known signature of substantial electronic correlations [1]. Specifically, the ab plane response of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ is governed by many body effects leading to pronounced frequency and temperature dependences of $\Gamma_{ab}(\omega)$ shown in the inset of Fig. 2(b). Moreover, giant absolute values of $\Gamma_{ab}(\omega)$ of $\text{FeTe}_{0.55}\text{Se}_{0.45}$ indicate that the many body effects are particularly strong in this compound. Our inference of the pivotal role of strong correlations in $\text{FeTe}_{0.55}\text{Se}_{0.45}$ is further supported by an observation of a large energy scale associated with the spectral weight transfer in the interlayer conductivity as discussed above.

Now we discuss the optical response in the superconducting state. The onset of superconductivity is identified by the change in $R_c(\omega)$ across T_c . Upon entering the superconducting state, $R_c(\omega)$ becomes larger (smaller) than $R_c(\omega)$ just above T_c below 37 cm^{-1} (between 37 and 200 cm^{-1}) as shown in Fig. 1. For s -wave superconductor, the reflectance becomes unity below the gap energy. Although $R_c(\omega)$ of

FeTe_{0.55}Se_{0.45} shows steep increase, it does not reach unity down to low-frequency cutoff of our data. Accordingly, while $\sigma_c(\omega)$ below T_c shows weak depression below 200 cm⁻¹, it does not exhibit a clear gaplike structure.

It is interesting to note that the c axis optical spectra of Ba_{0.67}K_{0.33}Fe₂As₂ also experienced little change across T_c [19]. In $\sigma_c(\omega)$ of Ba_{0.67}K_{0.33}Fe₂As₂, the gap opening was not observed but a large Drude-like response remained, which was attributed to the nodes in the superconducting gap [19]. Recent heat transport measurements on Ba(Fe_{1-x}Co_x)₂As₂ indeed revealed the appearance of the accidental nodes in the Fermi surfaces that contribute strongly to the c axis conduction as x moves away from the concentration of maximal T_c [50].

Previous studies on Fe(Te,Se) system indicated that the superconducting order parameter should be nodeless [51–54]. At this point, we defer further discussion of the c axis optical response in the superconducting state. In order to obtain the concrete information on the gap energy and the superfluid density in the c direction, the experiment in tera-hertz frequency region is highly desired.

In conclusion, we have demonstrated that the normal-state c axis electronic response of FeTe_{0.55}Se_{0.45} is incoherent and its behavior is reminiscent of the pseudogap formation in the underdoped cuprates. The incoherent c axis response of FeTe_{0.55}Se_{0.45} is in sharp contrast to the coherent c axis behavior of Ba_{1-x}K_xFe₂As₂. We show that the significant difference between the c axis responses of the two families of Fe-based superconductors correlates with the variation in the magnitude of the ab plane scattering rate. These findings indicate that among the Fe-based superconductors Fe(Te,Se) system is an exceptional case in which the strong electronic correlation governs the electronic structure [55–57].

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