High-energy electronic interaction in the 3*d* band of high-temperature iron-based superconductors

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One of the most unique and robust experimental facts about iron-based superconductors is the renormalization of the electronic band dispersion by factor of 3 and more near the Fermi level. Obviously related to the electron pairing, this prominent deviation from the band theory lacks understanding. Experimentally studying the entire spectrum of the valence electrons in iron arsenides, we have found an unexpected depletion of the spectral weight in the middle of the iron-derived band, which is accompanied by a drastic increase of the scattering rate. At the same time, the measured arsenic-derived band exhibits very good agreement with theoretical calculations. We show that the low-energy Fermi velocity renormalization should be viewed as a part of the modification of the spectral function by a strong electronic interaction. Such an interaction with an energy scale of the whole *d* band appears to be a hallmark of many families of unconventional superconductors.

There is a fundamental problem in condensed matter physics: The Hamiltonian of any solid is extremely complicated due to the large amount of involved particles. It is actually not trivial that one-electron mean-field theory (MFT) gives astonishingly accurate predictions for the electronic properties of a vast majority of common materials [1–3]. On the other hand, the one-electron approach spectacularly fails for many compounds that are currently under the scrutiny of modern condensed matter physics, especially for those with potentially useful extraordinary properties [4,5]. One recent example is the class of iron-based high-temperature superconductors [6,7], where numerous experimental techniques have established that the distribution of the electronic states at the Fermi level is compressed three and more times as compared to the MFT predictions [8–13]. Since there are many candidates not accounted for by the MFT approximation, which could lead to the renormalization at low energies, and very little is known experimentally about the nature of the force capable of triple band squeezing, the problem remains unsolved. In this Rapid Communication we apply angle-resolved photoemis-

sion spectroscopy (ARPES) to study the electronic structure of iron-based superconductors covering a much larger interval of binding energies than it is usually done, and show that not only electronic states at the Fermi level are renormalized, but the whole structure of the iron 3*d* band is changed with respect to MFT by a strong interaction of an energy scale comparable to the total bandwidth.

In Fig. 1 we compare ARPES spectra recorded along the high-symmetry directions with the corresponding results of the local density approximation (LDA) band structure calculations for a NaFeAs compound. At the bottom of the valence band, between 2 and 5 eV binding energies (ω), the photoemission intensity closely follows the calculated dispersions which correspond to the *p* bands of arsenic. In this region there are well-defined dispersion features with moderate broadening and no appreciable renormalization. Near $\omega \approx 2$ eV, where the bottom of the iron 3*d* band is located, we observe the features with significantly larger scattering, but still a noticeable dispersion and energy position similar to the original nonrenormalized LDA bands. A comparison of the dispersions at even lower binding energies demonstrates that the experimental features change rapidly from smeared out and weakly defined to intense and well-discernible ones when going from 500 meV to 0. It is clearly seen that they are located much closer to the Fermi level than the calculations predict [compare Fig. 1(a) with Figs. 1(b) and 1(c) and Fig. 1(e) with Figs. 1(f) and 1(g), respectively]. In some spectra one is able to see a kink in the dispersion at about 500 meV [Figs. 1(c), 1(g) and 1(h)]. See the Supplemental Material [14]

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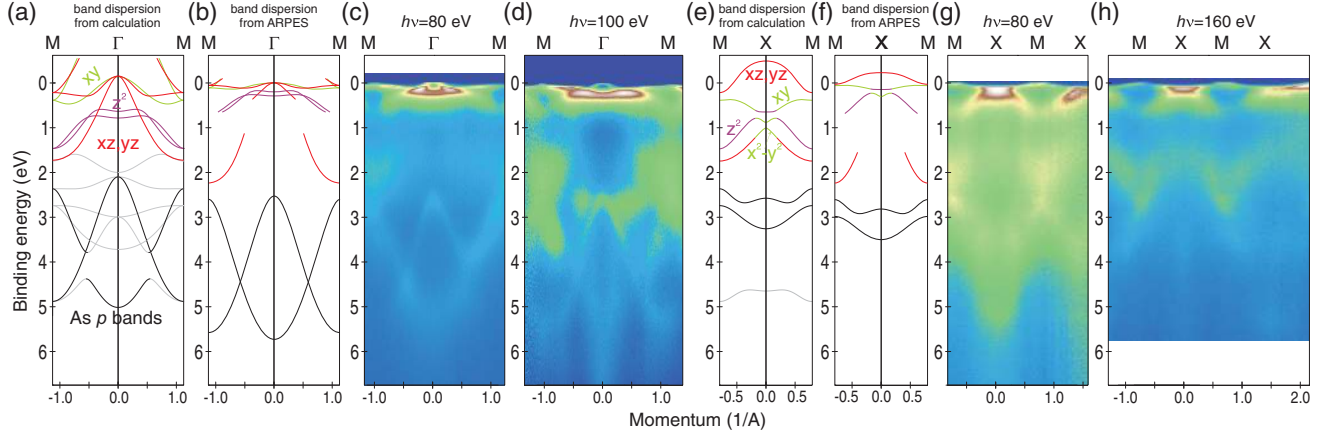
FIG. 1. Theoretically calculated and derived from ARPES band dispersion in NaFeAs. (a) Calculated band dispersion in the $M\Gamma M$ direction. Color coding of the electronic state orbital composition is indicated in the figure. Orbital polarization of the electronic states in this direction is very strong. (b) Contours of the band dispersion, extracted from an ARPES intensity distribution. Extraction of experimental dispersion curves relies on the data measured at different experimental conditions; typical spectra are shown in (c) and (d). (e)–(h) present the same information for the $M X M$ direction.

for a more detailed identification of the dispersive bands in the vicinity of the Fermi level; the basic result of such matching is that the experimental bands at low binding energies are the ones predicted by LDA, but renormalized by an average factor of 3.

Next, we try to understand the origin of the above-described anomalies and deviations from the one-electron picture. For a start, we employ a rather simple model where electrons of the original MFT bands interact with a hypothetical bosonic spectrum [15] according to the Eliashberg formalism. Within this approach, the self-energy, which encapsulates the many-body effects of electronic interactions, is defined by the following formulas, $\Sigma'(\omega) = \int_0^\infty \alpha^2 F(\Omega) \ln |\frac{\omega+\Omega}{\omega-\Omega}| d\Omega$, $\Sigma''(\omega) = -\pi \int_0^\omega \alpha^2 F(\Omega) d\Omega$, where $\Sigma(\omega) = \Sigma'(\omega) + i \Sigma''(\omega)$ is the self-energy, and $\alpha^2 F(\Omega)$ is the Eliashberg function.

In Fig. 2 we compare the calculated spectral function with the experimental energy-momentum distribution of the photoemission intensity. The bosonic spectrum $\alpha^2 F(\Omega)$ was assumed to be of a simplest single-peak shape. In the Supplemental Material [14] we present a convenient analytic form for $\alpha^2 F(\Omega)$, allowing for an explicit integration of the expressions for Σ' and Σ''. To make the comparison more transparent, the experimental conditions were chosen in such a way that the photoemission matrix elements highlight one of the bands, while all others are suppressed. The model captures many important features seen in the experimental spectra. First of all, these are the sharp and strongly renormalized dispersions close to the Fermi level. The fast increase of the electronic scattering rate with binding energy at a correct energy scale is reproduced, too—well-defined dispersions vanish below 0.5 eV. Finally, the smeared out spectral weight is also distributed around the contours of bare dispersion. This result clearly implies that combining the LDA calculations with the simple treatment of the self-energy satisfactorily reproduces the experimental spectral function of the whole valence band on an energy scale of up to 6 eV. Obviously, $\alpha^2 F(\Omega)$ does not necessarily represent here a particular external bosonic excitation—it could well be just a convenient representation

of purely electron-electron interactions, and it remains an open question whether the corresponding bosonic excitations can be singled out.
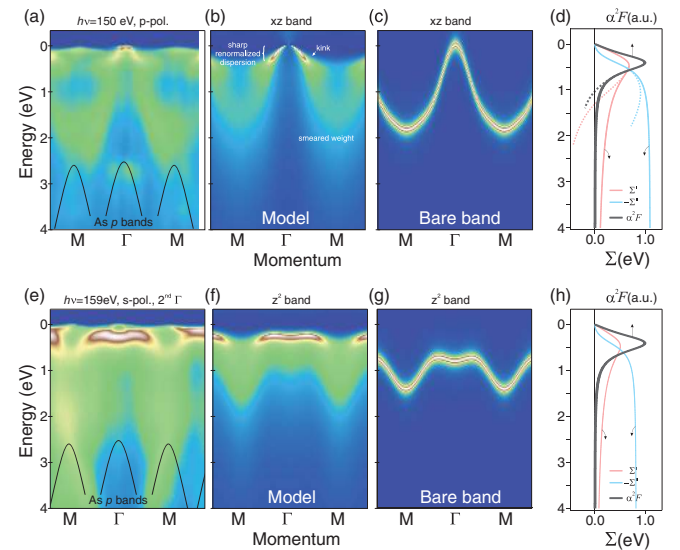

FIG. 2. Model for the spectral function based on the bare dispersion and electronic interaction with a hypothetical bosonic spectrum of single-peak form. Experimental conditions were chosen to highlight one of the bands and suppress the intensity from all others. (a) Experimental data, recorded at a photon energy of 150 eV with vertical light polarization. (b) Spectral function, obtained for the lower-lying xz/yz band and $\lambda = 2$. (c) Spectral function for the bare band. (d) Bosonic spectrum $\alpha^2 F(\omega)$ and self-energy $\Sigma(\omega)$ used in the model. Dashed lines schematically represent the situation where Σ' changes sign and the value of Σ'' decreases at high ω—this would further improve the agreement between the model and experiment at the bottom of the iron band. (e) Experimental data, recorded at a photon energy of 159 eV with horizontal light polarization in the second Brillouin zone. (f) Spectral function, obtained for the z^2 band and $\lambda = 1.6$. (g) Spectral function for the bare band. (h) Corresponding $\alpha^2 F(\omega)$ and $\Sigma(\omega)$.

(a) predominantly xz band

NaFeAs   LiFeAs   (Ba,Rb)Fe₂As₂   NdFeAs(O,F)   (La,Na)Fe₂As₂

80 eV, in-plane light polarization

(b) predominantly z² band

NaFeAs   (Ba,K)Fe₂As₂   (Ba,Na)Fe₂As₂   (Ba,Co)Fe₂As₂   (La,Na)Fe₂As₂
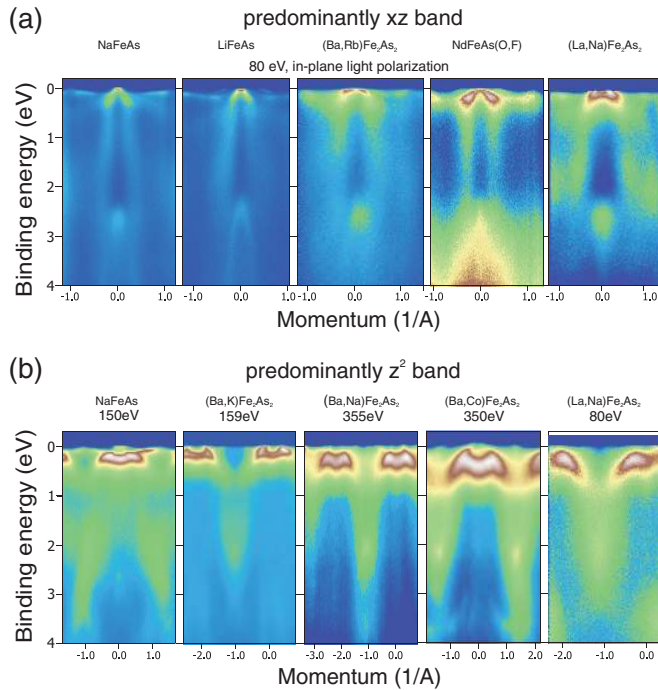150eV   159eV   355eV   350eV   80eV

FIG. 3. Experimental data taken from various iron-based superconductors, revealing common signatures of strong electron interactions in the spectral function. (a) Data recorded from NaFeAs, LiFeAs, (Ba,Rb)Fe₂As₂, NdFeAs(O,F), and (La,Na)Fe₂As₂. All spectra were taken along the ΓM direction, Brillouin zone diagonal, exclusively with the light of 80 eV energy and polarization lying in the ab plane. These experimental conditions highlight the spectral function related to the original xz band. Note the similarities in all important aspects for the spectra of different materials and compare to the corresponding model, (b) and (c). (b) Experimental data recorded from NaFeAs, (La,Na)Fe₂As₂, and hole- and electron-doped BaFe₂As₂ along the ΓM direction. Experimental conditions were tuned in order to highlight the spectral weight originating from the original z^2 band. The presented data reveal signatures of strong electron interactions and a similar spectral shape (fat letter M) is observed for all studied materials; also see (f) and (g).

In order to illustrate the universality of the high-energy anomaly for a large class of materials, we show ARPES spectra recorded from different iron arsenides of 111, 122, and 1111 crystalline structures with electron, hole, or no doping in Fig. 3. In Fig. 3(a) solely the energy-momentum cuts passing through the Γ point, recorded with 80 eV photons of vertical polarization, are presented. At these conditions the most contribution to the photoemission intensity comes from the xz band, while the matrix element for other bands is largely suppressed. The observed intensity distributions are very similar to each other and possess all signatures of strong electron interactions of a high-energy scale, discussed above [also see Figs. 2(a) and 2(b)]—most prominently, the region of intense well-defined dispersion near the Fermi level ends sharply, leaving only a broad spectral weight at higher binding energies. Figure 3(b) shows data recorded with experimental parameters highlighting the z^2 band. The spectral weight here also exhibits all the features, inherent to the interacting electronic systems. In particular, the intensity in the shape of the letter M centered at zero momentum is seen in all cases.

One may notice that the experimental spectral function slightly deviates from the model in the region between 1.5 and 2 eV: The contours of the smeared spectral weight distribution are somewhat below the original bare bands and the electronic scattering at the bottoms of these bands is somewhat lower [Figs. 2(a) and 2(b)]. Both these discrepancies can be removed by allowing a *decrease* of the scattering rate after it passes the maximum, which would cause a simultaneous *change of sign* of the real part of the self-energy [Fig. 2(d)]. Interestingly, the same effect is observed when one describes the spectra of cuprates, ruthenates, and vanadates using the self-energy formalism [16–18]. Within our model such behavior of the self-energy would necessarily imply that the bosonic spectrum $\alpha^2 F$ becomes *negative*, starting from a particular energy. The peculiarities of the experimental spectra, including a high-energy kink, are very reminiscent (both in shape and energy scale) of the features present in the spectral function, calculated for strongly correlated electronic systems [5,19–22]. Together, it means that while the intensity distribution corresponding to the Fe $3d$ band, as it appears in iron pnictides and chalcogenides, can be described by weak-coupling equations surprisingly adequately, at the same time it inherits some signatures peculiar to the strong-coupling approach.

The observation of high-energy anomalies in the experimental spectral function such as a kink in dispersion and a stripe of intensity depletion as well as the possibility to track intensity variations at the contours of original MFT bands allow us to determine the Eliashberg function rather precisely. What could be the physical nature of the introduced effective bosonic spectrum $\alpha^2 F$? A phononic origin can be ruled out, as the typical energies of phononic modes have more than an order of magnitude lower energy [23,24]. One of the obvious candidates would be the spin-fluctuation spectrum, as strong electron coupling to the spin resonance mode below T_c has been detected for many iron-based superconductors [25–27]. However, the spin-fluctuation spectrum has a maximum at about 200 meV, and does not extend as high as 500 meV [28,29]. Consequently, although both spin fluctuations and phonons certainly make contributions to the band renormalization at lower binding energies, they cannot be a source of the high-energy anomalies and band renormalization on the largest energy scale of the $3d$ band discussed here. Among other theoretically considered possibilities, the Coulomb interaction in the forms of (i) the well-known on-site repulsion U and (ii) recently proposed to be important in iron-based superconductors Hund's coupling J [30–32] are reasonable candidates for explanations of high-energy anomalies in the electronic spectrum. More theoretical and experimental work is obviously needed to understand the origin and details of the spectral anomalies introduced here.

It is instructive to recall that most of the electronic systems are "normal"—their spectra do not exhibit such strong anomalies and departures from MFT. We show ARPES spectra for a number of renowned materials in Fig. 4(a). The band dispersion can be traced down to a 5 eV binding energy and even deeper, and the agreement with MFT including the energy bandwidth is nearly perfect. A good intuitive quantity for an illustration of the electronic interaction strength is the scattering rate Σ''. A plot of the binding energy dependence $\Sigma''(\omega)$ for
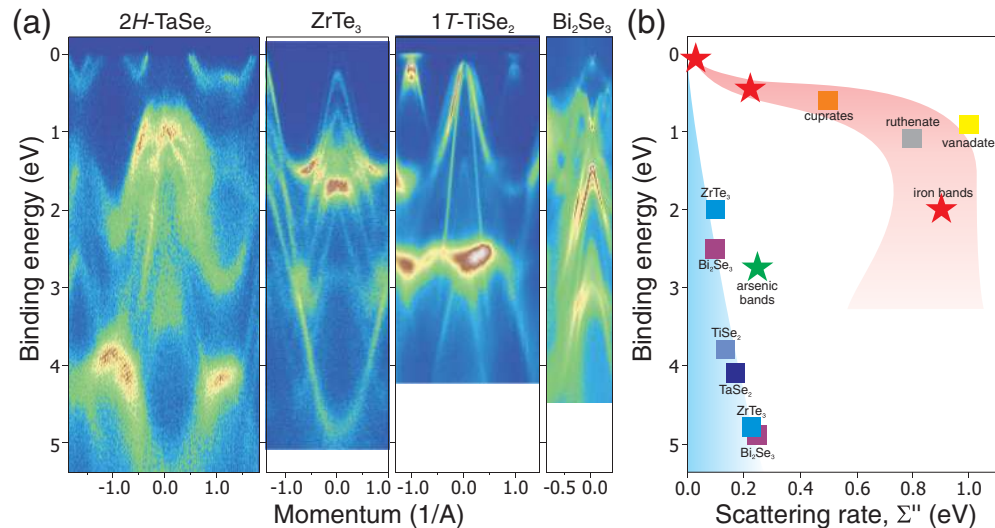
FIG. 4. Dependence of the scattering rate on the binding energy in different compounds. In the materials with relatively weak electronic interactions, such as $TaSe_2$, $ZrTe_3$, $TiSe_2$, and Bi_2Se_3, the spectra are sharp, and the band dispersion remains well discernible down to 5 eV and further from the Fermi level. In contrast, for the materials referred to as "correlated," or featuring strong electron interactions, such as iron-based, cuprate and ruthenate superconductors, and vanadates, the scattering rate grows very fast, and sharp dispersing features are observed only within several hundreds of meV from the Fermi level. (a) ARPES spectra of $TaSe_2$, $ZrTe_3$, $TiSe_2$, and Bi_2Se_3. (b) Scattering rate, determined from the width of energy distribution curves; data for cuprates from Refs. [16,33]; for ruthenates from Ref. [17]; for vanadates from Ref. [18].

several materials is adduced in Fig. 4(b). There is a drastically different behavior of the scattering rates for groups of materials with and without substantial electronic interactions.

Certainly, the most interesting question is the relation of the above-discussed large-scale electronic interaction to the electron pairing. It is *a priori* clear that the introduced effective bosonic spectrum can hardly be considered as a pairing interaction in a conventional sense. Moreover, if one makes an attempt to estimate T_c with the parameters of the extracted spectrum, $\lambda \approx 2$, $\omega_{average} \approx 0.5$ eV, then with the original BCS formula one arrives at $T_c \approx \omega_{average} \exp^{-1/\lambda} = 3600$ K, and at 970 K with McMillan's expression [37]—as expected, one gets values that are way too high. On the other hand, there is a rather clear hint for the importance of such electronic interactions for high-temperature superconductivity: It is present in all studied high-T_c superconductors. Finally, there are a number of materials isostructural to iron arsenides which were synthesized with complete substitution of one or more elements, e.g., $BaCo_2As_2$ [38], $BaNi_2As_2$ [39], and $SrPd_2Ge_2$

[40]. In all of these materials superconductivity is either absent, or T_c is low and superconductivity is believed to be of a conventional phonon origin. Remarkably, the band renormalization peculiar to iron-based superconductors, in all these materials, is not observed. Thus, there is a strong empirical indication for strong electronic interactions on the energy scale of the whole d band to be a necessary requisite for unconventional high-temperature superconductivity.

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[14] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.96.060501 for a more detailed identification of the dispersive bands in the vicinity of the Fermi level, derivation of convenient formulas for the self-energy, and for details of the spectral function analysis in a wide range of binding energy.

[15] From the practical side there are good reasons to operate the notion of a *hypothetical* bosonic spectrum here: (i) Real, and imaginary, parts of the self-energy, introduced by the presented integrals, always appear to be Kramers-Kronig consistent—otherwise, one would have to control the consistency separately; (ii) the self-energy, describing the experiment, corresponds to the $\alpha^2 F$ of a rather simple single-peak shape. From a bit more general perspective, it is not completely surprising that the interaction of a given particle with the rest of the electronic system can be approximated in terms of a scattering on some (broad) bosonic spectrum. Certainly, we do not keep in mind any well-defined excitations, but rather think that it may be possible to single out some (let them be poorly defined) corresponding modes.

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