

## Incoherent $c$ -Axis Interplane Response of the Iron Chalcogenide $\text{FeTe}_{0.55}\text{Se}_{0.45}$ Superconductor from Infrared Spectroscopy

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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 to 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  $\text{CuO}_2$  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 [2]. Band calculations predicted that all the families of Fe-based superconductors share common cylinder-like 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  $\text{AFe}_2\text{As}_2$  ( $A = \text{Ba}, \text{Sr}, \text{and Eu}$ ) [17,18] and  $\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2$  [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 ( $T$ ). 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  $T$ . 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 was confirmed by the diamagnetic response 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  $c$ -axis reflectance was measured between 25 and 30 000  $\text{cm}^{-1}$  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].

Figure 1 displays  $c$ -axis reflectance  $R_c(\omega)$  at different  $T$ . 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  $T$  dependence of  $R_c(\omega)$  in the normal state is quite different from that of  $R_{ab}(\omega)$ . As  $T$  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  $T$  at all frequencies. The disparate  $T$  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. At 295 K,  $\sigma_{ab}(\omega)$  is rather flat and shows weak metallic up-turn below 100  $\text{cm}^{-1}$ . As  $T$  decreases, the Drude-like behavior becomes evident [16]. In contrast,  $\sigma_c(\omega)$  shows no Drude

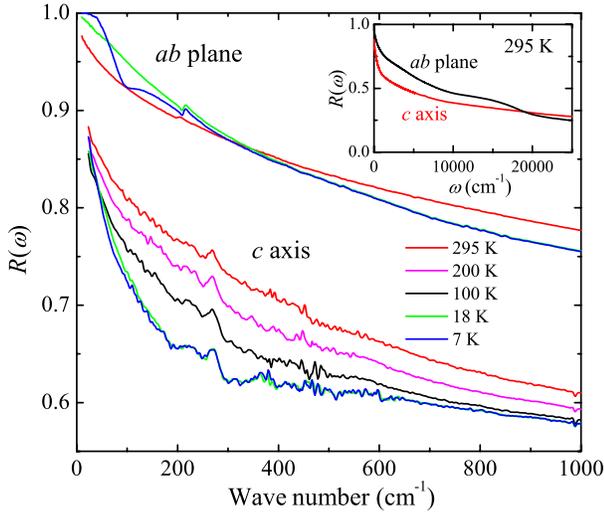


FIG. 1 (color online).  $T$ -dependent  $c$ -axis reflectance  $R_c(\omega)$  of  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  with  $T_c = 14$  K. The  $ab$ -plane reflectance  $R_{ab}(\omega)$  at 295, 18, and 6 K is also plotted for comparison [16]. The inset shows  $R_{ab}(\omega)$  and  $R_c(\omega)$  over broad frequency up to  $25\,000\text{ cm}^{-1}$ .

peak at any  $T$ . Moreover,  $\sigma_c(\omega)$  becomes suppressed as  $T$  is lowered and the spectral weight (SW) is transferred to higher energy. We will discuss the energy scale of the SW 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  $160\text{ }\Omega^{-1}\text{ cm}^{-1}$  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  $T$  [1,23,24]. In addition, the absolute value of  $\sigma_c(\omega \rightarrow 0)$  of  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  just above  $T_c$  lies between those of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.90}$  ( $\sim 90\text{ }\Omega^{-1}\text{ cm}^{-1}$ ) and  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$  ( $\sim 200\text{ }\Omega^{-1}\text{ cm}^{-1}$ ) [24].

Apart from similarities in the form of  $\sigma_c(\omega)$ , the  $T$  dependence of interlayer SW in  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  parallels the behavior registered in the underdoped cuprates [1]. In both classes of materials, the SW is removed from low frequencies and is shifted to higher energy. In the cuprate literature this effect is usually referred to as the  $c$ -axis pseudogap reflecting strongly suppressed but finite conductivity at low  $T$  [1,24]. An immediate consequence of the pseudogap formation (or the SW transfer to high frequency) for the  $c$ -axis transport is the semiconducting  $T$  dependence of resistivity. Figure 2(a) shows that it is the SW transfer that is responsible for the semiconducting trend of the interlayer resistivity  $\rho_c$  of  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  mirroring the behavior of the underdoped cuprates.

We stress that the SW removed from far-infrared conductivity in  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  is transferred to *higher* energies and not to the region below the low-frequency cutoff of our

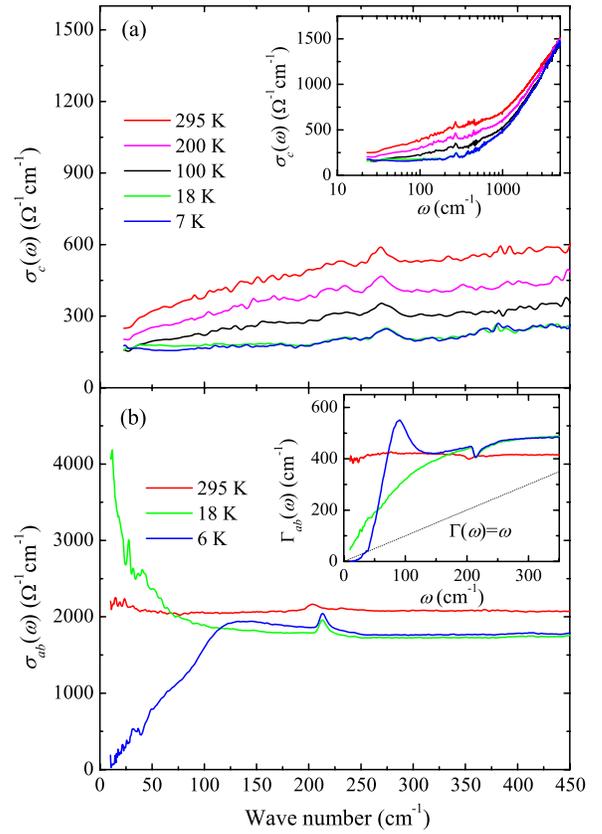


FIG. 2 (color online). (a)  $T$ -dependent  $c$ -axis optical conductivity  $\sigma_c(\omega)$ . The peak at about  $268\text{ cm}^{-1}$  is due to the infrared-active phonon mode [57].  $\sigma_c(\omega)$  in the far-infrared region becomes clearly depressed with decreasing  $T$ , and the suppressed SW seems not to be recovered by  $5000\text{ cm}^{-1}$ , as shown in the inset. (b)  $T$ -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 to  $\Gamma(\omega) = \omega$ .

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 [25]). The oscillator strength sum rule implies a conservation of the global SW. 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  $5000\text{ cm}^{-1}$ . The large energy scale of the SW 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 [26].

It is worth noting that the behavior of  $\sigma_c(\omega)$  in other layered selenides  $2H\text{-NbSe}_2$  and  $2H\text{-TaSe}_2$  is quite different from that in  $\text{FeTe}_{0.55}\text{Se}_{0.45}$ . In  $2H\text{-NbSe}_2$ ,  $\sigma_c(\omega)$  shows a Drude-like response at all  $T$  [27]. In  $\sigma_c(\omega)$  of  $2H\text{-TaSe}_2$ , there occurs a SW transfer from high to low frequencies due to a charge-density wave (CDW) transition [28]. Above the CDW transition,  $\sigma_c(\omega)$  of  $2H\text{-TaSe}_2$  is flat. At low  $T$ , the SW lost by the CDW gap opening is

shifted to a narrow Drude-like peak indicating coherent  $c$ -axis response [28].

The incoherent interlayer electrodynamics of  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  should be contrasted to the coherent interlayer responses of  $\text{SrFe}_2\text{As}_2$  and  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ . In their  $\sigma_c(\omega)$ , the Drude response was clearly observed at low  $T$  [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  $(\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$ ,  $\text{Sr}_2\text{RuO}_4$ , and the overdoped cuprates [1,29,30]. 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 the conducting  $ab$  plane [31,32]. The inset of Fig. 2(b) shows  $ab$ -plane scattering rate  $\Gamma_{ab}(\omega)$  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. The data presented in Fig. 3, which 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 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  $\omega_0$ . 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  $\omega_0$  does not make an impact on the inferences made out of Fig. 3. Especially for the cuprates, the value of  $\omega_0$  falls into the regime where  $\Gamma_{ab}(\omega)$  shows linear frequency dependence [33–36]. 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 gray bar dissipation becomes progressively stronger. The materials showing coherent  $c$ -axis response (blue symbols), including  $(\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$ ,  $2H\text{-NbSe}_2$ , and  $\text{BaFe}_2\text{As}_2$ , all occupy the weak dissipation territory [27,37–40]. In the same territory one also finds a number of moderately and heavily overdoped cuprates that are notorious for their coherent interlayer electrodynamics [1,33–35,41,42]. As the doping level of the cuprates is decreased toward optimally doped and underdoped phases, the magnitude of  $\Gamma_{ab}(\omega)$  increases continuously [33,34,36]. As soon as  $\Gamma_{ab}(\omega)$  crosses over to the strong dissipation regime, the character of the interlayer transport is modified as well, revealing marked incoherent trends [1,43,44]. These latter materials are labeled with red

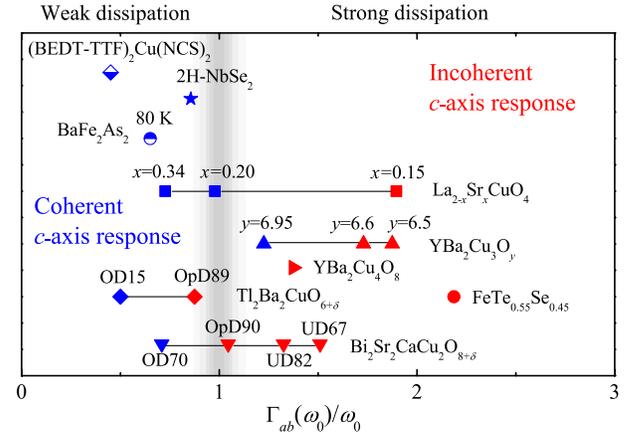


FIG. 3 (color online).  $ab$ -plane scattering rate  $\Gamma_{ab}(\omega_0)$  divided by  $\omega_0$ .  $\omega_0$  is the frequency that accounts for 30% of the intraband SW. The gray bar denotes the region close to  $\Gamma_{ab}(\omega_0) \sim \omega_0$ . The left and right sides of this region represent 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 SW in  $\sigma_c(\omega)$  at low  $T$  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 SW in  $\sigma_c(\omega)$  and  $d\rho_c/dT > 0$ . Data points:  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  (Ref. [16]),  $\text{BaFe}_2\text{As}_2$  (Refs. [38,39]),  $2H\text{-NbSe}_2$  (Ref. [27]),  $(\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$  (Refs. [29,37]), cuprates (Refs. [33–36,41–44]). (UD, underdoped; OpD, optimally doped; OD, overdoped. The number denotes  $T_c$  of each compound.)

symbols and with one exception they all are located in the strong dissipation region of Fig. 3 [33–36,45].

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 [46–48]. 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  $T$  dependences of  $\Gamma_{ab}(\omega)$ . 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 SW 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  $37\text{ cm}^{-1}$  (between  $37$  and  $200\text{ cm}^{-1}$ ) as shown in Fig. 1. For  $s$ -wave superconductor, the reflectance becomes unity below the gap energy. Although  $R_c(\omega)$  of  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  shows steep increase, it does not reach unity down to the low-frequency

cutoff of our data. Accordingly, while  $\sigma_c(\omega)$  below  $T_c$  shows weak depression below  $200\text{ cm}^{-1}$ , it does not exhibit a clear gaplike structure.

It is interesting to note that the  $c$ -axis optical spectra of  $\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2$  also experienced little change across  $T_c$  [19]. In  $\sigma_c(\omega)$  of  $\text{Ba}_{0.67}\text{K}_{0.33}\text{Fe}_2\text{As}_2$ , 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  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  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  $T_c$  [49].

Previous studies on the  $\text{Fe}(\text{Te},\text{Se})$  system indicated that the superconducting order parameter should be nodeless [50–53]. 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 the terahertz frequency region is highly desired.

In conclusion, we have demonstrated that the normal-state  $c$ -axis electronic response of  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  is incoherent and its behavior is reminiscent of the pseudogap formation in the underdoped cuprates. The incoherent  $c$ -axis response of  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  is in sharp contrast to the coherent  $c$ -axis behavior of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ . We show that the significant difference between the  $c$ -axis responses of the two families of Fe-based superconductors correlates to the variation in the magnitude of the  $ab$ -plane scattering rate. These findings indicate that among the Fe-based superconductors, the  $\text{Fe}(\text{Te},\text{Se})$  system is an exceptional case in which the strong electronic correlation governs the electronic structure [54–56].

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