Evidence of the influence of magnetism on pseudogap states in the high resolution spectra of $\text{EuFe}_2\text{As}_2$

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Employing state of the art high resolution photoemission spectroscopy, we studied the temperature evolution of the electronic structure of $\text{EuFe}_2\text{As}_2$, an unique pnictide, where antiferromagnetism of Eu layer survives within the superconducting phase due to ‘FeAs’ layers achieved via substitution and/or pressure. High energy and angle resolution helped to reveal pseudogap-quasiparticle features having primarily As 4p character and spin density wave transition induced band folding in the electronic structure. A weakly dispersing feature of dominant As 4p character is discovered around 80 meV that becomes weaker in intensity below 20 K manifesting influence of antiferromagnetic order on conduction electrons. These results provide an evidence of a link between the pseudogap states and magnetism that could be revealed employing high resolutions.

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Interplay of magnetism and superconductivity is a long standing issue in material research and believed to play the key role in deriving the unconventional superconductivity [1]. Discovery of superconductivity in Fe-based compounds [2, 3] via suppression of spin density wave (SDW) state, further reinforces this connection and are believed to be good candidates for such studies. These superconductors are usually classified into the following categories based on their composition and structure, namely ‘1111’, ‘122’, ‘111’ and ‘11’ families [4]. ‘FeAs’ layers play the key role in deriving the materials properties of these systems. Among them, ‘122’ class of materials, $\text{AFe}_2\text{As}_2$ ($\text{A} = \text{Ba, Sr, Ca and Eu}$) have drawn significant attention as high quality single crystals can be grown easily in the whole composition range and one can easily tailor the properties of the materials via insertion of varied elements in the layers intermediate to the ‘FeAs’ layers.

$\text{EuFe}_2\text{As}_2$ is a special compound in this class as the spacer atom, Eu has large moment leading to two magnetic transitions, (i) SDW-type antiferromagnetic transition around 190 K due to Fe-moments and (ii) antiferromagnetic (AFM) ordering of the Eu-moments at 20 K. The versatility of $\text{EuFe}_2\text{As}_2$ is that superconductivity (SC) can be achieved by substituting foreign elements in any of the sites. For example, substitution of K [5, 6] and Na [7] at Eu sites leads to superconductivity with transition temperature, $T_C$, as high as 30 K and 34.7 K, respectively. Fe site substitution by Co results in superconductivity [8, 9]. Ni substitution at Fe sites is multifaceted; while Ni-doped $\text{CaFe}_2\text{As}_2$ is superconducting [10], Eu moments align ferromagnetically in Ni-doped $\text{EuFe}_2\text{As}_2$ at low temperatures and no superconductivity was observed [11]. The phase diagram of P-doping at As sites is also interesting [12, 13]; a narrow low-doped regime exhibit coexistence of superconductivity and AFM order, while a somewhat higher doping leads to ferromagnetic order of Eu-moments and superconductivity disappears. Application of pressure leads to superconductivity even in the parent compound [14]. Evidently, $\text{EuFe}_2\text{As}_2$ provides an ideal playground to achieve insight on the interplay of superconductivity and magnetism. Here, we studied the evolution of the electronic structure as a function of temperature and photon energy employing high resolution photoemission spectroscopy. Experimental results provide signature of pseudogap, features related to magnetic order and evidence of a link between them.

High quality single crystalline, $\text{EuFe}_2\text{As}_2$ was grown using Sn-flux [15]. The photoemission measurements were carried out on cleaved sample surface using monochromatic photon sources and Gammadata Scienta, R4000 WAL analyzer at a base pressure better than 3×10$^{-11}$ torr. The temperature variation down to 10 K on the sample was achieved using an open cycle liquid helium cryostat, LT-3M from Advanced Research Systems, USA.

We probed the evolution of the electronic structure of $\text{EuFe}_2\text{As}_2$ as a function of temperature and photon energy employing high energy resolution in the angle integrated photoemission (AIPES) and 10 meV for angle-resolved photoemission spectroscopy (ARPES) with angle resolution set to 0.3°.

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photoemission cross-section of Eu 4f states \(\sigma(Eu4f)\) compared to all other contributions in the valence band [10]. Thus, the features in 1-2 eV energy range can be attributed primarily to the Eu 4f photoemission [17] and the intensities near the Fermi level, \(\epsilon_F\), are due to Fe 3d - As 4p hybridized bands. The change in lineshape and peak positions reflect the sensitivity of the spectra to the photoemission cross section of constituent electronic states.

Change in temperature across the AFM transition of the Eu moments leads to a decrease in intensity near \(\epsilon_F\) - this is evident in the subtracted spectra (30 K - 10 K) shown in the lower panel of Fig. 1(a) exhibiting three distinct features, A, B and C. The feature, A appearing around 50 meV (see inset for clarity) has similar intensity in both He I and He II spectra suggesting mixed character of these states. The feature, B at about 200 meV is strong in He I spectrum and absent in He II spectrum. At He I and He II energies, the values of \(\sigma(Fe3d)\) are 4.833 and 8.751, and that of \(\sigma(As4p)\) are 3.856 and 0.2949, respectively [10]. Clearly, As 4p is significantly stronger in He I energies, while Fe 3d contributions are stronger in He II energies. This indicates dominance of As 4p character in feature B. The feature, C appearing around 0.5 eV is weak and observed in the He I spectrum indicating its As 4p character. The intensity changes beyond 1 eV is visible primarily in He II spectrum presenting the changes in Eu 4f intensities. Clearly, the electronic states near \(\epsilon_F\) possess finite As 4p contributions due to Eu 4f-As 4p and Fe 3d-As 4p hybridizations, and are highly sensitive to the AFM ordering. Similar intensity of the feature A indicates participation of Fe 3d states in the AFM ordering.

In Fig. 1(b) and 1(c), we investigate the spectral density of states (SDOS) obtained by symmetrization \((I(\epsilon - \epsilon_F) + I(\epsilon_F - \epsilon))\) of the experimental data, which provides a good estimation of the intensity at \(\epsilon_F\). The SDOS at both the photon energies (He I and He II) exhibit significant reduction of intensity at \(\epsilon_F\) across SDW transition. The high energy resolution enabled to reveal an additional gap at \(\epsilon_F\) across the AFM transition at 20 K and a quasiparticle peak appears around 20 meV in both He I and He II spectra as evidenced in the rescaled spectra in Fig. 1(c). While AFM transition can lead to a gap/depletion at \(\epsilon_F\) [18, 13], the spectral evolution exhibiting a pseudogap-quasiparticle peak structure in Fig. 1(c) resembles the widely known spectral evolution in various unconventional superconductors exhibiting precursor effects [20, 21] noting the fact that EuFe\(_2\)As\(_2\) is highly susceptible to show superconductivity as it exhibits superconductivity under application of pressure and/or suitable substitution at any of the three sites.

Various angle resolved photoemission (ARPES) studies of EuFe\(_2\)As\(_2\) showed that the temperature change across the SDW transition (\(\sim 190\) K) leads to the formation of tiny hole and electron pockets near \((\pi, \pi)\) [22]. The overlap of the back-folded and non-folded bands in the SDW phase forms small ‘droplets’ in the Fermi surface [22]. Both, the bulk measurements [17] and ARPES studies [22] suggested that Eu and FeAs sublattices are nearly decoupled as no noticeable change was observed across the antiferromagnetic ordering. However, the high energy resolution employed in this study helped to reveal strong evidence of the influence of antiferromagnetic transition on the conduction electrons responsible for the electronic properties. In order to probe this in more detail, we study the energy bands around the \(\Gamma\) point along (0,0)-\((\pi, \pi)\) direction as a function of temperature and photon energy.

In Fig. 2(a), 2(b) and 2(c), we show the He II ARPES data at 300 K, 30 K and 10 K, respectively - the lower panels show their second derivative. The band dispersions at 300 K (paramagnetic phase) are consistent
FIG. 2: (color online) He II ARPES data along (0,0)-(π,π)-direction at (a) 300 K, (b) 30 K and (c) 10 K. The lower panel show the second derivative of the same data. Lines are guide to the eye. The corresponding EDCs at (d) 300 K, (e) 30 K and (f) 10 K. (g) EDC at Γ at different temperatures. (h) MDC at 30 K. The points in (h) and (i) represent the peak positions obtained by fitting MDCs shown in (h).

with the band structure results [22]. At 30 K, the region around (π,π) modifies significantly - the dispersion around (π,π) becomes very similar to that at Γ point indicating band folding due to the supercell formation in the SDW phase evidencing signature of a hole pocket around (π,π). We calculated SDOS by dividing the experimental spectra by the resolution broadened Fermi-Dirac distribution function. The corresponding energy distribution curves (EDCs) are shown in Fig. 2(d), 2(e) and 2(f) for the spectra at 300 K, 30 K and 10 K, respectively. Three distinct bands denoted by α, β and γ are discernible in the spectra. While the α band folds back around 150 meV binding energy at Γ point, β and γ bands cross $\epsilon_F$ forming hole pockets around Γ point [22,23].

The energy band dispersions of β and γ bands were derived via fitting the momentum distribution curves (MDC). The peak positions are shown in Fig. 2(h) and 2(i). The estimated values of $k_B T_\text{F}$ for the β and γ bands are about 0.1 Å$^{-1}$ and 0.2 Å$^{-1}$, respectively. The effective mass ($m^*/m_e = h^2/(\partial^2 \epsilon/\partial k^2)$) of the charge carrier is found to be 2.5 and 6.1 for the β and γ bands, respectively, consistent with the values estimated from quantum oscillation studies in similar systems [24]. The electronic states forming the γ band are found to be more correlated than others and play active role in the SDW transition. The Fermi velocities for the two bands are $6.5 \times 10^2$ and $4.5 \times 10^4$ m/sec, respectively. These values remain almost the same across the antiferromagnetic transition at 20 K.

The EDC around Γ point exhibit interesting evolution with temperature. At 300 K, EDC exhibits large intensity near $\epsilon_F$. The 30 K data, on the other hand, exhibits a dip at $\epsilon_F$, which becomes more prominent at 10 K consistent with the ARPES data. This is shown with better clarity in Fig. 2(g), where we show the EDCs at Γ point at various temperatures. The α band appearing around 150 meV remains almost unchanged in the whole temperature range studied. An additional feature (see the
observed at citations, respectively. The largest change in SDOS is weak dispersion suggests hybridization of these states shown in Figs. 4(c) and 4(d) for He I
K observed in He II feature, responding electronic states. In order to investigate the portion of the As 4
4+3 electronic states is enhanced significantly at He II and He II moments. The symmetrized Spectral DOS are similar systems, resemble well the experimental data. The δ bands possessing d_{z^2} character appear at higher binding energies. The α and β bands possess (d_{xz}, d_{yz}) and d_{xy} character, respectively and are less influenced by the magnetic transitions - a small shift (∼20 meV) of the β band is observed across the AFM transition at 20 K. Fig. 3(g) indicates (d_{xz}, d_{yz}) symmetry of the γ band, which is responsible for the SDW transition. This is understandable as the As layers appear above and below the Fe-layers and hence, the hybridization mediating interccrete coupling will be dominated by (d_{xz}, d_{yz}) electronic states.

Although the energy bands in the vicinity of ε_F possess dominant Fe 3d character, the contribution of As 4p states is significant due to hybridization among the corresponding electronic states. In order to investigate the feature, κ further, we compare the EDCs at 30 K and 10 K observed in He I and He II spectra in Fig. 4. While the photoemission intensities reduce with temperature in both the photon energies, the largest change is observed in the He I spectra. The relative photoemission cross section of the As 4p states is enhanced significantly at He I energy compared to that at He II energy [12]. Thus, the feature κ can be attributed primarily to As 4p states. Weak dispersion suggests hybridization of these states with the weakly dispersing Eu 4f states. This characterization is further justified by the fact that the intensity of the κ band reduces significantly with the decrease in temperature across the AFM transition temperature due to Eu 4f moments. The symmetrized Spectral DOS are shown in Figs. 4(c) and 4(d) for He I and He II excitations, respectively. The largest change in SDOS is observed at k-points, where the β and γ bands cross the Fermi level - the intensities at all other k-points are almost identical. The change in intensity is largest in the He I spectra indicating again large As 4p character of these electronic states.

In summary, we studied the temperature evolution of the electronic structure of EuFe_2As_2, an ideal compound to study the interplay between magnetism and superconductivity generally known to be two mutually exclusive phenomena. The signature of pseudogap-quasiparticle feature, typical of various superconducting materials is manifested in the high resolution data at low temperatures; these features possess large As 4p character. The angle resolved data demonstrate band folding due to the spin density wave transition. Distinct signature of an additional weakly dispersing feature is manifested in the low temperature data possessing large As 4p character that reduces in intensity across antiferromagnetic tran-
sition in the Eu layer. Evidently, the Eu 4f-As 4p and Fe 3d-As 4p hybridizations are significant and play major role in deriving the electronic properties. These results demonstrate evidence of a link of antiferromagnetism and pseudogap states that could be revealed due to the employment of high resolutions.

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