

## Electronic correlations and unusual superconducting response in the optical properties of the iron chalcogenide $\text{FeTe}_{0.55}\text{Se}_{0.45}$

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The in-plane complex optical properties of the iron-chalcogenide superconductor  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  have been determined above and below the critical temperature  $T_c=14$  K. At room temperature the conductivity is described by a weakly interacting Fermi liquid; however, below 100 K the scattering rate develops a frequency dependence in the terahertz region, signaling the increasingly correlated nature of this material. We estimate the dc conductivity  $\sigma_{\text{dc}}(T \geq T_c) \approx 3500 \pm 400 \text{ } \Omega^{-1} \text{ cm}^{-1}$  and the superfluid density  $\rho_{s0} \approx 9 \pm 1 \times 10^6 \text{ cm}^{-2}$ , which places this material close to the scaling line  $\rho_{s0}/8 \approx 8.1\sigma_{\text{dc}}T_c$  for a BCS dirty-limit superconductor. Below  $T_c$  the optical conductivity reveals two gap features at  $\Delta_{1,2} \approx 2.5$  and 5.1 meV.

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The surprising discovery of superconductivity in the iron-arsenic  $\text{LaFeAsO}_{1-x}\text{F}_x$  (1111) pnictide compound has prompted an intense investigation of this class of materials.<sup>1,2</sup> The critical temperature  $T_c$  may be increased above 50 K through rare-earth substitutions.<sup>3</sup> While the mechanism for superconductivity in many metals and alloys is mediated by lattice vibrations,<sup>4</sup> the high values for  $T_c$  and the strong interplay between the magnetism and the lattice suggest that the superconductivity in this class of materials is not phonon mediated.<sup>5</sup> In addition to searching for higher values of  $T_c$  in the 1111 family of materials, considerable effort has been made looking for superconductivity in other structurally simpler Fe-based systems. In metallic  $\text{BaFe}_2\text{As}_2$  the application of pressure yields  $T_c \approx 29$  K while Co and Ni doping yields  $T_c \approx 23$  K at ambient pressure.<sup>6-8</sup> Superconductivity has also been observed in the As-free iron-chalcogenide  $\text{FeSe}$  compound with  $T_c=8$  K, which increases to  $T_c=27$  K with the application of pressure.<sup>9,10</sup> By introducing Te, the critical temperature in  $\text{FeTe}_{1-x}\text{Se}_x$  at ambient pressure reaches a maximum  $T_c=14$  K for  $x=0.45$ . Despite these structural differences, the band structure of these materials is similar, with a minimal description consisting of an electron band ( $\beta$ ) at the  $M$  point and a hole band ( $\alpha$ ) at the center of the Brillouin zone.<sup>11</sup> There have been a number of studies of the  $\text{Fe}_{1+x}\text{Te}$  and  $\text{FeTe}_{1-x}\text{Se}_x$  materials, including transport,<sup>12-15</sup> tunneling<sup>16</sup> and angle-resolved photoemission,<sup>17-20</sup> with particular emphasis placed on the magnetic properties.<sup>20-25</sup> While the optical properties of the superconducting iron-pnictide materials have been investigated in some detail,<sup>26-32</sup> in comparison the iron-chalcogenide materials remain relatively unexplored.<sup>13</sup>

In this work we examine the in-plane complex optical properties of superconducting  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  above and below  $T_c$ . Over much of the normal state the material is a weakly interacting Fermi liquid and the transport is Drude-like. However, close to  $T_c$  the Drude picture breaks down and the scattering rate adopts a strong frequency dependence, signaling the increasingly correlated nature of this material.<sup>19,33</sup> The onset of superconductivity is clearly observed in the optical properties below  $T_c$ , and the optical conductivity suggests that in addition to a prominent gap feature at  $\approx 5.1$  meV, a second gap opens at  $\approx 2.5$  meV.

Single crystals with good cleavage planes (001) were grown by a unidirectional solidification method with a nominal composition of  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  and a critical temperature determined by magnetic susceptibility of  $T_c=14$  K with a transition width of  $\approx 1$  K. The reflectance from the cleaved surface of a mm-sized single crystal has been measured at a near-normal angle of incidence for several temperatures above and below  $T_c$  over a wide frequency range ( $\sim 2$  meV to 4 eV) for light polarized in the  $a$ - $b$  planes using an *in situ* overcoating technique.<sup>34</sup> The reflectance in the terahertz and far-infrared region ( $1 \text{ THz}=33.4 \text{ cm}^{-1}$ ) is shown in Fig. 1 (the extended unit cell of  $\text{FeTe}$  is shown in the inset). At room temperature, the reflectance displays the typical metallic form in the Hagen-Rubens regime  $R \propto 1 - \sqrt{\omega}$ ; however, just above  $T_c$  the reflectance develops a striking linear-frequency dependence. Below  $T_c$  the formation of a superconducting condensate and the opening of a gap in the spectrum of excitations is clearly visible. The reflectance is a complex quantity consisting of an amplitude and a phase,  $\tilde{r} = \sqrt{R}e^{i\theta}$ ; because only the amplitude  $R = \tilde{r}\tilde{r}^*$  is measured it is

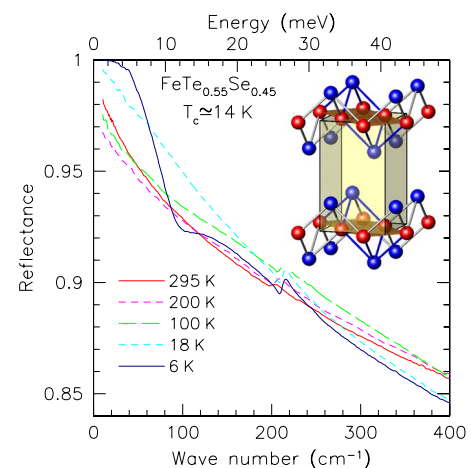


FIG. 1. (Color online) The reflectance of  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  in the far-infrared region for light polarized in the Fe-Te planes at several temperatures above and below  $T_c$ . Inset: the extended unit cell of  $\text{FeTe}$  in the tetragonal  $P4/nmm$  space group showing the tetrahedrally coordinated Te above and below the Fe planes.

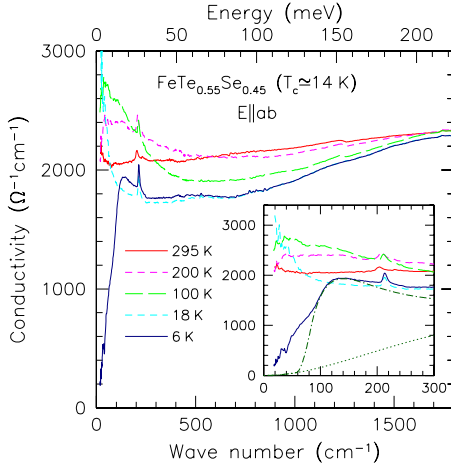


FIG. 2. (Color online) The real part of the in-plane optical conductivity for  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  at several temperatures above and below  $T_c$  in the infrared region. Inset: the conductivity in the far-infrared region compared with a model calculation for a single isotropic gap ( $\Delta_0=4.5$  meV,  $1/\tau=4\Delta_0$ ) superimposed on the Lorentzian contribution.

often not intuitively obvious what changes in the reflectance imply. Consequently, the complex optical properties have been determined from a Kramers-Kronig analysis of the reflectance.<sup>35</sup>

The temperature dependence of the real part of the optical conductivity is shown in Fig. 2 in the infrared region; the far-infrared region is shown in the inset. At room temperature, the conductivity is relatively flat and structureless, except for a sharp feature associated with the infrared-active  $E_u$  mode at  $204$   $\text{cm}^{-1}$  which is due to the in-plane displacements of the Fe-Te(Se) atoms<sup>36</sup> (slightly higher than the  $E_u$  mode observed at  $187$   $\text{cm}^{-1}$  in our examination of  $\text{Fe}_{1.03}\text{Te}$ ). As the temperature is lowered there is a redistribution of the spectral weight [defined here as the weight under the conductivity curve over a given interval,  $\int_{\omega_0}^{\omega_c} \sigma_1(\omega, T) d\omega$ ] from high to low frequency. This response is not unusual for a metallic system where the scattering rate decreases with temperature. The optical conductivity is described by a Drude-Lorentz model for the dielectric function  $\tilde{\epsilon} = \epsilon_1 + i\epsilon_2$

$$\tilde{\epsilon}(\omega) = \epsilon_\infty - \frac{\omega_{p,D}^2}{\omega^2 + i\omega/\tau_D} + \sum_j \frac{\Omega_j^2}{\omega_j^2 - \omega^2 - i\omega\gamma_j}, \quad (1)$$

where  $\epsilon_\infty$  is the real part of the dielectric function at high frequency,  $\omega_{p,D}^2 = 4\pi n e^2 / m^*$  and  $1/\tau_D$  are the plasma frequency and scattering rate for the delocalized (Drude) carriers, respectively;  $\omega_j$ ,  $\gamma_j$ , and  $\Omega_j$  are the position, width, and strength of the  $j$ th vibration or excitation. The complex conductivity is  $\tilde{\sigma}(\omega) = \sigma_1 + i\sigma_2 = -i\omega[\tilde{\epsilon}(\omega) - \epsilon_\infty]/4\pi$ .

The optical conductivity may be reproduced quite well using this approach at 295, 200 and 100 K, with fitted values of  $\omega_{p,D} = 7200$   $\text{cm}^{-1}$  and  $1/\tau_D = 414, 363,$  and  $317$   $\text{cm}^{-1}$ , respectively ( $\pm 5\%$ ). To fit the midinfrared component, Lorentzian oscillators at the somewhat arbitrary positions of  $650$  and  $3000$   $\text{cm}^{-1}$  have been introduced, allowing the free-carrier component to be fit using a single Drude expression,

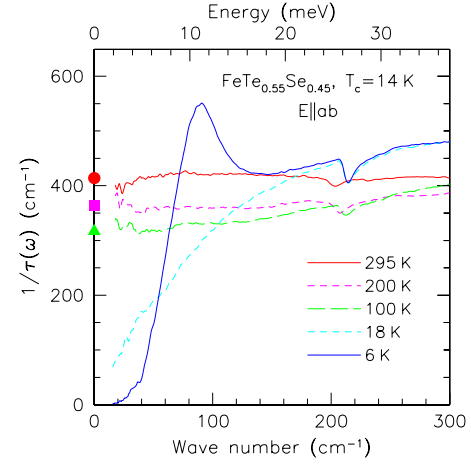


FIG. 3. (Color online) The in-plane frequency-dependent scattering rate of  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  for several temperatures above and below  $T_c$  in the far-infrared region. The values for  $1/\tau_D$  are shown at 295 (●), 200 (■), and 100 K (▲), respectively, where the scattering rate displays little temperature dependence. For  $T \geq T_c$  (18 K) at low frequency  $1/\tau(\omega) \propto \omega$  while for  $T < T_c$  large changes in the scattering rate are observed in response to the formation of superconducting gap(s).

as opposed to the two-Drude response that has recently been applied to some of the pnictide materials.<sup>32</sup> While this approach works well over most of the normal state, it fails for the optical conductivity just above  $T_c$  at 18 K, where the low-frequency component is not Drude-like. To address this problem, we consider the extended-Drude model in which the scattering rate takes on a frequency dependence. The experimentally determined scattering rate is<sup>37</sup>

$$\frac{1}{\tau(\omega)} = \frac{\omega_p^2}{4\pi} \text{Re} \left[ \frac{1}{\tilde{\sigma}(\omega)} \right]. \quad (2)$$

In this instance we set  $\omega_p = \omega_{p,D}$  and  $\epsilon_\infty = 4$  [although the choice of  $\epsilon_\infty$  has little effect upon  $1/\tau(\omega)$  in the far-infrared region]; the temperature dependence of  $1/\tau(\omega)$  is shown in Fig. 3 above and below  $T_c$ . At 295, 200, and 100 K the scattering rate displays little frequency dependence, and moreover  $1/\tau(\omega \rightarrow 0) \approx 1/\tau_D$ . This self-consistent behavior indicates that within this temperature range, the transport may be described as a weakly interacting Fermi liquid (Drude model). However, just above  $T_c$  at 18 K the scattering rate develops a linear-frequency dependence  $\approx 200$   $\text{cm}^{-1}$ , suggesting strong electronic correlations.<sup>19</sup> This may be due in part to magnetic correlations<sup>21</sup> that arise from the suppression of the magnetic transition in  $\text{Fe}_{1+\delta}\text{Te}$  at  $T_N \approx 70$  K in response to Se substitution.<sup>14</sup> We note that similar behavior of the scattering rate is observed in optimally doped cuprates where the electronic correlations may have a similar origin.<sup>38</sup> Dramatic changes are also observed in  $1/\tau(\omega)$  below  $T_c$  where the scattering rate is suppressed at low frequencies, but increases rapidly and overshoots the normal-state (18 K) value at about  $60$   $\text{cm}^{-1}$ , finally merging with the normal-state curve at about  $200$   $\text{cm}^{-1}$ ; this behavior is in rough agreement with a recently proposed sum rule for the scattering rate.<sup>39</sup>

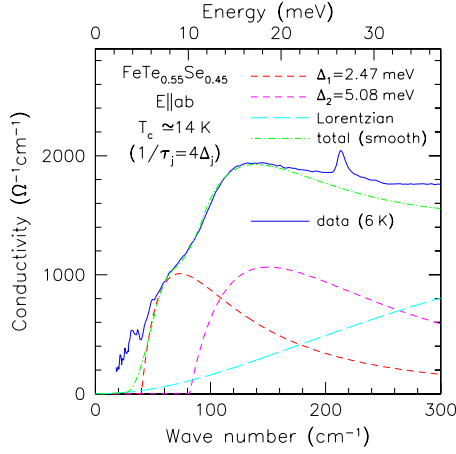


FIG. 4. (Color online) The in-plane optical conductivity of  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  shown at 6 K (solid line). The calculated optical conductivity with gaps of  $2\Delta_1 \approx 5$  meV and  $2\Delta_2 \approx 10.2$  meV for  $T \ll T_c$  (short-dashed lines) is superimposed on the contribution from the bound excitations in the mid-infrared (long-dashed line); the smoothed linear combination of the three curves (dot-dash line) is in good agreement with the measured data below  $200$   $\text{cm}^{-1}$ .

Returning to the optical conductivity in Fig. 2, below  $T_c$  there is a dramatic suppression of the low-frequency conductivity and a commensurate loss of spectral weight which is shown in more detail in the inset. The loss of spectral weight is associated with the formation of a superconducting condensate, whose strength may be calculated from the Ferrell-Glover-Tinkham sum rule:  $\int_0^{\omega_c} [\sigma_1(\omega, T \geq T_c) - \sigma_1(\omega, T \ll T_c)] d\omega = \omega_{p,S}^2/8$ . Here  $\omega_{p,S}^2 = 4\pi n_s e^2/m^*$  is the superconducting plasma frequency and the cut-off frequency  $\omega_c \approx 150$   $\text{cm}^{-1}$  is chosen so that the integral converges smoothly; the superfluid density is  $\rho_{s0} \equiv \omega_{p,S}^2$ . The sum rule yields  $\omega_{p,S} = 3000 \pm 200$   $\text{cm}^{-1}$ , indicating that less than one-fifth of the free carriers in the normal state have condensed ( $\omega_{p,S}^2/\omega_{p,D}^2 \leq 0.18$ ). The superfluid density can also be expressed as an effective penetration depth  $\lambda_0 = 5300 \pm 300$  Å, which is in good agreement with recent tunnel-diode measurement on  $\text{FeTe}_{0.63}\text{Se}_{0.37}$  (Ref. 40). From the estimate  $\sigma_{dc} \equiv \sigma_1(\omega \rightarrow 0) = 3500 \pm 400$   $\Omega^{-1} \text{cm}^{-1}$  for  $T \geq T_c$ , this compound is observed to fall on the general scaling line<sup>41</sup> for a BCS superconductor with the condition that  $1/\tau \geq 2\Delta$  (the “dirty limit”),  $\rho_{s0}/8 \approx 8.1\sigma_{dc}T_c$ .

The detailed optical conductivity below  $T_c$  at 6 K is shown in Fig. 4. In addition to the strong suppression of the conductivity below  $\sim 120$   $\text{cm}^{-1}$ , there is also a prominent shoulder at  $\sim 60$   $\text{cm}^{-1}$ . Below  $T_c$  the optical conductivity has been calculated using a Mattis-Bardeen formalism for the contribution from the gapped excitations,<sup>35,42</sup> as well as the low-frequency tail of the bound midinfrared excitations modeled by Lorentzian oscillators. The Mattis-Bardeen approach assumes that  $l \leq \xi_0$ , where the mean-free path  $l = v_F \tau$  ( $v_F$  is the Fermi velocity) and the coherence length is  $\xi_0 = \hbar v_F / \pi \Delta_0$  for an isotropic superconducting gap  $\Delta_0$ ; this may also be expressed as  $1/\tau \geq 2\Delta_0$ . This approach is motivated by the observation that less than one-fifth of the free carriers collapse into the condensate, a condition which indicates that these materials are not in the clean limit. Initially, only a

single isotropic gap  $\Delta_0 \approx 4.5$  meV was considered; however, this failed to accurately reproduce the residual conductivity observed at low frequency (inset in Fig. 2). To properly model the optical conductivity, two gaps at  $\Delta_1 \approx 2.5$  meV and  $\Delta_2 \approx 5.1$  meV are used. For the purposes of this calculation we have assumed a moderate amount of disorder scattering,  $1/\tau_j = 4\Delta_j$ . The optical conductivity for each of the gaps is shown in Fig. 4 for  $T=0$ ; the smoothed linear combination of the gaps and the Lorentzian tails is in good agreement with the experimental data. The observation of two gap features is consistent with a number of recent theoretical works that propose that  $s$ -wave gaps form on each band, possibly with a sign change between them ( $s^\pm$ ), in this model the gap on the electron band may be an extended  $s$  wave with nodes.<sup>43,44</sup> The strong reduction of the conductivity at low frequency for  $T \ll T_c$  suggests the absence of nodes. It is possible that disorder may lift the nodes, resulting in a nodeless extended  $s$ -wave gap.<sup>45,46</sup> The optical results provide estimates of the gap amplitudes but do not distinguish between  $s^\pm$  and extended  $s$  wave. The optical gaps  $2\Delta_j \approx 40$  and  $82$   $\text{cm}^{-1}$  are similar to or larger than the low-frequency scattering rate observed at 18 K,  $1/\tau(\omega \rightarrow 0) = 40$   $\text{cm}^{-1}$ . While this might seem to cast doubt on the validity of the Mattis-Bardeen approach, we note that the strong frequency dependence of the scattering rate at this temperature complicates matters. If we consider the value of the  $1/\tau(\omega)$  in the region of the optical gaps  $2\Delta_j$  where the scattering should be important, we find from Fig. 3 that the scattering rate is then larger than the gap amplitude

$$\frac{1/\tau_j(2\Delta_j)}{2\Delta_j} \approx 3,$$

which is actually larger than the ratio used in the calculation, indicating that the Mattis-Bardeen approach is correct. Finally, we note that while  $2\Delta_1/k_B T_c \approx 4$  is close to the value of 3.5 expected in the BCS weak-coupling limit,<sup>4</sup>  $2\Delta_2/k_B T_c \approx 8.4$  is significantly larger.

To summarize, the optical properties of  $\text{FeTe}_{0.55}\text{Se}_{0.45}$  ( $T_c = 14$  K) have been examined for light polarized in the Fe-Te(Se) planes above and below  $T_c$ . Well above  $T_c$  the transport may be described by a weakly interacting Fermi liquid (Drude model); however, this picture breaks down close to  $T_c$  when the scattering rate takes on a strong frequency dependence, similar to what is observed in the cuprate superconductors. Below  $T_c$ , less than one-fifth of the free carriers collapse into the condensate ( $\lambda_0 \approx 5300$  Å), indicating that this material is in the dirty limit, and indeed this material falls on the general scaling line predicted for a BCS dirty-limit superconductor. To successfully model the optical conductivity, two gaps of  $\Delta_1 \approx 2.5$  meV and  $\Delta_2 \approx 5.1$  meV are considered using a Mattis-Bardeen formalism (with moderate disorder scattering), suggesting either an  $s^\pm$  or a nodeless extended  $s$ -wave gap.

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