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Strong pressure-dependent electron-phonon coupling in FeSe

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We have computed the correlated electronic structure of FeSe and its dependence on the A_{1g} mode versus compression. Using the self-consistent density functional theory-dynamical mean field theory (DFT-DMFT) with continuous time quantum Monte Carlo, we find that there is greatly enhanced coupling between some correlated electron states and the A_{1g} lattice distortion. Superconductivity in FeSe shows a very strong sensitivity to pressure, with an increase in T_c of almost a factor of 5 within a few GPa, followed by a drop, despite monotonic pressure dependence of almost all electronic properties. We find that the maximum A_{1g} deformation potential behaves similar to the experimental T_c. In contrast, the maximum deformation potential in DFT for this mode increases monotonically with increasing pressure.

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So far there is no predictive theory for superconductivity in the cuprate and iron superconductors, hence these superconductors can be classified as nonconventional superconductors, since there is a well-developed, predictive theory for electron-phonon superconductors whose normal state is well represented by conventional density functional theory (DFT) [1–3]. The unconventional superconductors are very sensitive to applied pressure, so pressure provides additional routes to test different theories and develop a better understanding of high T_c superconductors [4–6]. Here we study superconductivity under applied pressure in pure FeSe; unlike cuprates, superconductivity in FeSe arises without doping. FeSe is an ideal system to study the electron pairing mechanism due to the simplicity of its crystal and electronic structure. It shows a very strong enhancement of T_c upon application of modest pressure with dramatic increase of T_c from 8 to $\sim$37 K [4,7,8] and then decreases upon further application of pressure. Why does T_c increase with pressure and then decrease for rather small lattice compression? This question was addressed in Ref. [8], where it was found that applied pressure (P) intensified antiferromagnetic spin fluctuations. However, this did not explain the decrease in T_c with further compression.

The discovery of the iron superconductors showed that high T_c is not specific to the cuprates, and suggests a wider field of potential high-T_c materials [9]. Although DFT gives many properties reasonably accurately for both cuprates [10–12] and Fe superconductors [13–15], there is also significant indication of the importance of correlations and fluctuating local moments beyond DFT, especially for the Fe superconductors, which are paramagnetic metals in room temperature, and dynamical mean field theory (DMFT) has proved to be a good approximation [16–22].

While most studies suggest a spin fluctuation coupling mechanism for superconductivity in Fe superconductors similarly to cuprates [23–25], strong coupling phonons have also been proposed to play a role in both sets of materials [3,26–30]. The study of strong electron-phonon coupling in correlated solids is in its infancy due to extreme computational complexity. Only in the non-self-consistent Hubbard I and LDA + U approximations has it proved tractable so far [31]. The role of lattice vibrations in the mechanism of superconductivity in unconventional superconductors is still controversial. The observation of strong electron-phonon and spin-phonon coupling, both in cuprates [32–35] and iron superconductors [36–43], indicates that the electron-phonon coupling (EPC) may play an important role in unconventional superconductors, at least to explain the observed Fe-isotope effect [44], the anomalous temperature dependence of the local Fe-As displacement [45], gap anisotropy, and the correlation of T_c with the Fe-anion height [46]. These observations also suggest polaron- and/or bipolaron-driven superconductivity in this material [47–52].

Here we examine the effects of pressure on the electronic structure and electron-phonon interaction (EPI) in FeSe using DMFT in combination with the DFT as implemented in [53].

Electronic and magnetic properties of FeSe are very sensitive to the position (z_{Se}) of the selenium layers with respect to the iron layers [15,46,54–58]. The magnetic ordering is found to be strongly affected by the chalcogen height [54]. An accurate estimation of z_{Se} is essential to study the electronic structure of FeSe. We optimized z_{Se} for $P = 0$, 3.4, 7.2, and 11 GPa and notice a monotonic increase with P. The reported experimental values of z_{Se} at ambient pressure are consistently estimated to $z_{Se} = 0.267$ [59]. Our DFT+DMFT computations give a value of $z_{Se} = 0.27$ at ambient pressure (Fig. S2 in the Supplemental Material [60]), whereas LDA and spin-polarized generalized gradient approximation (GGA) give 0.234 [13] and 0.26, respectively. The inclusion of local spin fluctuations by DMFT is hence crucial to describe the structural properties in the disordered paramagnetic state, and has a similar effect on z_{Se} as the presence of long-ranged order in standard DFT.

We compute the DFT-DMFT spectral function $[A(\omega,k)]$ and the Fermi surface (FS) for the optimized value of z_{Se} at different pressures and summarized them in the Fig. 1. A dramatic change in $A(\omega,k)$ is noticed when the pressure is increased from 0 to 3.4 GPa. There are three DFT-DMFT bands crossing the Fermi energy (E_F) from the Γ to the X point at $P = 0$, while there are only two bands crossing E_F at $P = 3.4$ GPa. The strong three-dimensional band which crosses E_F from Γ to Z at $P = 0$, does not cross E_F above 3.4 GPa.

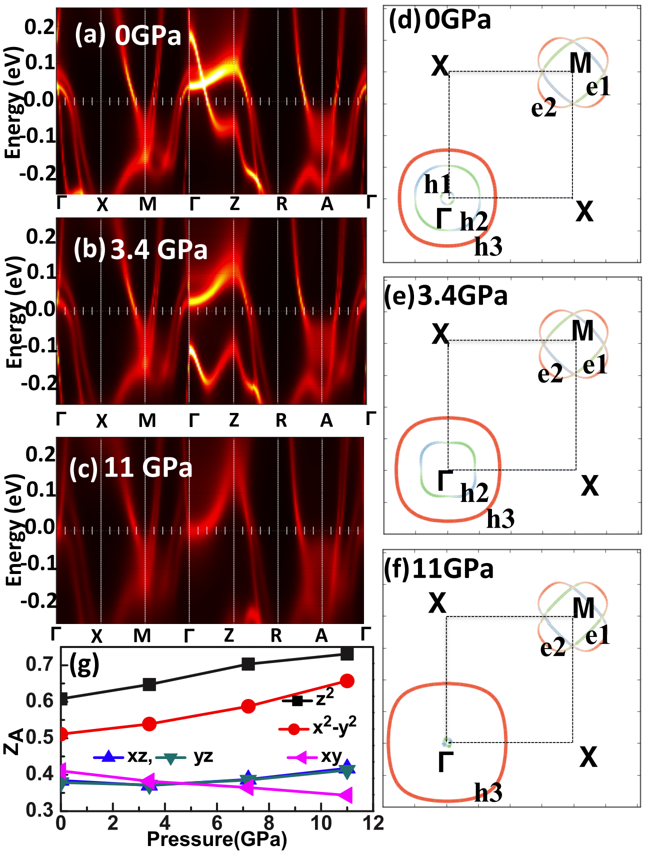

FIG. 1. (Color online) Pressure evolution of DFT-DMFT spectral function [left panel, (a)–(c)] and the Fermi surface on the $k_z = 0$ plane [right panel, (d)–(f)]. The Fermi surface is colored in red, green, and blue according to its dominating orbital character of xy, xz, and yz, respectively. (g) Quasiparticle weight as a function of pressure for the Fe-d orbitals.

Further, we consider the orbital resolved FS at the $k_z = 0$ plane as a function of pressure. From Figs. 1(d)–1(f) we notice that the electron pockets (e1 and e2) at the M point do not change much with the increase of pressure, whereas the hole pockets (h1, h2, and h3) at the Γ point changes significantly. The number of hole pockets reduces from three to one by increasing pressure. The outer hole pockets h3 are mainly of xy character, while the inner hole pockets h1 and h2 consist of xz and yz character. With increasing pressure three hole pockets behave differently. The outer hole pocket h3 expands, while the inner hole pocket h2 shrinks with increasing pressure. The inner hole pocket h1, which shows most k_z dependence, vanishes above $P = 3.4$ GPa.

It is important to know how the electron correlation changes with increasing pressure. In order to investigate the degree of correlation, we calculated $Z_A = (1 - \frac{\delta\Sigma}{\delta\omega})^{-1}_{\omega=0}$. In a Fermi liquid it is the quasiparticle weight, which is unity for the noninteracting system, and is much smaller than unity for the strongly correlated system. We have calculated Z_A for all the Fe-d orbitals and plotted them as a function of P in Fig. 1(g). Though the d_{z^2},$d_{x^2-y^2}$ orbital becomes less correlated with the increase of P, the t_{2g} orbitals (d_{xz}, d_{yz}, and d_{xy}) remain correlated, and in particular the d_{xy} orbital, which carries more magnetic moment, becomes even more correlated with increasing pressure. The predicted Z_A of 0.41 for the d_{xy} orbital at $P = 0$ GPa and at $T = 300$ K differs from 0.287 calculated at $T = 116$ K in Ref. [21] due to the use of slightly different lattice parameters and z_{Se}. By comparing results with Ref. [21], we found that among other t_{2g} orbitals, d_{xy} shows the strongest temperature dependence in Z_A at $P = 0$ GPa.

Calculated electron density of states, optical conductivity both along the ab-plane and the c-axis shows a monotonic behavior as a function of compression (details in the Supplemental Material [60]). We now turn our discussion to the electron-phonon coupling and polaron formation in FeSe. The coupling between electronic states and atomic displacements (the electron-phonon matrix element) and hence λ is directly related to the shift in the energy eigenvalues at E_F. We calculate this shift by calculating δE (details in the Supplemental Material [60]). We estimate the average as well as the maximum value of the deformation potential ($\mathcal{D} = \frac{\delta E}{\delta Q}$) upon Se atom displacement (δQ) in z_{Se} of a particular phonon mode (A_{1g} distortion) for the entire FS. In Fig. 2(a) we have plotted the deformation potential for both DFT and DFT-DMFT methods. The equation of states (pressure points on the axis) are obtained from the experiment [61]. First we notice that the average $\mathcal{D}$ increases in DFT-DMFT over that obtained in DFT for all pressure. At $P = 0$ the average $\mathcal{D}^2$ increases ~1.5 times in DFT-DMFT; $\mathcal{D}$ is 0.84 eV/Å in DFT-DMFT, while 0.69 eV/Å in GGA. A similar increase was found by Boeri *et al.* [28] when magnetic softness was included in their calculations. At $P = 3.4$ GPa the average $\mathcal{D}^2$ increases ~2.25 times in DFT-DMFT. However, this is still not sufficient to obtain 37 K but not small enough to ignore as suggested earlier by Boeri *et al.* [28].

More interesting is the pressure dependence of the maximum $\mathcal{D}$. At ambient pressure we notice that the maximum deformation potential ($\mathcal{D}^m$) in DFT-DMFT is about three times higher than that obtained in GGA. One band is very sensitive to this deformation; it crosses the E_F for $P = 0$, 1.4, 2.6, and -2.0 GPa [indicated by h1 in Fig. 1(d) for $P = 0$]. This sensitive pocket gives a very high value of $\mathcal{D}^m$ for this pressure range within DFT-DMFT method, where the experimental T_c is also observed to be high. $\mathcal{D}^m$ obtained from the GGA is found to be very small at $P = 0$. We found that the $\mathcal{D}$ obtained from DFT-DMFT is different for different parts of the FS (details in the Supplemental Material [60]). For example, at $P = 0$, h1 gives $\mathcal{D}^m$ of ~4.4 eV/Å, while e1 gives only ~1.5 eV/Å with DMFT method. The largest contribution in the enhanced deformation potential is from the hole pocket (h1), located in the Γ region. A nonuniform and anisotropic nature of the electron-phonon coupling has also been seen in the cuprates, where the average EPC was found to be one order of magnitude smaller than the maximum [62]. This is consistent with our results.

With increasing pressure, maximum value of the deformation potential within DFT-DMFT then decreases and remains almost unchanged after $P = 3.4$ GPa; whereas the maximum $\mathcal{D}$ within DFT for this mode is insensitive to increasing pressure except at $P = 11$ GPa, where the experimental T_c is found to decrease. At $P = 11$ GPa, the sensitive band crosses the E_F for the GGA, which leads to a high $\mathcal{D}^m$ for GGA. For pressure above 3.4 GPa, we notice that the maximum $\mathcal{D}$ in

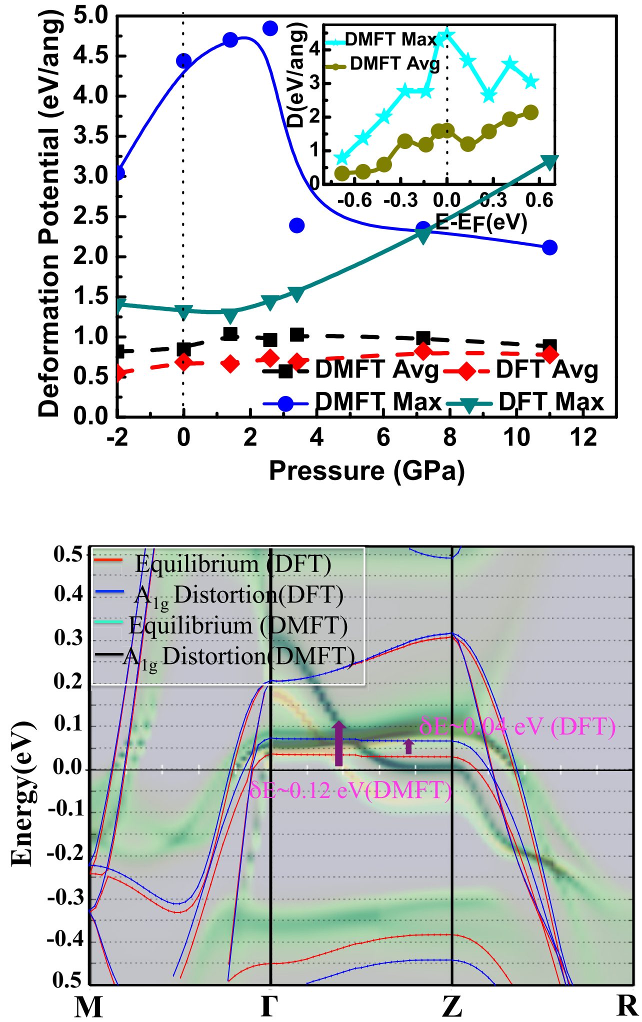

FIG. 2. (Color online) (a) Maximum and Fermi surface average of deformation potentials ($\mathcal{D}$) for the A_{1g} distortion computed in DFT-DMFT, and DFT as a function of pressure indicates the presence of strong EPC in FeSe; inset shows deformation potential as a function of E_F at $P = 0$. (b) Demonstration of huge local electron-lattice coupling for the A_{1g} distortion in our DFT-DMFT computations at $P = 0$ GPa for a selective part of the Brillouin zone; red and blue lines represent GGA bands. The common Fermi energy is considered for the equilibrium position and denoted by the single horizontal line for both DFT and DFT-DMFT methods.

DMFT is from the electron pocket centered at the M point. $\mathcal{D}^m$ also strongly depends on the E_F. The inset of Fig. 2(a) shows the behavior of maximum and average values of $\mathcal{D}$ as a function of E_F at $P = 0$ calculated with DFT-DMFT. So the movement of E_F due to defect or pressure can significantly change the FS topology and hence the $\mathcal{D}$.

The momentum resolved spectral function $A(\omega,k)$ is shown in Fig. 2(b) for both equilibrium position and A_{1g} distortion between the high symmetric points, where the most sensitive band crosses the E_F. The solid red and blue lines represent corresponding GGA bands for equilibrium position and A_{1g} distortion, respectively. From Fig. 2(b), we notice that at $P = 0$, the shift in energy (δE) over the atomic displacement of 0.0276 Å is $\sim$0.12 eV in DFT-DMFT and $\sim$0.04 eV in GGA, respectively. So as reflected from Fig. 2, the $\mathcal{D}$ is about three times higher in DFT-DMFT for this particular region of the Brillouin zone. If we notice carefully in Fig. 2(b), the shift of the bands due to A_{1g} distortion is very nonuniform in DFT-DMFT; a strong deformation potential is noticed only in Γ to Z region, while for the other part of the Brillouin zone, deformation potential is found to be small. This leads to a strong nonuniform EPC at $P = 0$, which is reflected in Fig. 2(a) where maximum $\mathcal{D}$ is found to be about three times higher than the average. We found this similar nonuniform EPC for $P = 1.4$, 2.6, and -2.0 GPa.

We estimate λ using $\mathcal{D}$ (see Supplemental Material [60] for details). While the average λ is still small, the maximum λ in DFT-DMFT is found to be 0.98 at $P = 0$. At $P = 2.6$ GPa, the maximum λ reaches 1.159. We found that only certain electronic states have very strong λ, while the average λ is not strong enough to explain 37 K. So the conventional electron-phonon mechanism seems unlikely. On the other hand, this also indicates that local EPC can be important and one can use a polaron model, where a single electron can strongly couple with the lattice and form polarons. Formation of polarons has been experimentally found in both Fe superconductors [42,63,64] and cuprates [49]. The anomalous temperature dependence of the local Fe-As displacement, observed in Ref. [45], indicates that local rather than global electron-lattice interaction is present in Fe-based superconductors and as suggested in Ref. [48], polaron formation is responsible for the observed anomalies [45]. Though the formation of polarons depends on a lot of factors, such as the band filling, temperature, EPC strength, phonon frequency, etc., our results suggest the use of a polaron model. We consider the electronic state corresponding to maximum λ ($\sim$1) forms a polaron, which is a quasiparticle consisting of an electron and the surrounding lattice distortion. Then the polaronic binding energy (E_p) will linearly depend [65,66] on maximum λ and hence on the square of the maximum deformation potential. Taking the polaronic band into account, Alexandrov and Mott [65] described that T_c exponentially depends on the function of E_p. Under hydrostatic pressure, we found that electronic properties change monotonically, while only $|\mathcal{D}^m|^2$ (and hence E_p) initially grows (up to 3.4 GPa) and then drops, similarly to experimental T_c. This indicates that a strong local EPC plays an important role in Fe-based superconductors.

It is important to mention that T_c was found to increase rapidly for the low pressure range (0–3 GPa) and can reach up to 27 K at 1.48 GPa [67]. The disagreement in the pressure dependence of experimental T_c and our DFT-DMFT calculation of maximum $\mathcal{D}$ can be due to the presence of the mixed phase in low temperature crystal structure in experiment, while our calculations are based on room temperature tetragonal (PbO-type) structure.

The behavior of the DFT-DMFT deformation potential with pressure hints that superconductivity in FeSe may have partially phonon or polaron origin and local EPI plays a very important role in superconductivity in the unconventional superconductors. Analysis of the contributions of each many-body state reveals that charge fluctuations due to correlations and charge transfer from Fe to Se are coupled to the A_{1g} mode.

Our computations predict that applied pressure significantly changes the FS around the Γ point. We show the Fermi surface average of the deformation potential is enhanced up to 50% in DFT-DMFT when compared with standard DFT, and is still not high enough to give high T_c in FeSe. This reflects and confirms that a simple electron-phonon coupling mechanism seems unlikely as relevant from many experimental findings. Calculated electronic properties show a monotonic behavior with applied pressure. We found a strong enhancement of the coupling between correlated electronic states along the high symmetric Γ point and A_{1g} lattice distortion. The maximum deformation potential within GGA for this mode is almost insensitive to increasing pressure except at high pressure where the experimental T_c is found to decrease. The corresponding DFT-DMFT deformation potential behaves similarly to experimental T_c, which increases first and then decreases after a certain pressure.

To explain high T_c by conventional strong coupling EPI enhanced in the DFT-DMFT method, these enhancements would need to be on average just as large for all modes. More importantly the difference (addition) between interband and intraband coupling has to be enhanced to support the $s_{\pm}$ (s_{++}) pairing symmetry. Alternatively, as shown here, if only certain modes become very strongly coupled with certain electronic states, this could give sufficient coupling for polaron formation which requires only certain modes to have extreme coupling leading to condensation of either polaronic and/or bipolaronic states or Cooper pairs [29,42,49,50]. For high-T_c cuprates, Müller *et al.* showed that the normal state polaron can form Cooper pairs and can be responsible for superconductivity [49]. Such polaronic states may involve spin as well as charge fluctuations [29], leading to a problem that still requires significant development for a predictive theory. Even if the giant correlated EPI we predict is not directly responsible for superconductivity, it should help explain some experimental signatures related to EPI in iron-based superconductors [30,45].

The high-T_c superconductors so far are all rather bad metals in the normal state, have low densities of state at E_F, are ionic metals containing transition-metal ions, and are quasi-two-dimensional and highly anisotropic, so it seems any competitive theory should explain why. Spin fluctuations may play a major role, but it seems that is not enough to explain all of these common features. The fact that they are anisotropic bad metals with low densities of state may be because this decreases the screening of the attractive interaction, partly from poorly screened Coulomb fluctuations from phonons or polarons. Correlations may enhance the EPI as we found here, and in addition lead to lower entropy from fluctuations among multiplets in the normal state leading to higher T_c's.

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