The discovery of high-temperature superconductivity in the ferropnictides has stimulated great interest in investigating their physical properties [1]. As in the cuprates, the superconducting state of the ferropnictides is located in the vicinity of a magnetically ordered state. The parent ferropnictide compounds exhibit stripe-type antiferromagnetic (AFM) spin-density-wave (SDW) order [2,3,4,5], while suppressing the AFM order by various means gives rise to the superconductivity [6,7,8,9]. It is generally believed that the proximity to magnetism plays a crucial role in establishing electronic pairing [10].

Numerous efforts have been made to elucidate the electronic structure changes across the SDW transition of the ternary ferropnictide $A$Fe$_2$As$_2$ ($A=$Ba, Sr, or Eu) compounds. In angle-resolved photoemission spectroscopy (ARPES) studies on BaFe$_2$As$_2$ and SrFe$_2$As$_2$, the electronic structure changes across the SDW transition were initially explained in terms of the exchange splitting [11,12], even though no uniform exchange splitting is expected in an antiferromagnet. Later ARPES studies, optical and quantum oscillation measurements as well as transport data indicated substantial, albeit incomplete, gapping of the Fermi surfaces due to the SDW gap opening [13,14,15,16,17,18]. In particular, an optical study by Hu et al. showed the appearance of two gap-like features (GLFs) in the SDW states of BaFe$_2$As$_2$ and SrFe$_2$As$_2$, reflecting the multiband nature of the ferropnictides [13]. However, Wu et al. reported that the optical conductivity spectra $\sigma(\omega)$ of EuFe$_2$As$_2$ showed only one GLF in the SDW state [14]. Hsieh et al., using polarization-dependent ARPES on SrFe$_2$As$_2$, reported two sets of Fermi surfaces, nested and non-nested, and concluded that only the nested bands exhibited a partial gap opening in the SDW state [18].

The small crystal field splitting and large bandwidth of the ferropnictide compounds make multiband effects crucial in the formation of the electronic structure both in the vicinity of the Fermi energy and at higher energies [19,20,21]. This complicates the interpretation of the electronic structure. Moreover, particularly in the 122 family, the large magnetic moments cause reconstruction of the electronic bands in the SDW state not only in the vicinity of the Fermi level (as a simplistic nesting picture would imply), but over the entire bandwidth. The multiband and multi-orbital nature of the electronic structure suggests an interesting interplay between the orthorhombic anisotropy of the magnetic order (even neglecting the small structural distortion) and the orbital character of the bands near the Fermi level. However, to date, few studies have addressed the effect of magnetism on the multiple electronic bands. In light of this, understanding the changes of the individual electronic bands due to magnetism, and the effects of these changes on spectroscopic quantities such as optical conductivity, is of great interest.

In this Letter, we investigate the evolution of the electronic structure of EuFe$_2$As$_2$ using optical spectroscopy and first-principles calculations. We observe a drastic decrease of the Drude response and the formation of two GLFs in the $\sigma(\omega)$ across the SDW transition. The GLFs exhibit disparate temperature ($T$) dependences, with one GLF magnitude and spectral weight (SW) in quantitative agreement with the BCS weak-coupling behavior, while the other is much less $T$-dependent. Based on first-principles calculations, we identify the two corresponding classes of optical transitions: one is related to the transition within the spin-minority bands and is therefore sensitive to the long-range magnetic order, and the other between the spin-majority and the spin-minority (on the site with the opposite magnetization) states. The second transition is mostly defined by the local magnetic moment, and less sensitive to the long-range ordering.

High-quality EuFe$_2$As$_2$ single crystals were grown using the flux technique. The resistivity of EuFe$_2$As$_2$ showed an anomaly at $T_N=190$ K, which is associated with the paramagnetic (PM)-to-AFM transition. Near-normal incident re-
reflectance spectra $R(\omega)$ were measured between 5 meV and 6.5 eV, using a liquid-He cooled cryostat. The in-plane $\sigma(\omega)$ were obtained using the Kramers-Kronig transformation of $R(\omega)$. In the calculations, the experimental tetragonal and orthorhombic lattice constants were used for the PM and AFM states, respectively [22].

Figure 1 displays the $T$-dependent $\sigma(\omega)$ of EuFe$_2$As$_2$. As $T$ decreases across the SDW transition, the broad Drude-like response becomes strongly suppressed and develops into a gap-like structure. These $T$-dependent $\sigma(\omega)$ of EuFe$_2$As$_2$ reveal a couple of intriguing features. First, in the SDW state, there is a very sharp Drude response in the frequency region below about 0.02 eV. Its small SW and even smaller width, are consistent with a large reduction of the free carrier concentration, due to the opening of a partial gap at the Fermi surface, and an even stronger reduction of the scattering rate, as noticed before [23, 24, 25]. Second, unlike the weight transfer for $\sigma(\omega)$ of single-band density wave systems, two GLFs (peaks $\alpha$ and $\beta$) appear below the SDW transition that receive the SW lost due to the gapping of the Fermi surface. This clearly reflects the multiband nature of the ferropnictides.

The evolution of these two peaks with $T$ provides insights into the characteristic responses of the individual bands to magnetism. To obtain such information, we analyzed the $\sigma(\omega)$ using the Drude-Lorentz oscillator model. Overall, we used two Drude ($D$) terms and three Lorentz oscillators for peak $\alpha$, peak $\beta$, and a high energy interband transition. The second Drude peak is not necessary for fitting below $T_N$, but if we include it, it comes out with a smaller weight and a very large width, and, consequently, with a large uncertainty. According to the Hall data [17], the hole mobility in the SDW state is drastically smaller than the electron mobility. Thus, we tentatively attribute the first, narrow Drude peak to electrons, and the second, broad and with the SW corresponding to 1.5-2 times smaller Fermi velocity, to holes. Note also that above $T_N$ the intensity of the low-energy peak $\alpha$ is zero within the fitting uncertainty.

Next we turn to the peak $\alpha$, which is well-defined below $T_N$. We find $2\Delta_\alpha/k_BT_N \sim 3.4$, where $\Delta_\alpha$ is taken equal to the peak position $\omega_\alpha$ at 20 K. Note that this value agrees well with the mean-field BCS ratio. Also, the $T$ dependences of $\omega_\alpha$ and $SW_\alpha$ follow the BCS formula quite well, as shown with dashed lines in Figs. 2(a) and 2(b). Conversely, for the peak $\beta$, the $2\omega_\beta(20\text{ K})/k_BT_N$ value is about 10.9, and there are hardly any $T$ dependence in $\omega_\beta$ and $SW_\beta$. Above $T_N$, the SW$\beta$ abruptly decreases, but its position is about the same as that below $T_N$. Importantly, the total SW is reasonably well conserved across the transition, with the SW removed from the Drude peaks in the SDW phase, and transferred into the GLF $\alpha$ and $\beta$. The overall picture suggests that the peak $\alpha$ is associated with partial gapping that evolves in a mean-field manner, while the bands related to the peak $\beta$ are almost fully gapped and the evolution is a more local phenomenon and involves wholesale alteration of the band structure.

This can be compared with Hsieh et al.’s suggestion that two different kinds of bands, nested and non-nested bands, exist in SrFe$_2$As$_2$ [18]. They argued that the nested bands experience partial gapping, but that the non-nested bands were assumed to have no substantial change upon entering the SDW state. However, the SDW gap these authors observed in their ARPES experiment was rather small and not consistent with results from our (and others) optical studies (and also too small for the observed SDW magnitude of $\sim 1\mu_B$). As shown in Fig. 2(b), most of the carriers present above $T_N$ at the Fermi level disappear below $T_N$, and 90% of the Drude SW
is shifted into the interband peaks $\alpha$ and $\beta$. The discrepancy with the ARPES results most likely is caused by the sensitivity of ARPES (but not optics) to the surface states, which likely have a smaller magnetic moment than in the bulk.

To directly identify the electronic structure changes in EuFe$_2$As$_2$ due to the SDW transition, we performed all-electron density-functional calculations using the local density approximation, as implemented in the WIEN2K code. To simulate the actual small experimental magnetic moment of $\sim 1 \mu_B$, we applied an artificial negative $U$ of $-0.03$ Ry, which reduced the calculated staggered magnetization from $1.65 \mu_B$ to $1.12 \mu_B$ (this procedure has no physical justification and is only employed to emulate the reduction of the ordered moment due to quantum fluctuations).

Figure 3(a) shows the band structure of the PM state in the energy region between $-1$ and $1$ eV, where the bands have predominantly Fe $d$ character. Near the zone center $\Gamma = (0, 0, 0)$, three hole bands cross the Fermi level $E_F$, while near the $N = (\pi, \pi, 0)$ point, two electron bands cross the $E_F$. As shown in Fig. 3(b), the band structure changes drastically upon entering the AFM state. In the AFM state, there are far fewer $E_F$ crossings, and as is evident from Fig. 3(b), these are almost entirely of $d_{yz}$ character, which is consistent with recent results from a laser-ARPES study [26]. This therefore reflects the gaping of the bands of this character lies approximately $0.055$ eV in the measured band-structure changes across the ordering transition.

Evidently, magnetism affects the band structure on the energy scale of several eV. A comparison of the Fe $d_{xz}$ spin-up and spin-down band character plots in Figs. 3(c) and 3(d) reveals that the spin-up bands of this character lie approximately $1$ eV higher than the spin-down bands, and this energy difference carries over to the bands with other orbital characters (not shown) as well. This energy difference can be quantitatively interpreted in terms of exchange splitting, based on the local Fe Hund coupling of $\sim 0.8$ eV and the calculated staggered magnetization of $1.12 \mu_B$. This exchange splitting, not the Hubbard $U$ or Fermi surface nesting, drives the main band-structure changes across the ordering transition.

Moreover, these band-structure changes and Hund coupling can be directly related to the features at about $0.2$ eV and $0.055$ eV in the measured $\sigma(\omega)$. Presented in Figs. 3(d) and 3(e) are bands with the Fe $d_{xy}$ spin-down character projection and the $d_{xy}$ projection for the same spin on an antiferromagnetically aligned adjacent Fe, respectively. The latter is identical by symmetry to the spin-up projection on the same Fe, therefore they are separated by roughly the exchange splitting.

We immediately notice a flat band in the former projection at approximately $-0.4$ eV and the latter at $0.2$ eV, suggesting the existence of an $0.6$ eV feature, which is clearly observed in the calculated $\sigma(\omega)$ (Fig. 3(g)). This feature results from transitions between the majority occupied Fe spin-down $d_{xz}$ orbital on one Fe atom, Fe-1, and the minority unoccupied spin-down $d_{xy}$ orbital on an adjacent Fe atom with the opposite spin, Fe-2. The parallel nature of the bands then results in a large contribution to the joint density of states (JDOS) at $0.6$ eV, which drives the larger gap. (There is of course a similar effect linking the occupied spin-up Fe-2 orbital state with the unoccupied spin-up Fe-1 orbital state.) As this interaction occurs in real space, it is sensitive to the local exchange splitting, and is therefore the harbinger of localized moments. Factoring in the band mass renormalization in the pnictides.

![Figure 3](color online). (a) Band structure of the PM tetragonal state. (b) Band structures of the AFM orthorhombic state. The contributions of the $xy$, $yz$, and $xz$ orbital states are depicted by the open squares, circles, and triangles. The size of the symbol indicates the fraction of the orbital character of bands. The symmetry points are $\Gamma = (0, 0, 0)$, $H = (2\pi, 0, 0)$, $N = (\pi, \pi, 0)$, and $P = (\pi/2, \pi/2, \pi/2)$ in the PM Brillouin zone for (a) and AFM Brillouin zone for (b)-(f). Note that the AFM Brillouin zone is rotated by $\pi/4$ relative to that of the tetragonal state and is downfolded. (c) The contribution of Fe-1 $d_{xz}$ spin-up character. (d) The contribution of Fe-1 $d_{yz}$ spin-down character. (e) The Fe-2 $d_{xy}$ spin-down character. Here the Fe-2 atom is adjacent to Fe-1 in the direction of the ordering vector. (f) The Fe-1 $d_{yz}$ spin-up character. (g) Calculated in-plane $\sigma(\omega)$ for the PM and AFM states.
of approximately 3, as measured through ARPES \[27\], one concludes that this is the peak $\beta$ at about 0.2 eV in the measured $\sigma(\omega)$. Supporting this assignment is the fact that this peak is experimentally observed to have little $T$ dependence in the SDW state and to change abruptly its weight (number of instant AFM bonds), but not its position (local moment magnitude) across the magnetic transition.

The origin of the peak $\alpha$ at about 0.055 eV in the measured $\sigma(\omega)$ is somewhat more complex. The $d_{xz}$ spin-down band in Fig. 3(f) is the one that provided metallic carriers at the $E_F$. In the plot of the $d_{xz}$ spin-up character (Fig. 3(c)) one sees a band parallel to this band, but displaced down by approximately 0.25 eV. These parallel bands provide a distinct contribution to JDOS and result in a feature at 0.25 eV in the calculated $\sigma(\omega)$. Unlike the larger gap, however, this gap originates from transitions between two same-spin orbitals within the same-spin sublattice, and is therefore insensitive to the local exchange splitting (we have verified that reducing the local moment in the calculations suppresses the frequency of the higher-energy peak, but affects little the lower-energy feature). This gap is thus an itinerant-type SDW one, despite substantial band-structure effects. Considering the mass renormalization factor, we conclude that this smaller peak is the peak $\alpha$ seen at 0.055 eV in the experiment, which obeys markedly mean-field BCS-type behavior.

The magnetism in the ferropnictides is thus neither fully local nor fully itinerant, but has elements of both, as discussed in Ref. \[28\]. Our experiment shows that this duality translates into two distinctively different sets of optical features. One is a low-energy feature that monitors the true long range order manifesting itself in restructuring the Fermi surface, and thereby gapping out the Drude SW. This spatial ordering is comparable to the conventional spin-Peierls SDW, except it develops on the background of the large local moments, which produce substantial band-structure changes. The higher energy feature is less sensitive to the long-range spatial order, but rather to the local exchange splitting. Intermediate-range ordering is sufficient to slow down the local fluctuations enough to make the local splitting visible in optics.

In summary, we have investigated the electronic structure of EuFe$_2$As$_2$ using optical spectroscopy and first-principles calculations. We observe drastic changes of the optical conductivity across the magnetic transition, which can be described as transfer of the spectral weight into two gap-like features. These are very sensitive to the symmetry breaking associated with the stripe antiferromagnetism. The higher-energy feature is associated with the optical transitions between two Fe atoms of the opposite spin. The scale is set by the local exchange splitting, therefore this transition shows little temperature dependence below $T_N$ and survives (with a reduced spectral weight) above $T_N$. The low-energy feature is associated with the transitions within the spin-minority states of the same-spin Fe’s. These are less sensitive to the magnitude of exchange splitting, but very sensitive to the long range order controlling the gapping of the Fermi surface. Thermally this feature behaves as a weak-coupling Fermi surface. This makes it a promising candidate for experiments probing the spin density wave.

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