

## Correlation strength, Lifshitz transition, and the emergence of a two-dimensional to three-dimensional crossover in FeSe under pressure

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We report a detailed theoretical study of the electronic structure, spectral properties, and lattice parameters of bulk FeSe under pressure using a fully charge self-consistent implementation of the density functional theory plus dynamical mean-field theory method (DFT+DMFT). In particular, we perform a structural optimization and compute the evolution of the lattice parameters (volume,  $c/a$  ratio, and the internal  $z$  position of Se) and the electronic structure of the tetragonal (space group  $P4/nmm$ ) unit cell of paramagnetic FeSe. Our results for the lattice parameters obtained by structural optimization using DFT+DMFT are in good quantitative agreement with experiment, implying a crucial importance of electron correlations in determining the correct lattice properties of FeSe. Most importantly, upon compression to 10 GPa our results reveal a topological change in the Fermi surface (Lifshitz transition) which is accompanied by a two- to three-dimensional crossover and a small reduction of the quasiparticle mass renormalization compared to ambient pressure. The behavior of the momentum-resolved magnetic susceptibility  $\chi(\mathbf{q})$  shows no topological changes of magnetic correlations under pressure but demonstrates a reduction of the degree of the in-plane  $(\pi, \pi)$  stripe-type nesting. Our results for the electronic structure and lattice parameters of FeSe are in good qualitative agreement with recent experiments on its isoelectronic counterpart  $\text{FeSe}_{1-x}\text{S}_x$ .

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### I. INTRODUCTION

Iron chalcogenides (FeCh) with the PbO crystal structure have attracted much attention recently due to their anomalous properties caused by the complex interplay between electronic, magnetic, and lattice degrees of freedom [1]. In this context, FeSe is one of the most well-known members of the so-called 11 family, which shows remarkable connections between magnetism, nematicity, and superconductivity. FeSe has the same planar structure of the FeCh tetrahedra planes as iron pnictides, bound together by the van der Waals interaction but without separating layers [2,3]. In fact, it is structurally the simplest among other Fe-based superconductors, with a superconducting phase emerging below a critical temperature  $T_c \sim 8$  K without applying doping and/or external pressure [4]. In contrast to iron pnictides, FeSe shows no long-range magnetic order but displays a strong enhancement of short-range spin fluctuations near  $T_c$ , with a resonance at the  $(\pi, \pi)$  magnetic vector (stripe type) in the spin excitation spectra [5]. Moreover, similar to many other Fe-based materials, it exhibits a tetragonal-to-orthorhombic transition to a nematic phase below  $T_s \sim 90$  K, with a spontaneously broken lattice rotational  $C_4$  symmetry in the  $ab$  plane [1,6,7].

Upon compression,  $T_s$  appears to decrease, followed by the emergence of a magnetically long range ordered phase at  $\sim 1$  GPa [8]. It leads to a dramatic increase of the critical temperature  $T_c$  to a maximum of about 37 K at  $\sim 6$  GPa, implying that  $T_c$  depends very sensitively on even a moderate change

of the lattice volume [5,9]. Furthermore, it has recently been shown that the electronic and lattice properties of bulk FeSe can be effectively tuned by the isoelectronic substitution of Se by S and Te, respectively. Namely, S has a smaller ionic radius than Se and hence results in a compression of the unit cell, while Te has a larger ionic radius, leading to an expansion of the lattice. In the expanded lattice  $T_c$  is found to increase up to 14 K [10], while tetragonal FeS has been reported to be a superconductor with  $T_c \sim 5$  K [11]. Moreover, superconductivity in FeSe can be boosted to  $\sim 40$  K and even to about 100 K by means of K intercalation [12] and in the case of a monolayer of FeSe on  $\text{SrTiO}_3$  [13]. Interestingly, while FeSe is not magnetically long range ordered, magnetism appears both upon compression (most likely consistent with a stripe-type  $(\pi, \pi)$  vector [14]) and upon expansion. For example, the end member FeTe has a long-range  $(\pi, 0)$  antiferromagnetic order [15]. These findings clearly demonstrate that isoelectronic tuning of the lattice of FeSe, e.g., by substitution, can be an effective means to control  $T_c$  in FeCh.

On the experimental and theoretical sides, much effort has been devoted to understanding the electronic properties of FeCh employing, e.g., angle-resolved photoemission spectroscopy (ARPES) measurements [1] and theoretical band structure calculations [16]. In particular, it has been shown that FeSe has the same Fermi surface (FS) topology as the pnictides, characterized by an in-plane stripe-type nesting  $(\pi, \pi)$ , consistent with  $s^\pm$  pairing symmetry [17]. A significant narrowing of the Fe  $3d$  bandwidth by a factor of  $\sim 2$  and a

large orbital-selective enhancement of the quasiparticle mass  $m^*/m$  in the range of  $\sim 3$ – $20$  were reported [18,19], implying the crucial importance of (orbital-selective) electronic correlations [20–24]. In addition, recent photoemission data show the formation of the lower Hubbard-band satellite at about  $-2$  eV in bulk FeSe which is absent in density functional theory (DFT) [22,24]. Furthermore, computations of the lattice parameters of iron-based superconductors within DFT highlight peculiar deviations with experiments not captured by standard band structure methods [23,25]. All this suggests a complex interplay between electronic correlations and the lattice, implying the crucial importance of the effect of electronic correlations in FeCh.

State-of-the-art methods for the calculation of the electronic structure of correlated materials using band structure methods combined with dynamical mean-field theory (DFT+DMFT) [26,27] have provided a good quantitative description of the electronic structure of FeCh [23,24,28,29]. In fact, recent DFT+DMFT calculations showed a significant orbital-dependent mass enhancement in the range of 2–5 and demonstrated the existence of a lower Hubbard band at about  $-1.5$  to  $-2.0$  eV below the Fermi level of FeSe [22–24,28]. Electronic correlations in FeSe show a strong sensitivity to variation of the crystal-structure parameters. For example, both  $T_c$  and the correlation strength are found to be affected by an isostructural change in the lattice volume [19]. Indeed, upon expansion of the lattice caused by substitution of Se for Te,  $T_c$  increases up to  $\sim 14$  K. It also leads to a pronounced damping of quasiparticle coherence, i.e., to an enhancement of the strength of electronic correlations [19]. By contrast, upon compression of the lattice, e.g., upon partial substitution of Se by S, the electronic correlations become weaker, as can be expected for a correlated metal [30]. This is associated with a substantial increase in the bandwidth; that is, tetragonal FeS is less correlated than FeSe. We also note that  $T_c$  is about 5 K in bulk FeS, which is close to  $\sim 8$  K in FeSe [11]. Interestingly, it has recently been proposed that in FeS the electron-phonon interaction is significant [31].

Moreover, the above-mentioned changes in the lattice volume have a strong effect on the FS. In fact, in  $\text{Fe}(\text{Se}_{1-x}\text{Te}_x)$ , i.e., upon expansion of the lattice, experiments observe a suppression of the spectral weight associated with the FS pockets near the  $M$  point of the Brillouin zone [19]. This behavior is accompanied by the appearance of spectral weight near the  $X$  point, indicating the existence of a topological change in the FS (Lifshitz transition). We note that this effect, as well as a  $(\pi, \pi)$  to  $(\pi, 0)$  reconstruction of magnetic correlations upon going from FeSe to FeTe, has been well established by recent DFT+DMFT calculations [23]. Furthermore, compression of the lattice due to chemical (positive) pressure causes a damping of the spectral weight associated with the innermost FS hole pocket at the  $\Gamma$  point [32]. This suggests the possibility of superconductivity in FeCh without coherent quasiparticles in a hole band forming the inner cylinder of the FS. Overall, these results reveal a subtle interplay between the electronic, magnetic, and structural properties of FeSe and demonstrate the crucial importance of electronic correlations.

In our previous reports [23] based on the DFT+DMFT approach, we studied the electronic structure and phase stability of FeSe under *negative* pressure, which can be achieved

experimentally through partial substitution of Se by Te. In agreement with experiment FeSe was shown to exhibit a remarkable change in the electronic structure and the magnetic and lattice properties upon expansion of the lattice. Namely, a structural transition from a collapsed-tetragonal to tetragonal phase was found to occur, which was accompanied by a reconstruction of the electronic structure (Lifshitz transition) and by a  $(\pi, \pi)$  to  $(\pi, 0)$  change in magnetic correlations. Furthermore, there is a crossover from Fermi-liquid to non-Fermi-liquid behavior [23]. At the same time the synergy of electronic structure, magnetism, and the lattice of FeSe under *external* pressure has remained essentially unexplored. For that reason we will address that question in our present investigation to provide a microscopic explanation of the properties of FeSe under pressure.

In this paper, we focus on the interplay between the electronic structure and the magnetic and structural properties of the tetragonal ( $P4/nmm$ ) FeSe under external (positive) pressure. In particular, we study the effect of pressure on the unit-cell shape, band structure, magnetic properties, and Fermi surface of FeSe, employing a fully charge self-consistent implementation of the DFT+DMFT method [33]. We perform a structural optimization and compute the evolution of the lattice parameters, i.e., the volume, the  $c/a$  ratio, the internal  $z$  position of Se (which is related to the height of Se above the Fe plane), and the electronic structure of the tetragonal unit cell (space group  $P4/nmm$ ) of paramagnetic FeSe. Our results demonstrate that electronic correlations need to be taken into account in the structural optimization of tetragonal FeSe at ambient and positive pressure. We show that the lattice parameters of FeSe obtained within DFT+DMFT, i.e., when taking into account the effect of electronic correlations, are in good agreement with experimental data. Most importantly, our results reveal a topological change in the Fermi surface (Lifshitz transition) which is accompanied by a two- to three-dimensional crossover. We obtain a small reduction in the quasiparticle mass renormalization  $m^*/m$  by about 5% for the  $e$  states and less than 1% for the  $t_2$  states compared to ambient pressure. While the behavior of the momentum-resolved static susceptibility  $\chi(\mathbf{q})$  shows no topological changes in magnetic correlations under pressure, it demonstrates a reduction in the degree of the in-plane  $(\pi, \pi)$  stripe-type nesting. Our results for the electronic structure and lattice parameters of FeSe under pressure are in line with recent experiments on its isoelectronic and isostructural counterpart  $\text{FeSe}_{1-x}\text{S}_x$ . Overall, our results show the sensitivity of the electronic structure and magnetic properties of FeCh with respect to a small variation of the lattice. This is associated with a peculiarity of its correlated band structure, namely, the proximity of a Van Hove singularity associated with the  $xy$  and  $xz/yz$  Fe  $3d$  orbitals to the Fermi level [23,34].

## II. METHOD

To study the electronic structure, spectral properties, and phase stability of bulk FeSe under pressure we make use of the DFT+DMFT computational approach [27]. We employ a fully charge self-consistent DFT+DMFT scheme [33] and perform direct structural optimization of the tetragonal (space group  $P4/nmm$ ) unit cell of FeSe, calculating the total energy as

a function of volume, lattice parameter  $c/a$ , and the internal  $z_{\text{Se}}$  position. For simplicity  $z_{\text{Se}}$  was fixed to its experimental value  $z_{\text{Se}} = 0.266$  [6] in some of these calculations. In order to determine the pressure, we fit our total-energy results to the third-order Birch-Murnaghan equation of state [35]. The calculations are performed for the tetragonal ( $P4/nmm$ ) crystal structure of paramagnetic FeSe at an electronic temperature  $T = 290$  K. We note that according to experimental data, bulk FeSe undergoes a tetragonal-to-orthorhombic (nematic) phase transition at sufficiently lower temperature  $\sim 90$  K (at ambient pressure) [1,6,7].

The DFT+DMFT approach is implemented within plane-wave pseudopotentials with the generalized gradient approximation (GGA) in DFT [36]. For the partially filled Fe  $3d$  and Se  $4p$  orbitals we construct a basis set of Wannier functions using the projection procedure on a local atomic-centered symmetry-constrained basis set within a window spanning both the Fe  $3d$  and Se  $4p$  bands [37]. To solve the realistic many-body problem, we employ the continuous-time hybridization-expansion (segment) quantum Monte Carlo algorithm [38]. In accordance with previous studies of the pnictides and chalcogenides, we use the average Coulomb interaction  $U = 3.5$  eV and Hund's exchange  $J = 0.85$  eV parameters for the Fe  $3d$  orbitals [23,39]. The  $U$  and  $J$  values are assumed to remain constant upon variation of the lattice volume. The Coulomb interaction is treated in the density-density approximation. Spin-orbit coupling is neglected in these calculations. Furthermore, we employ the fully localized double-counting correction, evaluated from the self-consistently determined local occupations, to account for the electronic interactions already described by DFT. To compute spectral properties and renormalizations of the effective electron mass of the Fe  $3d$  orbitals, we calculate the real-axis self-energy  $\Sigma(\omega)$  using the Padé analytical continuation procedure [40]. The evolution of magnetic correlations under pressure is analyzed by calculating the momentum-resolved static susceptibility in the particle-hole bubble approximation:

$$\chi(\mathbf{q}) = -k_{\text{B}}T \sum_{\mathbf{k}, i\omega_n} \text{Tr} \hat{G}(\mathbf{k}, i\omega_n) \hat{G}(\mathbf{k} + \mathbf{q}, i\omega_n), \quad (1)$$

where  $T$  is the temperature,  $\omega_n$  is the fermionic Matsubara frequency, and  $\hat{G}(\mathbf{k}, i\omega_n)$  is the interacting lattice Green's function computed within DFT+DMFT.

### III. RESULTS

#### A. Crystal-structure optimization

We start by performing a structural optimization of tetragonal ( $P4/nmm$ ) FeSe at ambient pressure. To this end, we compute the total energy as a function of volume, lattice parameter  $c/a$ , and the internal position of Se  $z_{\text{Se}}$  (the space-group symmetry remains unchanged,  $P4/nmm$ ) using the charge self-consistent DFT+DMFT method. The equilibrium structural parameters are determined by calculating the minimum of the total-energy functional. Our results are summarized in Fig. 1. We note that the equilibrium lattice parameters of FeSe computed within the nonmagnetic GGA (nm-GGA) using the plane-wave pseudopotential approach, i.e.,  $V = 563$  a.u.<sup>3</sup>,  $c/a = 1.671$ , and  $z_{\text{Se}} = 0.227$ , are substantially

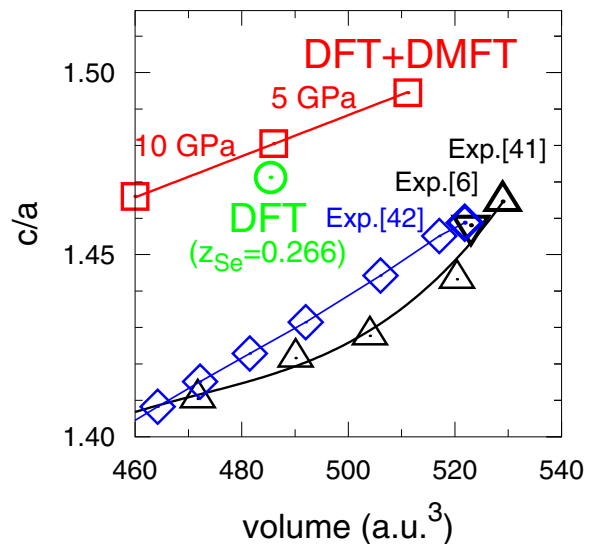


FIG. 1. Evolution of the lattice volume and  $c/a$  ratio of paramagnetic tetragonal FeSe under pressure obtained by DFT+DMFT at  $T = 290$  K (squares) compared to the experimental results (triangles [41], rotated squares [42], inverted triangle [6]). Bold triangles and square mark the cell parameters at ambient pressure. The nm-GGA result with  $z_{\text{Se}}$  fixed to its experimental value is shown by a circle. Note that a fully relaxed result obtained within nm-GGA would be out of the range of the plot (see text).

(by  $\sim 6\%$ – $15\%$ ) off the experimental values; this is out of the range of the plot of the  $c/a$  ratio vs volume shown in Fig. 1 [6,41,42]. Fixing the  $z$  position of Se to its experimental value  $z_{\text{Se}} = 0.266$ , the equilibrium lattice volume and the  $c/a$  ratio values calculated within nm-GGA are 485 a.u.<sup>3</sup> and 1.471, respectively, which is about 8% lower (volume) and  $\sim 1\%$  larger ( $c/a$ ) than those found in experiment (see Fig. 1). We note, however, that this remarkable failure of a nonmagnetic DFT approach is not that surprising in view of the importance of short-range magnetic correlations reported in FeSe [43]. In fact, the structural properties of FeSe obtained with DFT for particular (static) magnetic configurations agree better with experiment (see Table I in Ref. [44]).

By contrast, our results for the equilibrium lattice parameters obtained with DFT+DMFT agree well with experiment. In particular, we find the equilibrium volume to be 511 a.u.<sup>3</sup> and the  $c/a$  ratio to be 1.494, which agree within 3% with the experimental values. The calculated Se  $z$  fractional position  $z_{\text{Se}} = 0.261$  is in excellent agreement with experiment [6]. Furthermore, while in nm-GGA the results for the lattice parameters obtained with and without optimization of  $z_{\text{Se}}$  strongly differ, that is not the case within DFT+DMFT. Our result for the (instantaneous) local magnetic moment  $\sqrt{\langle \hat{m}_z^2 \rangle} \sim 1.86\mu_{\text{B}}$  is close to that found in the previous charge self-consistent DFT+DMFT calculations [23] with  $c/a$  fixed to its experimental value. The calculated bulk modulus is  $K_0 \sim 85$  GPa, which is comparable with that for iron pnictides. We note that this is much lower than the result obtained without electronic correlations (116 GPa).

Moreover, we observe that the  $c/a$  ratio obtained within DFT+DMFT is slightly overestimated by about 3%. This is presumably related to the van der Waals (attractive) interaction

between the electrically neutral FeSe layers, which is not taken into account in our calculations. It was shown previously that in the binary chalcogenides the local (repulsive) Coulomb interaction is important for calculating the in-plane lattice constant and the volume of the unit cell [23]. On the other hand, the van der Waals attraction was found to correct the overestimation of the out-of-plane lattice parameter in DFT [45].

Next, we calculate the evolution of the lattice parameters of FeSe under pressure (see Fig. 1). For simplicity, in these calculations the internal coordinate  $z_{\text{Se}}$  was fixed at its experimental value, which is very close to that obtained within DFT+DMFT at ambient pressure. Furthermore, for subsequent verification we perform an optimization of  $z_{\text{Se}}$  at a pressure of  $\sim 10$  GPa, which results in a weak  $\sim 2\%$  increase of  $z_{\text{Se}}$ , in agreement with experiment [41]. In Fig. 1 we display our results for the evolution of the  $c/a$  ratio and the lattice volume of FeSe upon compression. Our calculations exhibit a monotonous decrease in the  $c/a$  ratio and the lattice volume of FeSe, which can be approximated (up to the accuracy of the present DFT+DMFT calculations) by a straight line. The slope of this line, i.e., the relative dependence of the  $c/a$  on the lattice volume, is in good agreement with experimental data for the unit-cell parameters at 2–11 GPa [41,42]. However, the experimental data [41] show a substantial change (kink) in the slope of the  $c/a$  ratio vs lattice volume at about 2 GPa ( $V \sim 520$  a.u.<sup>3</sup>), which is not reproduced in our DFT+DMFT calculations. We attribute this discrepancy to the van der Waals attraction between the FeSe layers, which is not included in the present calculations. Moreover, the absence of short-range magnetic correlations within DMFT may also play a role.

Our results for the lattice parameters of FeSe under pressure are in overall good agreement with recent experimental data [41]. Moreover, our calculations demonstrate the crucial importance of electronic correlations in determining the structural properties of FeSe under pressure. For a pressure of  $\sim 10$  GPa, we obtain a lattice volume  $V \sim 460$  a.u.<sup>3</sup> and a ratio  $c/a = 1.464$ , which are  $< 1\%$  larger than the lattice volume and about 4% larger than  $c/a$  observed in experiment (see Table I), respectively. Our result for the local magnetic moment of  $1.82\mu_B$  is close to the result obtained at ambient pressure.

### B. Spectral properties and Fermi surface under pressure

We now compute the evolution of the spectral properties of FeSe under pressure. Our results for the Fe 3d spectral functions obtained with DFT+DMFT at ambient pressure and upon compression to  $\sim 10$  GPa are shown in Fig. 2. The former results are in good quantitative agreement with our previous DFT+DMFT study of FeSe [23]. In particular, all five Fe 3d orbitals form a band with a predominant contribution at the Fermi level originating from the  $t_2$  states. Moreover, in accordance with previous investigations, we observe a reduction of the Fe 3d bandwidth (in comparison to the DFT results) near the Fermi energy caused by electronic correlations. The lower Hubbard band associated with the Fe  $d_{xy}$  and  $d_{xz/yz}$  orbitals is located at about  $-1.5$  eV [22–24]. The Fe  $d_{xy}$  and  $d_{xz/yz}$  spectral functions exhibit a well-defined quasiparticle peak located below the Fermi level at about 0.2 eV, originating from the Van Hove singularity of the  $d_{xy}$  and  $d_{xz/yz}$  bands at the

TABLE I. Calculated structural parameters of tetragonal FeSe at ambient pressure and those at about  $\sim 10$  GPa as obtained by nm-GGA and DFT+DMFT in comparison with experiment [6,41]. Here  $h_{\text{Se}}$  is the height of Se above the basal Fe plane, and  $d_{\text{Fe-Se}}$  and  $d_{\text{Fe-Fe}}$  are the distances between the corresponding ions; a.p. indicates results obtained at ambient pressure.

$p$ (GPa)	$V$ (a.u. <sup>3</sup> )	$c/a$	$z_{\text{Se}}$	$d_{\text{Fe-Se}}$ (a.u.)	$d_{\text{Fe-Fe}}$ (a.u.)	$h_{\text{Se}}$ (a.u.)
DFT+DMFT						
a.p.	511	1.494	0.261	4.438	4.950	2.729
10	460	1.464	0.280	4.395	4.807	2.787
nm-GGA						
a.p.	563	1.671	0.227	4.366	4.920	2.638
10	433	1.402	0.282	4.307	4.781	2.670
Experiment (room temperature)						
a.p. [6]	523	1.458	0.266	4.498	5.024	2.759
a.p. [41]	529	1.464	0.288	4.657	5.036	3.001
8.5 [41]	456	1.406	0.319	4.614	4.858	3.080
11 [41]	447	1.397	0.306	4.487	4.824	2.915

$M$  point. Upon compression, we find no substantial spectral weight transfer, in remarkable contrast to the DFT+DMFT results obtained upon *expansion* of the lattice. Our results exhibit a weak increase in the bandwidth and a subtle splitting of the peaks associated with the  $e$  states located  $\sim 1$  eV above and below the Fermi level. However, the overall shape of the spectral functions shows only a weak pressure dependence. Under pressure, our results reveal no substantial reduction of the Fe 3d spectral weight at the Fermi level, which is less than 1%. These results agree well with an analysis of the band mass enhancement  $m^*/m = 1 - \partial \text{Re}\Sigma(\omega)/\partial \omega|_{\omega=0}$ , which provides a quantitative measure of the correlation strength. We calculate the derivative  $\partial \text{Re}\Sigma(\omega)/\partial \omega|_{\omega=0} = \partial \text{Im}\Sigma(i\omega)/\partial i\omega|_{i\omega=0}$  employing Padé extrapolation of the self-energy  $\Sigma(\omega)$  to  $\omega = 0$  eV. In particular, under a pressure of  $\sim 10$  GPa we obtain

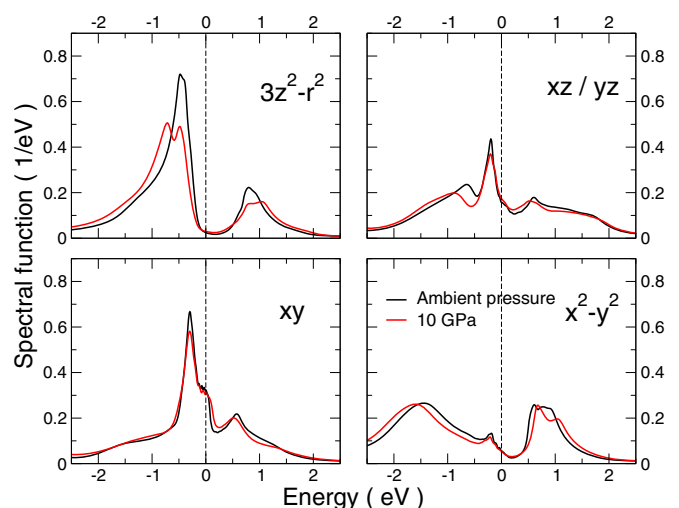


FIG. 2. Orbitaly resolved Fe 3d spectral functions of FeSe obtained within DFT+DMFT at  $T = 290$  K for ambient pressure and for about 10 GPa. The Fermi energy ( $E_F = 0$  eV) is shown by a vertical line.

TABLE II. Orbitally resolved Fe 3d band mass enhancement  $m^*/m$  of FeSe computed for ambient pressure (a.p.) and for  $p \sim 10$  GPa by DFT+DMFT at  $T = 290$  K.

$p$ (GPa)	$3z^2 - r^2$	$xz$	$yz$	$xy$	$x^2 - y^2$
a.p.	1.42	1.67	1.67	1.82	1.38
10	1.35	1.66	1.66	1.83	1.33

an  $\sim 5\%$  reduction of  $m^*/m$  for the  $e$  states and less than 1% for the  $t_2$  states, compared to the ambient pressure. Our results for  $m^*/m$  calculated for the different Fe 3d orbitals at ambient pressure and  $p \sim 10$  GPa are summarized in Table II.

To proceed further, we calculate the  $\mathbf{k}$ -resolved spectral functions of FeSe. In Fig. 3 we present our results for the FS of FeSe calculated for  $k_z = 0$  and  $k_x = k_y$  by integration of the spectral weight  $A(\mathbf{k}, \omega) \equiv -\frac{1}{\pi} \text{Im}[G(\mathbf{k}, \omega)]$  over a  $\pm 5$  meV energy window around the Fermi level. In agreement with previous studies, at ambient pressure we find two intersecting elliptical electron Fermi surfaces centered at the Brillouin zone  $M$  point, associated with the  $d_{xy}$  and  $d_{xz/yz}$  orbitals. In addition, there are three concentric holelike pockets at the  $\Gamma$  point (the two outer hole pockets are degenerate). The FS is seen to be remarkably expanded compared to the low-temperature ARPES measurements [1, 18, 19]. It was indeed proposed that this is related to a “blue-red shift” problem [34], i.e., inappropriate shifting upward of the electron bands at the  $M$  point and

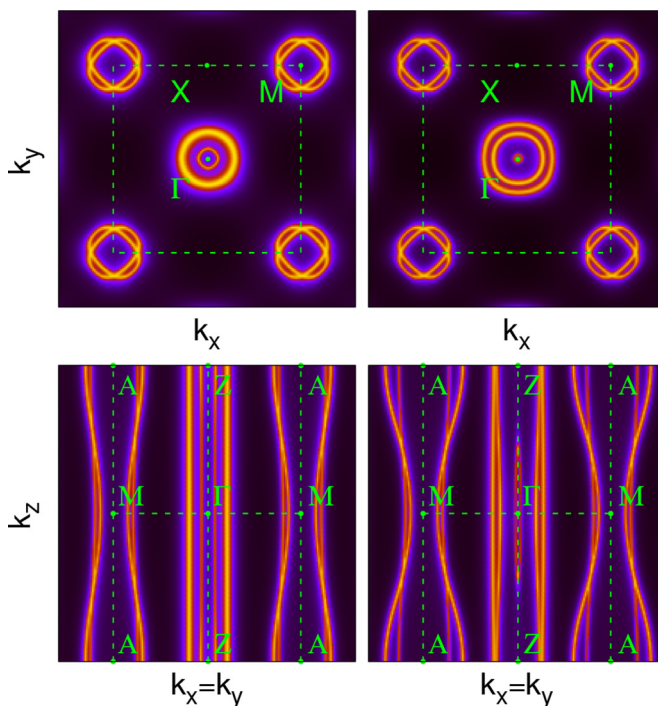


FIG. 3. Fermi surface sections in the  $k_z = 0$  (top) and  $k_x = k_y$  (bottom) planes of the tetragonal Brillouin zone of FeSe at ambient pressure (left column) and at  $p = 10$  GPa (right column) as computed by DFT+DMFT at  $T = 290$  K. Under pressure the inner holelike Fermi surface centered at the  $\Gamma$  point collapses halfway between the  $\Gamma$  and  $Z$  points, resulting in a closed ellipticlike three-dimensional FS (bottom right panel).

downward of the holelike bands at the  $\Gamma$  point, presumably due to a momentum dependence of the real part of self-energy  $\Sigma(\mathbf{k}, \omega)$  [the latter, by construction, is local,  $\mathbf{k}$ -independent  $\Sigma(\omega)$  in our single-site DFT+DMFT calculations]. We note, however, that our DFT+DMFT calculations are carried out at  $T = 290$  K, whereas a direct comparison of our result for the FS with recent room-temperature ARPES measurements [34] shows good quantitative agreement.

We find that bulk FeSe is a correlated metal with coherent quasiparticle behavior at the Fermi level (well-defined FS), which exhibits incoherent spectral weight at higher binding energies, in agreement with recent experiments [34]. We note that at ambient pressure, a structural optimization including relaxation of the  $z_{\text{Se}}$  coordinate affects neither the topology of the FS nor the in-plane nesting vector  $(\pi, \pi)$ , connecting the electronlike and holelike FS pockets. The latter is indicative of stripe-type spin excitations, consistent with a  $s^\pm$  pairing symmetry [17]. We also note that the size of the FS pockets depends only weakly on the structural optimization of the lattice.

Upon compression of the FeSe lattice, we observe a remarkable reconstruction of the electronic structure and of the FS, i.e., a Lifshitz transition (see Fig. 3, right panel). In particular, the inner holelike FS centered at the  $\Gamma$  point collapses halfway between the  $\Gamma$  and  $Z$  points, resulting in a closed ellipticlike three-dimensional FS, elongated in the  $k_z$  direction, around  $\Gamma$ . At the same time, the topology of the electronlike FS at the  $M$  point shows no qualitative change, while its cross section in the  $(k_x, k_y)$  plane now exhibits a substantial variation for different  $k_z$ . In fact, the elongation of the FS ellipses in the  $(k_x, k_y)$  cross section is small for  $k_z = 0$  and large for  $k_z = \pi$ . Overall, our results clearly show that bulk FeSe under pressure undergoes a Lifshitz transition which is associated with a two-to-three-dimensional crossover of the FS.

Bulk FeSe at about 10 GPa is seen to be a moderately correlated metal with coherent quasiparticle behavior at the Fermi level. The latter makes the electronic properties of FeSe particularly interesting since, under both positive and negative (chemical) pressure, it exhibits a Lifshitz transition. Interestingly, according to experiments  $T_c$  is boosted up to  $\sim 37$  K in FeSe under a pressure of about 9 GPa [5] and up to  $\sim 17$  K in  $\text{FeSe}_{1-x}\text{Te}_x$  upon negative chemical pressure caused by substituting Se by Te [10]. However, on a qualitative level, the Lifshitz transition upon positive pressure does not result in a coherence-incoherence crossover as it was found upon expansion of the lattice in Refs. [19, 23].

The change in the FS results in a corresponding change in magnetic correlations due to a suppression of the in-plane nesting vector  $Q_m = (\pi, \pi)$ . In particular, our results exhibit a small reduction of the magnetic correlations under (positive) pressure; that is,  $\chi(\mathbf{q})$  shows a small decrease due to the  $e$  states compared to the results at ambient pressure. At the same time the in-plane nesting  $Q_m$  vector remains of the  $(\pi, \pi)$  type, which is consistent with stripe-type spin fluctuations (see Fig. 4). This is in stark contrast to the  $(\pi, \pi)$  to  $(\pi, 0)$  reconstruction of the magnetic correlations in FeSe upon expansion of the lattice [negative chemical pressure in  $\text{Fe}(\text{Se}, \text{Te})$ ] previously found in [19, 23]. Furthermore, we note that the nm-GGA results reveal no substantial change in the FS of FeSe upon moderate compression or expansion of the

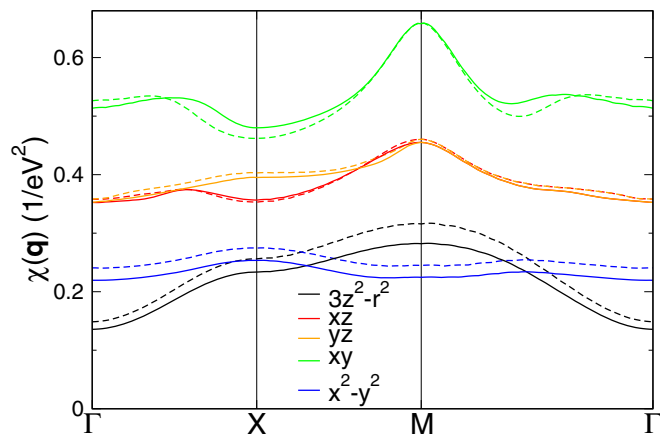


FIG. 4. Orbital contributions to the static spin susceptibility  $\chi(\mathbf{q})$  of paramagnetic FeSe at ambient pressure (dashed line) and  $p = 10$  GPa (solid line) calculated along the  $\Gamma$ - $X$ - $M$ - $\Gamma$  path using DFT+DMFT.

lattice. This highlights once more the great importance of electronic correlations, which are found to be responsible for the remarkable reconstruction of the electronic structure of bulk FeSe, associated with a Lifshitz transition upon both expansion *and* compression of the lattice volume.

To analyze our results further we calculated the electronic structure of bulk FeSe at ambient pressure and at about 10 GPa. In Fig. 5 we present our results for the orbitally resolved spectral function evaluated in the  $\Gamma$ - $X$ - $M$  plane of the Brillouin zone ( $k_z = 0$ ). The total spectral functions of FeSe computed in the  $\Gamma$ - $X$ - $M$  ( $k_z = 0$ ) and  $R$ - $Z$ - $A$  ( $k_z = \pi$ ) planes are shown in Fig. 6. In agreement with previous DFT+DMFT calculations, we obtain a substantial renormalization of the effective crystal-field splitting of the Fe  $3d$  bands with respect to the DFT

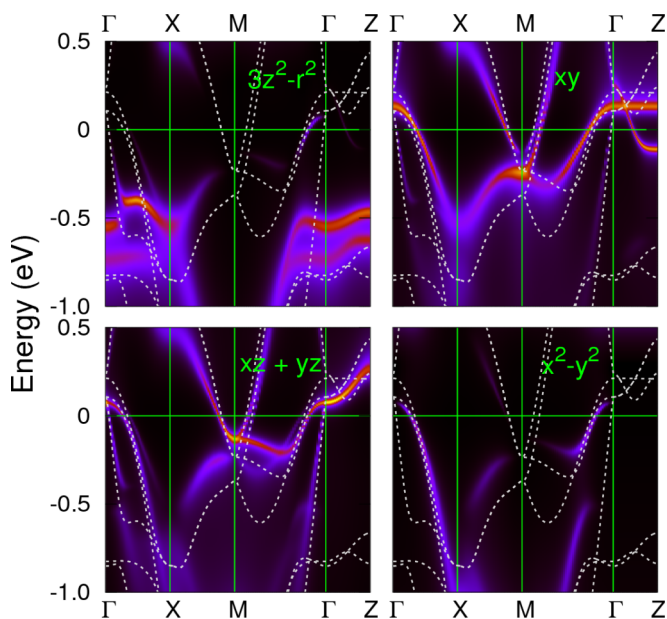


FIG. 5. Band structure of FeSe along the  $\Gamma$ - $X$ - $M$ - $\Gamma$ - $Z$  path at  $p = 10$  GPa as obtained by DFT+DMFT at  $T = 290$  K. The nm-GGA band structure results are depicted by dashed lines.

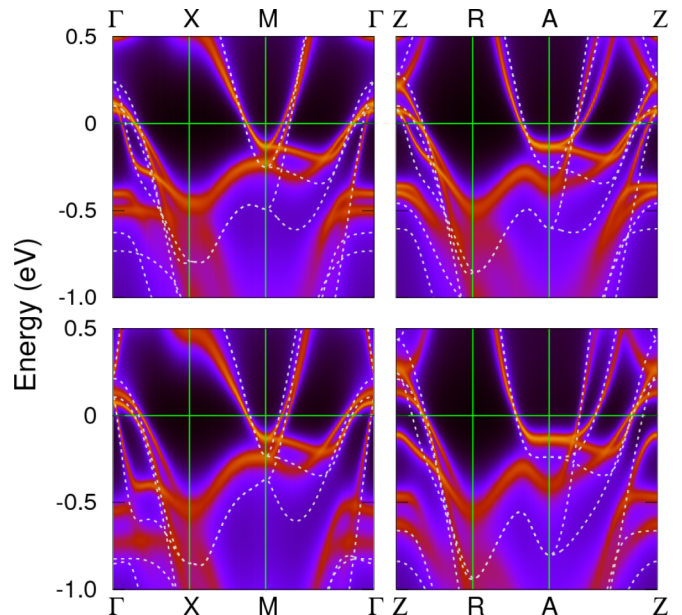


FIG. 6. Band structure of FeSe along the  $\Gamma$ - $X$ - $M$ - $\Gamma$  and  $Z$ - $R$ - $A$ - $Z$  directions at ambient pressure (top row) and  $p = 10$  GPa (bottom row) computed by DFT+DMFT at  $T = 290$  K (contours) and nm-GGA (dashed lines).

results caused by the strong energy and orbital dependence of the self-energy [22,23,28]. The latter leads to different shifts of the quasiparticle bands near the Fermi level. In particular, in agreement with the ARPES measurements, we observe a “blue-red shift” of the states near the  $M$  point (which are pushed upwards) and the hole pockets near the  $\Gamma$  point (pushed downwards). Both shift towards the Fermi level (see Fig. 6) due to orbital-selective correlation effects. In addition, our results clearly distinguish the Van Hove singularity at the  $M$  point, associated with the Fe  $d_{xy}$  and  $d_{xz/yz}$  orbitals (see Fig. 5).

We note that at ambient pressure the spectral function of FeSe evaluated in the  $\Gamma$ - $X$ - $M$  ( $k_z = 0$ ) and  $R$ - $Z$ - $A$  ( $k_z = \pi$ ) planes (see Fig. 6, top row) exhibits the same structure. It consists of three holelike bands near the center (at the  $\Gamma$  and  $Z$  points) and two electron-like bands near the Brillouin zone corners (at the  $M$  and  $A$  points). By contrast, under a pressure of  $\sim 10$  GPa, we observe an orbital-dependent shift of the energy bands near the  $Z$  point, resulting in a topological change in the FS (Lifshitz transition) along the  $\Gamma$ - $Z$  ( $R$ - $Z$ ) direction. In particular, the  $xy$  band shifts towards the Fermi level in the  $k_z = 0$  plane but still remains above the Fermi level (at the  $\Gamma$  point). On the other hand, for  $k_z = \pi$  the  $xy$  band is pushed below the Fermi level (near the  $Z$  point), resulting in the formation of a closed inner hole pocket in the  $\Gamma$ - $Z$  direction. Therefore we conclude that a microscopic mechanism of the Lifshitz transition which results in a two-dimensional to three-dimensional crossover in  $P4/nmm$  FeSe under pressure is associated with a pressure-induced shift of the  $d_{xy}$ -derived band. We note that the  $xy$  band exhibits the largest renormalization of the quasiparticle mass  $m^*/m$  [22–24,28]. Overall, our results show that renormalizations of the band structure of FeSe under pressure are weaker than those at ambient pressure, as expected for a typical correlated metal.

The reconstruction of the electronic structure and the evolution of the FS of FeSe under pressure agree reasonably well with available experiments. Moreover, our results are in line with the experimentally observed expansion of the FS of the  $\text{FeSe}_{1-x}\text{S}_x$  series upon addition of S [30,32]. In fact, tetragonal FeS is an isoelectronic counterpart of FeSe with a smaller lattice volume due to the smaller ionic radius of S with respect to Se. Therefore the properties of  $\text{FeSe}_{1-x}\text{S}_x$  can be regarded as those corresponding to FeSe under positive pressure. With this in mind, our results imply that FeSe under pressure and hence  $\text{FeSe}_{1-x}\text{S}_x$  are less correlated than FeSe, in agreement with recent studies [30]. An analysis of the FSs and magnetic correlations of FeSe under pressure in terms of  $\chi(\mathbf{q})$  suggests that  $\text{FeSe}_{1-x}\text{S}_x$  exhibits stripe-type spin excitations with in-plane  $(\pi, \pi)$  nesting. The latter are weakened by the damping of spin fluctuations upon compression of the lattice [32].

Under pressure, our results reveal the three holelike pockets of the FS in the vicinity of the  $\Gamma$  point, with the inner holelike FS pocket having a three-dimensional character. However, upon further compression it is seen that the three holelike FSs collapse to the two FSs, in agreement with recent studies of FeS [46]. Our results suggest that the pressure-induced increase in the critical temperature in FeSe, which is boosted to 37 K by applying a pressure of about 9 GPa [5], is not of spin-fluctuation origin. Indeed, they indicate that the anomalous behavior of FeSe upon variation of the lattice volume (both compression *and* expansion) is associated with a crossover between superconductivity mediated by spin fluctuations [upon expansion of the lattice in Fe(Se, Te)] and by phonons (upon compression). This conclusion is in qualitative agreement with the enhancement of the electron-phonon coupling in FeSe under positive pressure obtained within DFT+DMFT [28]. While the low-pressure superconducting phase of FeSe most likely has an  $s^\pm$  order parameter, one may expect a change of the symmetry, e.g., to a  $d$ -wave order parameter in the high-pressure phase [17,47].

#### IV. CONCLUSION

By employing a fully charge self-consistent implementation of the DFT+DMFT method we performed structural optimization and studied the electronic properties of the tetragonal ( $P4/nmm$ ) phase of paramagnetic FeSe under pressure at  $T = 290$  K. In agreement with previous studies our results demonstrate that electronic correlations need to be included to determine the correct lattice parameters of FeSe both in equilibrium and upon compression [23]. Our results for the lattice parameters are in overall good agreement with

experimental data. At ambient pressure, the calculated  $c/a$  ratio is about 3% larger than the experimental value, suggesting the possible importance of van der Waals attraction between the FeSe layers, which is absent in the present calculations. Upon compression to 10 GPa, the  $c/a$  ratio and lattice volume show a linearlike decrease by 2% and 10%, respectively, while the fractional coordinate of Se  $z_{\text{Se}}$  weakly increases by  $\sim 2\%$ .

Our results for the spectral properties exhibit a rather weak dependence of the bandwidth on the lattice volume, associated with a small change in the Fe-Se hybridization upon a moderate compression of the lattice up to 10 GPa. Most importantly, we obtained a drastic reconstruction of the Fermi surface topology, i.e., a Lifshitz transition, indicating a two-dimensional to three-dimensional crossover in the FS. In particular, the inner holelike pocket at the  $\Gamma$  point of the FS collapses halfway between the  $\Gamma$  and  $Z$  points, showing a three-dimensional character. We conclude that the microscopic mechanism of the Lifshitz transition is associated with a pressure-induced shift of the  $d_{xy}$  orbital in the  $Z$  point below the Fermi level. Our results are in good agreement with recent ARPES measurements for the  $\text{FeSe}_{1-x}\text{S}_x$  solid solution, which is regarded as an isoelectronic and isostructural (with a smaller lattice volume due to chemical pressure) counterpart of FeSe. Under pressure, we observed a small reduction in the quasiparticle mass renormalization  $m^*/m$  by about 5% for the  $e$  states and less than 1% for the  $t_2$  states compared to results at ambient pressure. This implies that  $\text{FeSe}_{1-x}\text{S}_x$  is less correlated than FeSe, in agreement with recent studies [30,32,46].

The behavior of the momentum-resolved magnetic susceptibility  $\chi(\mathbf{q})$  under pressure shows no topological change in magnetic correlations under pressure. The latter suggests that both FeSe under pressure and  $\text{FeSe}_{1-x}\text{S}_x$  exhibit stripe-type spin excitations. Moreover, an analysis of the FS and magnetic correlations exhibits a small reduction in the degree of the in-plane  $(\pi, \pi)$  nesting, which implies that magnetic correlations are weakened due to the damping of spin fluctuations under pressure. We speculate that the anomalous behavior of FeSe upon variation of the lattice volume under both expansion and compression is associated with a crossover between superconductivity mediated by spin fluctuations [upon expansion of the lattice in Fe(Se, Te)] and by phonons (upon compression).

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