

Understanding nematicity and band structure of FeSe by first-principles study and tight-binding model

Miao-Miao Li^{✉,*}, Xiu-Cai Jiang^{✉,*} and Yu-Zhong Zhang^{✉,†}

Shanghai Key Laboratory of Special Artificial Microstructure Materials and Technology, School of Physics Science and Engineering, *Tongji University*, Shanghai 200092, People's Republic of China



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We systematically investigate the effects of spin order and orbital polarization on the electronic and magnetic structures of nematic FeSe using the LDA + U method. While our calculations with on-site Coulomb repulsion $U \sim 4.5$ eV can account for both the nonmagnetic state at ambient pressure and the phase transition to a stripe antiferromagnetic state at moderate pressure as observed experimentally, neither static spin order nor static orbital polarization can describe the experimental band dispersions from angle-resolved photoemission spectroscopy (ARPES) studies, due to the underestimate of quantum fluctuations. By employing a particle swarm optimization algorithm, we construct a tight-binding model with inclusion of correlation effects through seeking appropriate renormalization factors of kinetic energies, which successfully reproduce the ARPES results at various temperatures, highlighting the importance of correlations in FeSe.

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I. INTRODUCTION

Iron-based superconductors (FeSCs) have attracted sustained scientific attention since their discovery [1–4]. FeSCs typically exhibit a phase diagram in which superconducting state emerges in proximity to nematic state [5–11]. Commonly, the superconducting transition temperature (T_c) reaches its maximum when nematicity is suppressed by either chemical doping or pressure, suggesting that nematicity plays a key role in establishing superconductivity. Especially, FeSe stands out among FeSCs for displaying nematicity at ambient conditions without long-range magnetic order [12–14], offering an ideal platform for studying the relationship between nematicity and superconductivity. However, a definitive conclusion about the electronic origin of its nematicity [15–18], whether spin [19–21], charge [22], orbital [23,24], degrees of freedom, or multipolar orders [25,26], has not yet been reached. And the description of energy spectrum obtained from angle-resolved photoemission spectroscopy (ARPES) experiments [16,17,27–46], which shows one elliptical hole pocket and two orthogonal electronic pockets around the center and corner of Brillouin zone (BZ), respectively, with one of the latter vanishing when the temperature is below 30 K [34,40,42–44], named the missing pocket, remains unsettled theoretically.

In current descriptions of nematicity, scientists primarily adopt two frameworks, spin order [19–21] and orbital order

[23,24]. In spin ordering scenarios, the introduction of magnetic order [47–50] inevitably induces orbital polarization, making it difficult to determine the hierarchical relationship between spin orders and orbital polarization. Additionally, such theories can hardly explain the experimental absence of long-range magnetic order that presents at moderate pressure [51–53] unambiguously. Orbital ordering approaches face greater challenges, since achieving orbital polarization without introducing spin order requires breaking conventional symmetry; for example, preconditioned wave function techniques [26,54] can construct pure orbital order but artificially introduce unobserved E-type order parameters, leading to predicted triclinic lattice distortions [55] that directly conflict with the experimentally observed orthorhombic to tetragonal transition [56]. To date, no unified framework has been established to systematically investigate the competition between spin order and orbital polarization.

In another phenomenological approach, nematic order parameters are introduced into C_4 symmetric tight-binding model (TBM) to generate a nematic state, with their specific values determined through comparison with ARPES band structures. Several order parameters have been proposed, including the parameters belonging to B_{1g} symmetry-breaking channel, such as ferro-orbital order (Φ_1) [57], d -wave (Φ_2) and extended- s -wave (Φ_3) bond orders [58], nonlocal d_{xy} nematic order (Φ_4) [59], unidirectional nematic bond order [31], and E-type order parameters [54,60]. The C_4 symmetric tight-binding model is typically parametrized from density functional theory (DFT) calculations [61] or high-temperature (HT) ARPES band structures [35]. Although these methods fit the experimental Fermi pockets well [59], they fail to describe the whole dispersion obtained from experiments due to the neglect of band renormalization effect caused by electronic correlations.

In this work, by employing LDA + U calculation, we systematically investigate the competition between spin order

*These authors contributed equally to this work.

†Contact author: yzzhang@tongji.edu.cn

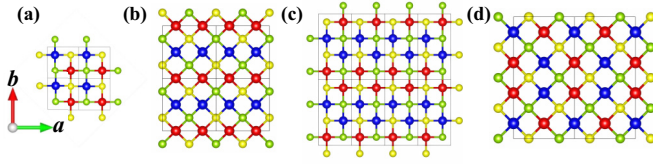


FIG. 1. Unit cells used in this work for (a) PM, AFM, (b) CAFM, (c) BAFM, and (d) PAFM, respectively. In panel (a), red and blue balls correspond to Fe1 and Fe2; in panels (b) and (c), they indicate spin-up and spin-down Fe atoms, respectively. The Fe atoms form a planar arrangement, while the Se atoms are distributed above (green) and below (yellow) this plane. $\mathbf{a}$ and $\mathbf{b}$ denote the basis vectors of lattice vectors in all cases.

and orbital polarization in FeSe to explain the absence of magnetism under ambient pressure and the emergence of stripe antiferromagnetic (AFM) order under moderate pressure. Our calculations indicate a Coulomb repulsion of approximately 4.5 eV within this system. We further demonstrate that either static spin order or static orbital polarization fails to describe the experimental band dispersions observed from ARPES studies, probably due to the underestimate of electronic correlation effects within the DFT method. To address this limitation, we develop a tight-binding model incorporating correlation effects through renormalized hopping parameters. Utilizing a particle swarm optimization (PSO) algorithm, we find optimal renormalization factors that accurately reconstruct the temperature-dependent ARPES band structures.

II. COMPUTATIONAL METHOD

We perform density functional theory calculations employing the full-potential linearized augmented plane-wave method as implemented in the WIEN2K code [62], where the exchange-correlation interactions are treated within the local-density approximation [63]. We incorporate on-site Coulomb repulsive interactions with around mean-field double-counting corrections [64], which is suitable for correlated metals [65,66]. Unless otherwise specified, the Hund's coupling is set to $J_H = U/10$ [67] in all our LDA + U calculations. To determine the magnetic ground state, we focused on following magnetic configurations, such as paramagnetic (PM), Néel antiferromagnetic, collinear antiferromagnetic (CAFM), bicollinear antiferromagnetic (BAFM), and pair-checkerboard antiferromagnetic (PAFM) orders, since Néel and stripe-type spin fluctuations are experimentally observed at ambient conditions [68], and CAFM order is detected under a moderate pressure [51,52]. Moreover, PAFM order is theoretically proposed as the ground state in FeSe [47,48], and BAFM order emerges in the classical mean-field phase diagrams [47]. The magnetic structures are illustrated in Fig. 1. We employ $\sqrt{2} \times \sqrt{2} \times 1$, $2 \times 2 \times 1$, and $2\sqrt{2} \times \sqrt{2} \times 1$ supercells for CAFM, BAFM, and PAFM, respectively, while using the primitive cell (PC) for other magnetic states. Brillouin zone integrations are performed using k -meshes of $30 \times 30 \times 12$ for the primitive cell, $22 \times 21 \times 22$ for CAFM, $15 \times 21 \times 30$ for BAFM, and $11 \times 21 \times 22$ for PAFM. The BZs for the primitive cell, CAFM, and PAFM configurations are illustrated in Fig. 2(a). We adopt the experimentally de-

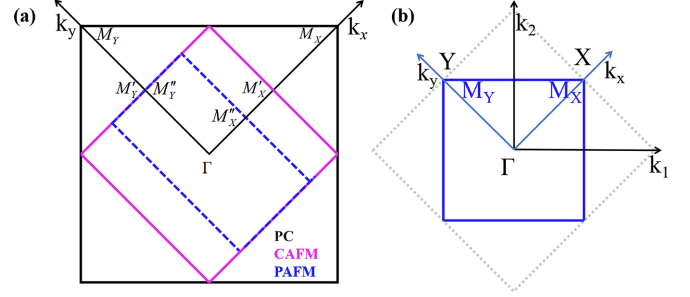


FIG. 2. (a) Brillouin zones used in DFT calculations: black, magenta, and blue represent the BZs for the PC, CAFM, and PAFM, respectively. (b) BZs for modeling: gray and blue correspond to the 1-Fe and 2-Fe unit cells, respectively.

termined lattice constants of the orthorhombic phase under various pressures, as reported in Ref. [69], and transform them into a tetragonal structure following the methodology outlined in Ref. [47]. This transformation is justified by the electronic origin of nematicity in the system.

In this study, we employ a tight-binding parametrization derived from DFT calculations on monolayer FeSe in the PM phase. The Hamiltonian is expressed as

$$\hat{H}_{\text{DFT}} = \sum_{\mathbf{k}\mathbf{R}\alpha\beta} t_{\mathbf{R}}^{\alpha\beta} \cdot e^{i\mathbf{k}\cdot\mathbf{R}} C_{\alpha}^{\dagger}(\mathbf{k}) C_{\beta}(\mathbf{k}), \quad (1)$$

where $t_{\mathbf{R}}^{\alpha\beta}$ represents the hopping parameters, which are obtained through a transformation from Bloch states to maximally localized Wannier functions using the WANNIER90 code [70,71]. Here, $\mathbf{R}$ denotes the lattice vectors connecting the hopping sites, while α and β represent ten 3d orbitals of two Fe atoms. The operators $C_{\alpha}^{\dagger}(\mathbf{k})$ and $C_{\alpha}(\mathbf{k})$ are the Fourier transforms of the creation and annihilation operators $C_{R\alpha}^{\dagger}$ and $C_{R\alpha}$, respectively, which act on the Wannier orbital α (spin indices are omitted for simplicity).

For tetragonal FeSe, the local coordinate axes of the two Fe atoms are set along Fe-Fe directions but rotated by 90° relative to each other due to the glide symmetry. However, ARPES measurements define the 3d orbitals with respect to a uniform local coordinate system $[k_x, k_y]$, as shown in Fig. 2(b) for both Fe atoms. To facilitate a direct comparison between the Wannier-derived band structure and experimental ARPES results, we align the local coordinate axes of both Fe atoms with the ARPES reference frame.

To incorporate the effects of electronic correlations, we introduce renormalization factors and modify the onsite energy according to the following expression:

$$\begin{aligned} \hat{H}_{\text{new}}^{\alpha\beta}(\mathbf{k}) = & Z_0^{\alpha\beta} \cdot (\hat{H}_{\text{DFT}}^{\alpha\beta}(\mathbf{k}) - \epsilon_{\text{DFT}}^{\alpha} \cdot \delta_{\alpha\beta}) + \epsilon_{\text{new}}^{\alpha} \cdot \delta_{\alpha\beta} \\ & + \sum_{\rho}^{\text{neighbor}} (Z_{\rho}^{\alpha\beta} - Z_0^{\alpha\beta}) \cdot t_{\rho}^{\alpha\beta} \cdot S(\rho), \end{aligned} \quad (2)$$

where $\hat{H}_{\text{DFT}}^{\alpha\beta}(\mathbf{k})$ ($\hat{H}_{\text{new}}^{\alpha\beta}(\mathbf{k})$) denotes the matrix elements of the DFT-derived (modified) Hamiltonian at momentum $\mathbf{k}$. The terms ϵ_{DFT} and ϵ_{new} represent the on-site energy from the DFT calculation and the modified Hamiltonian, respectively. The renormalization factors $Z_{\rho}^{\alpha\beta}$ adjust the hopping parameters $t_{\rho}^{\alpha\beta}$

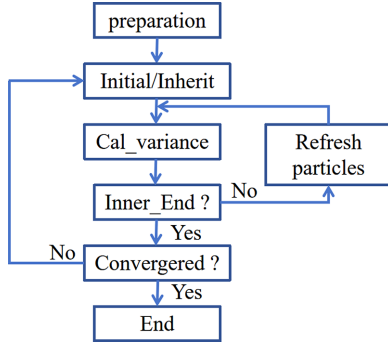


FIG. 3. Flowchart of the PSO algorithm. A double-loop search strategy is employed, where the inner loop identifies the best solution, which is then inherited by the subsequent outer loop until convergence is achieved.

between neighboring sites connected by ρ with a struct factor of $S(\rho)$ in 2-Fe unit cell, while $Z_0^{\alpha\beta}$ uniformly renormalizes the remaining hoppings unless otherwise specified. For diagonal elements ($\alpha = \beta$), the renormalization factors and hopping parameters are denoted as $Z_\rho^{\alpha\beta}$ and t_ρ^α , respectively. The term $Z_0^{\alpha\beta}$ is constrained to preserve C_4 symmetry. Hereafter, we denote $\mathbf{l}_1 = 1/2(\mathbf{a} + \mathbf{b})$ and $\mathbf{l}_2 = 1/2(\mathbf{a} - \mathbf{b})$, where $\mathbf{a}$ and $\mathbf{b}$ are the basis lattice vectors of primitive unit cell with two Fe atoms, as presented in Fig. 1(a). Then, the renormalization factors for nearest neighbors are $Z_{\mathbf{l}_1}^{\alpha\beta}$ and $Z_{\mathbf{l}_2}^{\alpha\beta}$, while the ones for next-nearest neighbors (NNNs) are $Z_{\text{NNN}}^{\alpha\beta}$.

To model the nematic state, we set $Z_\rho^{\alpha\beta}$ to be identical along the $\mathbf{a}$ and $\mathbf{b}$ directions but different along the $\mathbf{l}_1$ and $\mathbf{l}_2$ directions. Spin-orbit coupling (SOC) is included along the z axis [60] with a strength of $\lambda_{\text{SOC}} = 20$ meV, consistent with experimental findings [28]. The density of states (DOS) and orbital occupations are computed using a k -mesh of $1000 \times 1000 \times 1$. And the total number of electrons N is calculated within $N = \sum_{\mathbf{k}, n} f(E_n(\mathbf{k}) - \mu)$, where $E_n(\mathbf{k})$ is the energy of band n at momentum $\mathbf{k}$ and $f(E_n(\mathbf{k}) - \mu) = [1 + e^{(E_n(\mathbf{k}) - \mu)/(k_B T)}]^{-1}$ is the Fermi distribution function while μ is the chemical potential.

To determine the correct renormalization factors that yield a band structure consistent with experimental observations, we employ the PSO method, the flowchart of which is illustrated in Fig. 3. The procedure is outlined as follows:

(1) *Initialization.* Experimental data and DFT-derived parameters are prepared. A set of random particles, denoted as $\{X_1, \dots, X_{ip}, \dots, X_{Np}\}$, is initialized, where each particle $X_{ip} = \{\dots, Z_\rho^{\alpha\beta}, \dots, \epsilon_{\text{new}}, \dots\}$ represents a potential solution. Here, Np is the total number of particles. If a previously obtained high-quality solution exists, it can be incorporated into the initial particle set.

(2) *Fitness evaluation.* The variance (σ) between the band structure generated by the particles and the experimental band structure is calculated using the following expression:

$$\sigma = \sqrt{\frac{1}{N_k} \sum_{in} (\tilde{E}_n(k_i) - E_n(k_i))^2 + \Omega}, \quad (3)$$

where $\tilde{E}_n(k_i)$ and $E_n(k_i)$ are the n th eigenvalues of the searched Hamiltonian and the experimental Hamiltonian, re-

spectively, at the i th k -point along high-symmetry paths. N_k is the total number of such k -points. Ω is a penalty parameter that will be assigned a relatively large value when the shape of the Fermi surface or the dominant orbital components of the bands at high-symmetry points are not correctly reproduced.

(3) *Local and global optimization.* The local best position ($P_{p\text{best}}$) and the global best position ($P_{g\text{best}}$) are updated based on the variances computed in the current and previous iterations. If the current iteration number (t) is less than the preset maximum number of iterations (t_{max}), the particle velocities (v^{t+1}) are updated according to the PSO algorithm [72]:

$$v^{t+1} = \omega \cdot v^t + c_1 \cdot r_1 \cdot (P_{p\text{best}}^t - x^t) + c_2 \cdot r_2 \cdot (P_{g\text{best}}^t - x^t), \quad (4)$$

where $c_1 = 1.6$ and $c_2 = 1.8$ are positive constants, r_1 and r_2 are random numbers uniformly distributed in the range $[0, 1]$, and ω is the inertia weight that linearly decreases with iterations. We set $\omega_{\text{max}} = 2.0$ and $\omega_{\text{min}} = 0.4$. We retain the velocity direction of conventional PSO but implement step size control through velocity magnitude constraints by introducing a factor, γ , which adopts stage-specific values,

$$\gamma = \begin{cases} \gamma_{\text{max}}, & \text{if } t \leq t_{\text{max}}/3, \\ (\gamma_{\text{max}} + \gamma_{\text{min}})/2, & \text{if } t_{\text{max}}/3 < t \leq 2t_{\text{max}}/3, \\ \gamma_{\text{min}}, & \text{if } 2t_{\text{max}}/3 < t \leq t_{\text{max}}, \end{cases} \quad (5)$$

where $\gamma_{\text{max}} = 1.5$ and $\gamma_{\text{min}} = 0.01$. Then,

$$v^{t+1} = \gamma \cdot r_3 \cdot v^{t+1}, \quad (6)$$

where r_3 is another random number. Finally, the particle positions (x^{k+1}) are updated as follows:

$$x^{t+1} = x^t + v^{t+1}. \quad (7)$$

(4) *Termination of inner loop.* If the iteration limit is reached, the inner loop terminates, and the current global best position ($P_{g\text{best}}^t$) and its corresponding variance (σ^t) are carried over to the next outer loop as part of the initialized particles. This strategy enhances the likelihood of escaping local minima.

(5) *Convergence criteria.* The outer loop terminates when the convergence criteria are satisfied. The criteria are defined as

$$\Delta r = \|P_{g\text{best}}^t - P_{g\text{best}}^{t-1}\| \leq \sqrt{np} \times \delta, \quad (8)$$

$$\Delta \sigma = \sigma^t - \sigma^{t-1} \leq 10^{-7} \text{ eV}, \quad (9)$$

where np is the dimensionality of X_{ip} and $\delta = 10^{-4}$.

III. RESULTS

A. LDA + U on FeSe under ambient and pressure

We perform LDA + U calculations on FeSe under ambient conditions to investigate the evolution of its magnetic ground state with increasing on-site Coulomb interaction U (0–6.0 eV). Figure 4(a) shows total energies of all the magnetic states relative to that of PM state at each U , revealing that the ground state of FeSe undergoes a series of transitions, while Fig. 4(b) displays the magnetic moment (MM) per Fe atom and the orbital polarization (Δ) of the ground state. Here,

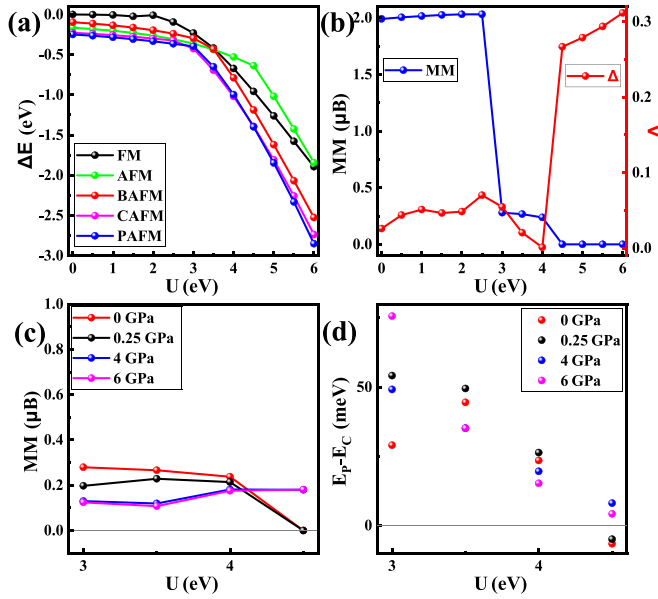


FIG. 4. LDA + U calculations under ambient and applied pressures. (a) Energy differences between magnetic states and the PM state. The FM, AFM, BAFM, CAFM, and PAFM states are represented by black, green, red, magenta, and blue balls, respectively. (b) Magnetic moment per Fe atom (blue balls) and orbital polarization between d_{xz} and d_{yz} orbitals (red balls) at ground state. Pressure-dependent LDA + U results: (c) magnetic moment per Fe atom and (d) ground-state energy relative to the CAFM state. Red, black, blue, and magenta balls correspond to ambient pressure, 0.25, 4, and 6 GPa, respectively.

$\Delta = |n_{xz} - n_{yz}|$, where n_{xz} and n_{yz} denote the electron occupations of the d_{xz} and d_{yz} orbitals, respectively. Specifically, the system undergoes transitions from a strong PAFM state, at $U = 0$ –2.5 eV, to a weak CAFM state, at $U = 3.0$ –4.0 eV, and finally to a strong orbital polarized state at $U \geq 4.5$ eV.

The orbital-resolved electron occupations are calculated as functions of U in Table I, where upward and downward arrows denote two spin species, respectively, while short horizontal dash indicates the absence of spin polarization. For $U \leq 2.5$ eV, electronic configurations obey the Hund's rule with parallel spins in all orbitals. Between $U = 2.5$ and 3.0 eV, the PAFM to CAFM transition occurs while the spin polarization of d_{xz} and d_{yz} orbitals inverts, breaking Hund's rule and sharply reducing the Fe magnetic moment as

TABLE I. Electron occupations for ground state of FeSe under different values of the on-site Coulomb repulsion U . Arrows in parentheses indicate the spin polarization directions of the corresponding orbitals, while the short horizontal dash signifies the absence of spin polarization.

U	d_{z^2}	d_{xz}	d_{yz}	$d_{x^2-y^2}$	d_{xy}
2.5 eV	1.332(↑)	1.138(↑)	1.068(↑)	1.356(↑)	1.239(↑)
3.0 eV	1.312(↑)	1.166(↓)	1.112(↓)	1.519(↑)	1.052(↑)
3.5 eV	1.325(↑)	1.166(↓)	1.112(↓)	1.551(↑)	1.040(↑)
4.0 eV	1.308(↑)	1.101(↓)	1.100(↓)	1.583(↑)	1.023(↑)
4.5 eV	1.480(–)	1.199(–)	0.932(–)	1.412(–)	1.073(–)

presented in Fig. 4(b). At $U \geq 4.5$ eV, all spin polarization disappears, resulting in a nonmagnetic state with strong orbital polarization that differs from the PM state. Between $U = 0$ and 2.5 eV, both orbital polarization and magnetic moment increase slightly with U . In the CAFM state ($U = 3.0$ –4.0 eV), orbital polarization weakens along with a gradual magnetic moment reduction, confirming spin order as the dominant driver of nematicity below $U = 4.0$ eV. At $U = 4.0$ eV, orbital polarization nearly vanishes ($\Delta \approx 0$) with a weak magnetic moment of $0.24 \mu_B$, indicating a nearly pure spin-ordered ground state. Above $U = 4.5$ eV, the system enters the strong orbital polarized state with vanishing magnetic moment. Thus, we achieve two ground states with either pure weak spin order or pure orbital polarization within a unified framework.

Our calculations reveal that the transition from CAFM to strong orbital polarized state occurs between $U = 4.0$ and 4.5 eV, corresponding to an effective Coulomb repulsion U_{eff} of 3.6–4.05 eV. Previous random phase approximation studies [67] estimate the U_{eff} in FeSe as ~ 4.24 eV, suggesting that the ground state of the system is likely dominated by the strongly orbital polarization.

Pressure-dependent calculations, as presented in Figs. 4(c) and 4(d), where total energies of PAFM state are relative to that of CAFM state at each U , reveal that at $U = 3.0$ –4.0 eV, weak CAFM persists under low to moderate pressures (0–6 GPa). At $U = 4.5$ eV, strong orbital polarized state with zero magnetic moment emerges under low pressure (0.25 GPa), while weak CAFM state reappears under moderate pressure (4 and 6 GPa). Experimentally [51–53], FeSe shows no magnetic moment below ~ 0.8 GPa and weak CAFM at 0.8–8 GPa. The experimental observations agree with our calculations at $U = 4.5$ eV, indicating competition between the strong orbital polarized and CAFM states, and suggesting the on-site Coulomb repulsion in FeSe proximity to $U \sim 4.5$ eV. It was proposed that the simultaneous excitation of different types of spin fluctuations in FeSe prevents their condensation at any wave vector, leading to a nonmagnetic ground state [47] under ambient pressure. In our calculations, orbital polarization prevails over spin order, stabilizing the nonmagnetic strong orbital polarized state and resulting in the experimental absence of magnetic order under ambient pressure.

Our calculations reveal that the ground state of FeSe under ambient pressure lies near the phase boundary between weak CAFM and strong orbital polarized state. To investigate its electronic structure, we calculate the band structures near this phase boundary at $U = 4.0$ and 4.5 eV, as displayed in Fig. 5. In the purely spin-ordered state, magnetic order induces band splitting—even weak magnetic order opens a large band gap as presented in Fig. 5(a). Previous theoretical studies also found that DFT calculations [48,50] incorporating magnetic order yield band structures inconsistent with experiments, indicating that static spin order is not the primary factor shaping the unique electronic structure of FeSe. In the state with purely orbital polarization as displayed in Fig. 5(b), bands along the Γ – M_x'' direction shift above the Fermi level, eliminating the hole pocket around Γ point, showing a clear discrepancy with experiments, likely due to overestimated orbital polarization. Experimentally, the saturated nematic orbital polarization is

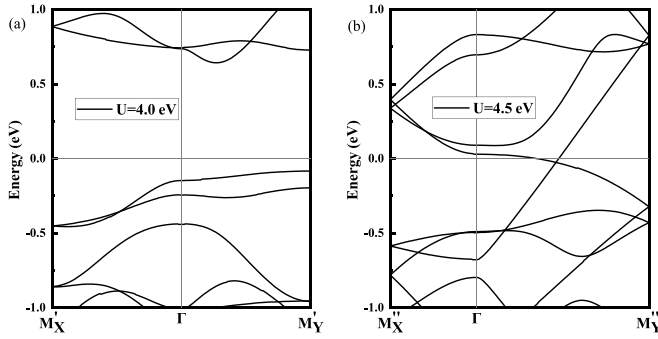


FIG. 5. Band structure of FeSe at the ground state from DFT + U calculations with (a) $U = 4.0$ eV and (b) $U = 4.5$ eV. The high-symmetry path is illustrated in Fig. 2(a).

~ 0.035 [24], whereas our calculation at $U = 4.5$ eV yields an orbital polarization of 0.267, far exceeding experimental values. This discrepancy likely stems from the neglect of correlation effects within the DFT method. In the next section, we will introduce correlation effects into the tight-binding model to suppress orbital polarization.

B. Fit nematic state at moderate temperature

To accurately reproduce band structure of the nematic phase at the moderate temperature (MT, ~ 60 K), we introduce orbital selectivity and nematicity into the renormalization of hopping parameters. For simplicity, we introduce anisotropy in renormalization factors for the nearest-neighbor hoppings, while all further hoppings are uniformly renormalized by Z_0 . Here, Z_0 satisfies C_4 symmetry with the following constraints: $Z_0^{xz,xz} = Z_0^{yz,yz}$, $Z_0^{z^2,xz} = Z_0^{z^2,yz}$, $Z_0^{xz,x^2-y^2} = Z_0^{yz,x^2-y^2}$, $Z_0^{xz,xy} = Z_0^{yz,xy}$, along with $Z_0^{\alpha\beta} = Z_0^{\beta\alpha}$. Under the assumption of isotropy for Z_0 , 11 independent nonzero parameters remain in Z_0 .

For hoppings from nearest neighbors, given d_{z^2} and $d_{x^2-y^2}$ bands far from the Fermi level, their renormalization is also assumed to be isotropic, which means $Z_{t_1}^{z^2} = Z_{t_2}^{z^2}$, $Z_{t_1}^{x^2-y^2} = Z_{t_2}^{x^2-y^2}$, and $Z_{t_1}^{z^2,x^2-y^2} = Z_{t_2}^{z^2,x^2-y^2}$, resulting in 15 nonzero parameters, rather than original 18 parameters (9 for single direction [61]). To better match experimental observations, we further set $Z_{t_1}^{yz,xy} \neq Z_{t_1}^{xy,xz}$ and $Z_{t_2}^{yz,xy} \neq Z_{t_2}^{xy,yz}$, which would reproduce the gap indicated by pink box in Fig. 6(a), thereby total renormalization parameters for nearest-neighbor hoppings becomes 17.

Additionally, the on-site energies for d_{xz} and d_{yz} orbitals are constrained as $\epsilon_{xz} = \epsilon_{yz}$, requiring four independent on-site energy parameters. Combining these, the total 32 independent nonzero parameters are employed to fit the ARPES bands at moderate temperature. And the renormalization factors are determined as presented in Table II.

The renormalization factors are determined by fitting the renormalized band dispersion and orbital weights along the high-symmetry path of $M_Y - \Gamma - M_X$ to the experimentally derived bands with specified orbital characters as overlaid in Fig. 2 of Ref. [40]. Although the experimental energy window does not include information on the $d_{x^2-y^2}$ orbital, there are implicit constraints between this orbital and the other three crucial orbitals— d_{xz} , d_{yz} , and d_{xy} . Accordingly, the renormal-

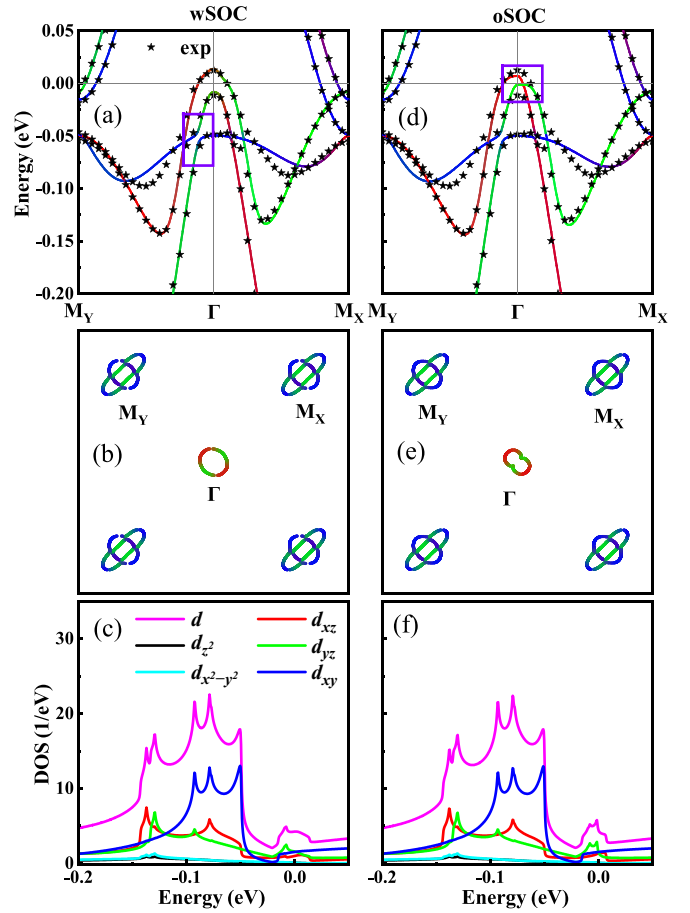


FIG. 6. The fitting results with and without spin-orbit coupling at moderate temperature. (a) Band structure, (b) Fermi surfaces, and (c) density of states of FeSe at MT states with SOC. Band structure is compared with experimental data from Ref. [40]. (d), (e) Results without SOC, derived from the calculations in panels (a)–(c), respectively. Magenta, black, cyan, red, green, and blue correspond to total $3d$, d_{z^2} , $d_{x^2-y^2}$, d_{xz} , d_{yz} , and d_{xy} orbitals, respectively.

ization factors for the $d_{x^2-y^2}$, d_{xz} , d_{yz} , and d_{xy} orbitals are optimally determined. To verify this, we do additional tests: We either increase or decrease the renormalization factor of the $d_{x^2-y^2}$ orbital by multiplying a factor of 1.2 or 0.8, respectively. In each case, the band structure and the size of Fermi surface around Γ are notably affected as illustrated in Fig. 7. Given the quasi-two-dimensional nature of FeSe and the experimental data from ARPES primarily in the Γ plane [40], we perform our calculations on a monolayer FeSe. The Fermi surface around Γ is constrained to an elliptical shape with a tolerance of 1%.

The fitting results for the MT state are presented in Fig. 6(a). The dispersion of bands associated with the d_{xz} and d_{yz} orbitals is well reproduced, while deviations are observed for the d_{xy} band, which is although the most accurate description as far as we know. The gap, indicated by pink box in Fig. 6(a), arising from the crossing of the d_{xy} band with the inner d_{yz} bands is not due to spin-orbit coupling but rather results from the folding effect induced by the inequivalence of the two Fe atoms [73] as found in scanning tunneling microscopy experiment [74]. However, the inner

TABLE II. The renormalization factors obtained from fitting the band structure of FeSe at moderate temperature, including (a) on-site energies ϵ , (b) Z_0 , (c) Z_{I_1} , and (d) Z_{I_2} .

(a)	d_{z^2}	d_{xz}	d_{yz}	$d_{x^2-y^2}$	d_{xy}
ϵ	-0.1683	0.0324	0.0324	-0.2036	0.0044
(b)	d_{z^2}	d_{xz}	d_{yz}	$d_{x^2-y^2}$	d_{xy}
d_{z^2}	0.9355	0.5228	0.5228	0.4428	0.9040
d_{xz}	H.c.	0.3363	0.5076	0.4090	0.1022
d_{yz}	H.c.	H.c.	0.3363	0.4090	0.1022
$d_{x^2-y^2}$	H.c.	H.c.	H.c.	0.7724	0.4868
d_{xy}	H.c.	H.c.	H.c.	H.c.	0.0941
(c)	d_{z^2}	d_{xz}	d_{yz}	$d_{x^2-y^2}$	d_{xy}
d_{z^2}	0.7478	0	0.4864	0.8864	0
d_{xz}	0	0.7011	0	0	0.2182
d_{yz}	H.c.	0	0.4450	0.4519	0
$d_{x^2-y^2}$	H.c.	0	H.c.	0.4737	0
d_{xy}	0	0.1959	0	0	0.7299
(d)	d_{z^2}	d_{xz}	d_{yz}	$d_{x^2-y^2}$	d_{xy}
d_{z^2}	0.7478	0.5122	0	0.8864	0
d_{xz}	H.c.	0.4050	0	0.4613	0
d_{yz}	0	0	0.5622	0	0.2458
$d_{x^2-y^2}$	H.c.	H.c.	0	0.4737	0
d_{xy}	0	0	0.1848	0	0.5012

electron band near the M_X point [Fig. 6(a)] exhibits a reduced d_{xz} orbital weight compared to experimental data [40]. The Fermi surfaces, as illustrated in Fig. 6(b), accurately reproduce the shrinkage of electron pockets around M . As shown in Fig. 6(c), the density of states near the Fermi energy is primarily contributed by the d_{xz} , d_{yz} , and d_{xy} orbitals, suggesting that a low-energy three-orbital model is suitable for describing FeSe [59,75].

The Hamiltonian describing the nematic phase is expressed as

$$H = H^{\text{HT}} + H_{\text{nem}}^T + H_{\text{cor}}^T + H_{\text{SOC}}. \quad (10)$$

Here, H^{HT} is the Hamiltonian describing HT phase and H_{cor}^T describes the temperature-dependent corrections that do not introduce symmetry breaking and its form will be discussed in

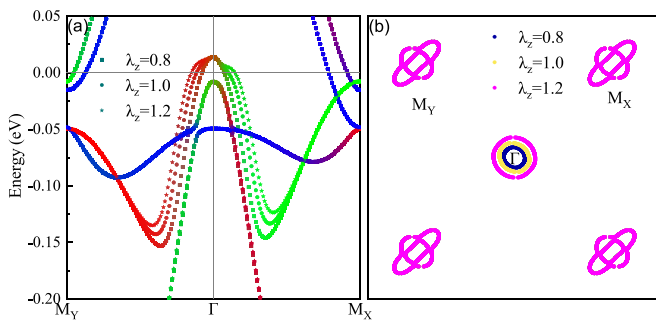


FIG. 7. The influence of the renormalization factor for the $d_{x^2-y^2}$ orbital on the electronic structure. (a) Band structures with the renormalization factors scaled by factors of $\lambda_z = 0.8$ (squares), $\lambda_z = 1.0$ (circles), and $\lambda_z = 1.2$ (pentagons), respectively. (b) Fermi surfaces for the same set of scaled renormalization factors, $\lambda_z = 0.8$ (dark blue), $\lambda_z = 1.0$ (dark yellow), and $\lambda_z = 1.2$ (magenta).

next section where H^{HT} is determined. H_{SOC} represents SOC following the form in Ref. [76]. H_{nem}^T is the nematic items that break the C_4 symmetry, which is also temperature dependent.

Our investigation further clarifies the impact of SOC on the electronic structure of FeSe. By excluding SOC from the MT solution, we observe in Fig. 6(d) that the d_{xz} and d_{yz} orbitals at the Γ point are energetically separated with a gap opened by the nematic order [58]. Notably, upon inclusion of SOC, a band crossing, highlighted in the purple box of Fig. 6(d), between the d_{xz} and d_{yz} bands disappears [see Fig. 6(a)]. This crossing gapped by SOC occurs near the Fermi level, thereby inducing significant modifications to the Fermi surface topology around the Γ point [40]. DOS analysis as displayed in Figs. 6(c) and 6(f) reveals that SOC suppresses the density of states near the Fermi level for both the d_{xz} and d_{yz} orbitals as predicted by Ref. [76].

All possible nematic order parameters within the B_{1g} irreducible representation of the $P4/nmm$ space group involving interactions between d_{xz} , d_{yz} , and d_{xy} orbitals within a single 2-Fe unit cell are Φ_1 – Φ_4 [59]. Based on their formalism in real space [59], H_{nem}^T is expressed as

$$H_{\text{nem}}^T = \sum_R \left\{ \frac{\Phi_1}{2} (C_{R,xz}^\dagger C_{R,xz} - C_{R,yz}^\dagger C_{R,yz}) + \frac{\Phi_2 + \Phi_3}{4} C_{R,xz}^\dagger C_{R+I_1,xz} + \frac{\Phi_2 - \Phi_3}{4} C_{R,yz}^\dagger C_{R+I_1,yz} - \frac{\Phi_2 - \Phi_3}{4} C_{R,xz}^\dagger C_{R+I_2,xz} - \frac{\Phi_2 + \Phi_3}{4} C_{R,yz}^\dagger C_{R+I_2,yz} + \frac{\Phi_4}{2} (C_{R,xy}^\dagger C_{R+I_1,xy} - C_{R,xy}^\dagger C_{R+I_2,xy}) + \text{H.c.} \right\}. \quad (11)$$

Here, only the anisotropy of hoppings from nearest neighbors is considered. Thus, the nematic order parameters, Φ_1 – Φ_4 can be determined as

$$\Phi_1 = \epsilon^{xz} - \epsilon^{yz}, \quad (12)$$

$$\Phi_2 = t_{I_1}^{xz} + t_{I_1}^{yz} - t_{I_2}^{xz} - t_{I_2}^{yz}, \quad (13)$$

$$\Phi_3 = t_{I_1}^{xz} + t_{I_2}^{xz} - t_{I_1}^{yz} - t_{I_2}^{yz}, \quad (14)$$

$$\Phi_4 = t_{I_1}^{xy} - t_{I_2}^{xy}. \quad (15)$$

The calculated nematic order parameters are $\Phi_1 = 0$, $\Phi_2 = 20.9$ meV, $\Phi_3 = -4.3$ meV, and $\Phi_4 = -8.3$ meV. E-type nematic order parameters, proposed by both DFT [54] and TBM studies [60], would induce a structural transition from the tetragonal to the monoclinic phase, which has not been observed in FeSe [55,56]. Therefore, this type of order parameter is excluded here. Additionally, $\Phi_1 = 0$ indicates that ferro-orbital order is not the origin of nematicity. And the corrections on hopping parameters due to the temperature effect will be determined in next section where the high-temperature phase is identified.

The electron occupations of this state are shown in Table III. The averaged orbital polarization Δ is 0.041 per Fe atom at MT, which is slightly higher than the value of 0.026 at low temperature (LT), as discussed in Sec. III D. This is consistent with the experimental observation of orbital polarization saturated at approximately 0.035 near 60 K, just

TABLE III. The electron occupations on each orbital of the states identified at different temperatures corresponds to the high-temperature, moderate-temperature, and low-temperature regimes, respectively.

Occ	d_{z^2}	d_{xz}	d_{yz}	$d_{x^2-y^2}$	d_{xy}
HT	1.395	1.134	1.134	1.215	1.168
MT	1.378	1.204	1.163	1.202	1.073
LT	1.378	1.219	1.245	1.188	0.994

below the structural transition temperature T_s [24]. In contrast to the DFT-derived ground state with strong orbital polarization at $U = 4.5$ eV, our calculations incorporating electronic correlations reveal a redistribution of electrons from the d_{z^2} and $d_{x^2-y^2}$ orbitals to the d_{yz} orbital, effectively suppressing orbital polarization. This mechanism enables us to successfully reconstruct the band structure of nematic FeSe with unprecedented accuracy.

The critical hopping parameters at the moderate temperature, including the on-site energies of the d_{xz} , d_{yz} , and d_{xy} orbitals, together with their intraorbital hoppings from nearest neighbors and next-nearest neighbors, are listed in Table IV.

To further verify whether the validity of the model we obtained, we calculate the dependence of ARPES intensity on different polarization of incident light. The details of the calculation are described in the Appendix. As showing in Fig. 8, the calculated ARPES signals are consistent with the experimental results [40] based on the selection rules [77].

C. Fit tetragonal state at high temperature

The high-temperature (~ 120 K) state serves as a suitable starting point for modeling the nematic phase of FeSe. Both standard density functional theory [29] and DFT combined with dynamical mean-field theory (DFT + DMFT) [78–80] calculations tend to overestimate the band dispersion and bandwidth, resulting in Fermi surfaces that are significantly larger than those observed experimentally. Despite extensive efforts [35,59,61,81] to develop precise tight-binding models that circumvent the shortcomings of *ab initio* approaches, an accurate phenomenological description of the band structure of FeSe is lacking. In this work, we adjust a minimal set of parameters based on the solution for the MT phase to construct a band structure for the HT phase that agrees with experimental data [40].

TABLE IV. The key parameters of the tight-binding model across various temperature regimes, including the on-site energy (ϵ) of the d_{xz} , d_{yz} , and d_{xy} orbitals, and intraorbital hoppings between nearest neighbors and next-nearest neighbors among d_{xz} , d_{yz} , and d_{xy} orbitals, respectively (all in eV).

High temperature				Moderate temperature				Low temperature			
Orbitals	d_{xz}	d_{yz}	d_{xy}	Orbitals	d_{xz}	d_{yz}	d_{xy}	Orbitals	d_{xz}	d_{yz}	d_{xy}
ϵ	0.0740	0.0740	-0.0172	ϵ	0.0324	0.0324	0.0044	ϵ	0.0483	-0.0484	0.0192
$t_{l_1}^{xz}$	$t_{l_1}^{yz}$	$t_{l_2}^{xz}$	$t_{l_2}^{yz}$	$t_{l_1}^{xz}$	$t_{l_1}^{yz}$	$t_{l_2}^{xz}$	$t_{l_2}^{yz}$	$t_{l_1}^{xz}$	$t_{l_1}^{yz}$	$t_{l_2}^{xz}$	$t_{l_2}^{yz}$
0.0599	0.1344	0.1344	0.0599	0.0420	0.1401	0.1275	0.0337	0.0552	0.1502	0.1349	0.0000
$t_{NNN}^{xz/yz}$	$t_{l_1}^{xy}$	$t_{l_2}^{xy}$	t_{NNN}^{xy}	$t_{NNN}^{xz/yz}$	$t_{l_1}^{xy}$	$t_{l_2}^{xy}$	t_{NNN}^{xy}	$t_{NNN}^{xz/yz}$	$t_{l_1}^{xy}$	$t_{l_2}^{xy}$	t_{NNN}^{xy}
0.0756	-0.0151	-0.0151	0.0069	0.0756	-0.0266	-0.0183	0.0076	0.0791	-0.0364	-0.0074	0.0000

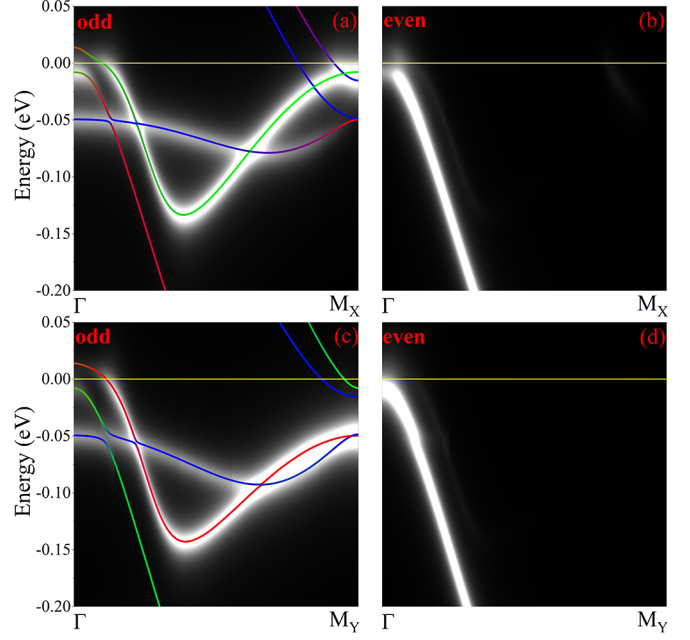


FIG. 8. Dependence of ARPES signals on the polarization of incident light. (a), (b) ARPES signal along Γ - M_X calculated with odd and even polarization, respectively. (c), (d) ARPES signal along Γ - M_Y calculated with odd and even polarization, respectively.

Based on the moderate-temperature phase identified in the previous section, we adjust a minimal set of parameters and employ the PSO method to search for the high-temperature phase. In contrast to the nematic phase at moderate temperatures, where nematicity originates from modifications to nearest-neighbor hoppings, we set $Z_{l_1}^{xz} = Z_{l_2}^{yz}$, $Z_{l_1}^{yz} = Z_{l_2}^{xz}$, $Z_{l_1}^{xy} = Z_{l_2}^{xy}$, and $Z_{l_1}^{xz,xy} = Z_{l_2}^{xy,xz} = Z_{l_2}^{yz,xy} = Z_{l_1}^{xy,yz}$, to enforce isotropy at HT. Simultaneously, we release the on-site energy parameters for $d_{xz/yz}$ and d_{xy} orbitals during the PSO search. This procedure introduces six additional free parameters. Subsequent investigations reveal that next-nearest-neighbor intraorbital hoppings of the d_{xy} orbital affect significantly the bands around the M point. Consequently, we further introduce one free parameter, Z_{NNN}^{xy} . By optimizing these seven parameters, we successfully fit the band structure at high temperature as shown in Fig. 9(a).

Then, the seven independent parameters are determined as $Z_{l_1}^{xz} = Z_{l_2}^{yz} = 1.0000$, $Z_{l_1}^{yz} = Z_{l_2}^{xz} = 0.4270$,

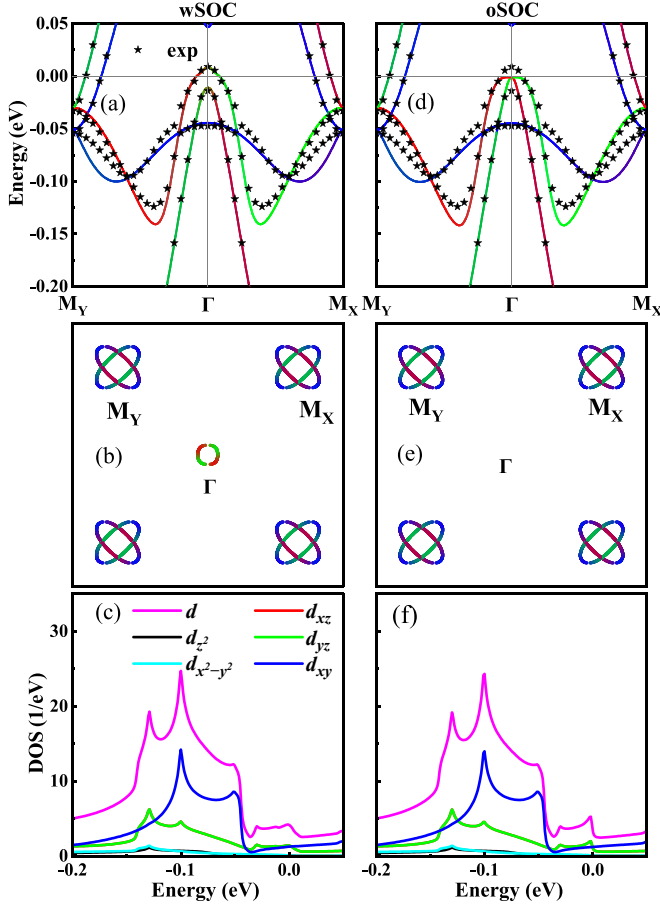


FIG. 9. The fitting results with and without spin-orbit coupling at high temperature. (a) Band structure, (b) Fermi surfaces, and (c) density of states of FeSe at HT state with SOC. Band structure is compared with experimental data from Ref. [40]. (d), (e) Results without SOC, derived from the calculations in panels (a)–(c), respectively. Magenta, black, cyan, red, green, and blue correspond to total $3d$, d_{z^2} , $d_{x^2-y^2}$, d_{xz} , d_{yz} , and d_{xy} orbitals, respectively.

$$Z_{l_1}^{xy} = Z_{l_2}^{xy} = 0.4155, \quad \epsilon_{xz} = \epsilon_{yz} = 0.0740, \quad \epsilon_{xy} = -0.1683, \\ Z_{l_1}^{xz,xy} = Z_{l_1}^{xy,xz} = Z_{l_2}^{yz,xy} = Z_{l_2}^{xy,yz} = 0.2320, \text{ and } Z_{\text{NNN}}^{xy} = 0.0863.$$

We also investigate the influence of SOC on the electronic structure of FeSe at this specific temperature. Due to the absence of nematic order, the band gap at Γ [Fig. 9(a)] opened by SOC precisely matches the SOC strength [58]. Contrary to previous understanding [58], SOC does not suppress one of the two hole pockets at the Γ point below the Fermi level to form a single hole pocket. Instead, it elevates one band above the Fermi level, thereby generating a hole Fermi pocket, as shown in Figs. 9(a), 9(b), 9(d), and 9(e). DOS analysis reveals that SOC suppresses the density of states near the Fermi level for both the d_{xz} and d_{yz} orbitals, as presented in Figs. 9(c) and 9(f).

Then, H_{cor}^T can be expressed as follows:

$$H_{\text{cor}}^T = \sum_{\alpha,i} \delta\epsilon_{\alpha} C_{i,\alpha}^{\dagger} C_{i,\alpha} + \sum_{\alpha\beta, \langle i,j \rangle} \delta t_{ij}^{\alpha\beta} C_{i,\alpha}^{\dagger} C_{j,\beta} + \text{H.c.}, \quad (16)$$

where $\delta\epsilon$ and δt denote corrections for onsite energies and hoppings between neighbor sites i and j , and $\alpha(\beta)$ denotes the

$3d$ orbitals. Here, we only analyze the intraorbital hoppings between the nearest neighbors involving d_{xz} , d_{yz} , and d_{xy} orbitals and the intraorbital hoppings between the next-nearest-neighbor d_{xy} orbitals. And the corrections induced by nematic parameters, Φ_1 – Φ_4 , and $H_{\text{cor}}^{\text{MT}}$ at moderate temperature can be identified, which are presented in Table V. As expected, $H_{\text{cor}}^{\text{MT}}$ preserves C_4 symmetry and is not negligible compared with corrections induced by nematic order parameters.

D. Fit nematic state at low temperature

The nematic state at low temperature (~ 15 K), characterized by a single electronic pocket around the M point, is crucial for unraveling the mechanisms for nematicity and superconductivity in FeSe [55]. In this section, we follow the same procedure as in the previous section but preserve nematicity by enforcing $Z_{l_1}^{xz} \neq Z_{l_2}^{yz}$, $Z_{l_1}^{yz} \neq Z_{l_2}^{xz}$, $Z_{l_1}^{xy} \neq Z_{l_2}^{xy}$, and $Z_{l_1}^{xz,xy} \neq Z_{l_1}^{xy,xz} \neq Z_{l_2}^{yz,xy} \neq Z_{l_2}^{xy,yz}$, resulting in ten independent parameters. We also include three on-site energy parameters for d_{xz} , d_{yz} , and d_{xy} orbitals and one renormalization factor parameter for the next-nearest-neighbor intraorbital hoppings of d_{xz} and d_{yz} orbitals ($Z_{\text{NNN}}^{xz} = Z_{\text{NNN}}^{yz}$). Additionally, renormalization factors for the next-nearest-neighbor intraorbital hoppings of d_{xy} orbital (Z_{NNN}^{xy}) are introduced due to its significant modulation of bands near M , yielding a total of 15 free parameters for the LT phase.

Through PSO optimization, we successfully identify the LT phase matching experimental observations. The 15 free parameters are determined as $Z_{l_1}^{xz} = 0.9210$, $Z_{l_2}^{yz} = 0.0007$, $Z_{l_1}^{yz} = 0.4776$, $Z_{l_2}^{xz} = 0.4287$, $Z_{l_1}^{xy} = 0.9995$, $Z_{l_2}^{xy} = 0.2019$, $Z_{l_1}^{xz,xy} = 0.3360$, $Z_{l_1}^{xy,xz} = 0.2710$, $Z_{l_2}^{yz,xy} = 0.2441$, $Z_{l_2}^{xy,yz} = 0.1325$, $\epsilon_{xz} = 0.0483$, $\epsilon_{yz} = -0.0484$, $\epsilon_{xy} = 0.0192$, $Z_{\text{NNN}}^{xz} = Z_{\text{NNN}}^{yz} = 0.3520$, and $Z_{\text{NNN}}^{xy} = 0$.

As shown in the Figs. 10(a) and 10(b), the calculated band structures match experimental observations and yield the Fermi surface with single electron pocket. The critical hopping parameters and parameter variations induced by order parameters and the temperature-dependent corrections $H_{\text{cor}}^{\text{LT}}$ are presented in the Tables IV and V, respectively. And order parameter magnitudes are determined as $\Phi_1 = 96.7$ meV, $\Phi_2 = 70.5$ meV, $\Phi_3 = 39.9$ meV, and $\Phi_4 = -29$ meV. Our calculations of electron occupations at this temperature (Table III) reveal a weaker orbital polarization between d_{xz} and d_{yz} orbitals compared to the moderate-temperature phase. This finding is consistent with experiments reporting a saturation of orbital polarization near 60 K [24].

It is worth mentioning that the charge occupancy of the d_{xz} orbital is higher than that of the d_{yz} orbital in the MT phase, which is expected to yield a negative signal in x-ray linear dichroism (XLD) measurements, while the relative charge occupancy is reversed in the LT phase, resulting in a positive XLD signal, as also observed [24,42]. This phenomenon is also captured in our model.

The impacts of SOC as presented in Figs. 10(a)–10(f) are the same as we discussed in Sec. III B. Comparisons of the fitting results for the high-temperature and low-temperature phases with independent experimental data [42] are shown in Fig. 11. Good agreement indicates effectiveness of our modeling.

TABLE V. Corrections to the key parameters of the tight-binding model by H_{corr}^T , at (a) moderate temperature and (b) low temperature, respectively, where values in parentheses denote corrections originating from the order parameters (all in eV).

(a)	$\delta\epsilon_{xz/yz}$	$\delta\epsilon_{xy}$	$\delta t_{\text{NN}}^{xy}$	$\delta t_{\text{NNN}}^{xy}$
MT	$-0.0416(\pm 0.0000)$	$0.0216(0.0000)$	$-0.0074(\pm 0.0042)$	$0.0007(\pm 0.0000)$
	$\delta t_{t_1}^{xz}$	$\delta t_{t_1}^{yz}$	$\delta t_{t_2}^{xz}$	$\delta t_{t_2}^{yz}$
	$-0.0221(0.0042)$	$-0.0006(0.0063)$	$-0.0006(-0.0063)$	$-0.0221(-0.0042)$
(b)	$\delta\epsilon_{xz/yz}$	$\delta\epsilon_{xy}$	$\delta t_{\text{NN}}^{xy}$	$\delta t_{\text{NNN}}^{xy}$
LT	$-0.0741(\pm 0.0484)$	$0.0364(0.0000)$	$0.0068(\pm 0.0145)$	$-0.0069(\pm 0.0000)$
	$\delta t_{t_1}^{xz}$	$\delta t_{t_1}^{yz}$	$\delta t_{t_2}^{xz}$	$\delta t_{t_2}^{yz}$
	$-0.0323(0.0276)$	$0.0082(0.0077)$	$0.0082(-0.0077)$	$-0.0323(-0.0276)$

IV. DISCUSSION

Our DFT calculations reveal the limitation of static spin order and static orbital polarization in reconstructing the band structure of FeSe, thereby conclusively establishing the indispensable role of electronic correlations in governing its

electronic structure. In this study, we systematically incorporate electronic correlations via renormalization of hopping parameters in the tight-binding model for paramagnetic FeSe and explicitly include spin-orbit coupling. This methodology yields the most accurate band structure of FeSe across three distinct temperature regimes achieved to date.

Previous attempts to describe the electronic structure of FeSe introduced orbitally selective renormalization factors [82–84], yet these efforts failed to adequately reproduce the band structure and could not capture the experimental observation of the single electron pocket. In contrast, theoretical calculations incorporating nonlocal interactions have partially reproduced such phenomenon [60]. In our work, we have gone beyond the existing orbital-dependent treatment of electronic correlations by further incorporating their spatial dependence—including both distance and directional dependence. This comprehensive approach accurately reproduces both the experimental band structure and Fermi surface, thereby demonstrating the essential role of spatially resolved electronic correlations in this system. However, a more complete description of the experimental observations would require the inclusion of dynamical correlations, which remains a focus for future work.

A comparison between Figs. 6(b) and 6(e) reveals significant changes in the Fermi pocket area. This primarily occurs because the relatively flat bands near the Γ point in the absence of spin-orbit coupling—although lying below the Fermi level—still contribute sufficient hole carriers due to thermal broadening effects, thereby maintaining occupation number conservation. At other temperatures, such as both high-temperature (~ 120 K) and low-temperature (~ 15 K) regimes, the thermal broadening effect can also induce noticeable variations in the Fermi pocket area.

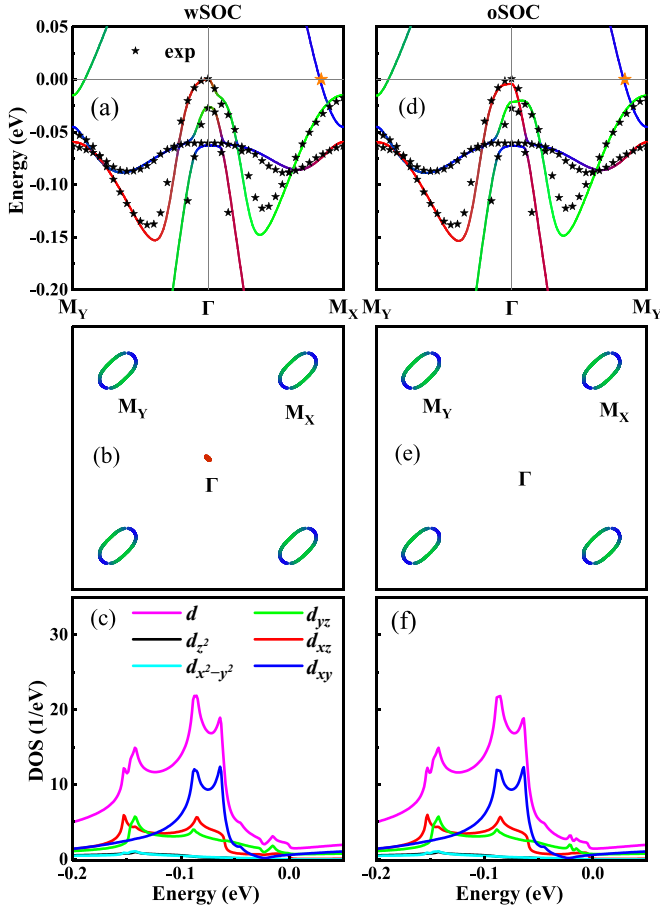


FIG. 10. The fitting results with and without spin-orbit coupling at low temperature. (a) Band structure, (b) Fermi surfaces, and (c) density of states of FeSe at LT state with SOC. The calculated band structure is benchmarked against experimental data from Ref. [41], with an additional data point (yellow) extracted from Ref. [42]. (d), (e) Results without SOC, derived from the calculations in panels (a)–(c), respectively. Magenta, black, cyan, red, green, and blue correspond to total 3d, d_{z^2} , $d_{x^2-y^2}$, d_{xz} , d_{yz} , and d_{xy} orbitals, respectively.

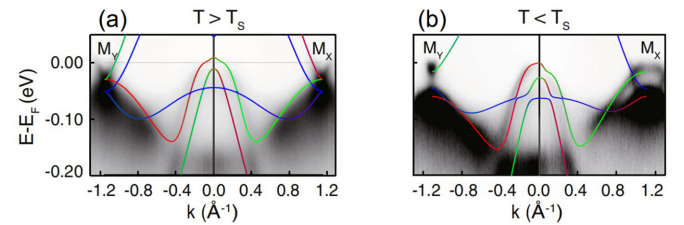


FIG. 11. Comparison between our calculated band structure and ARPES experiments from Ref. [42] at (a) high temperature and (b) low temperature, respectively.

By comparing the occupation of the DFT-derived ground state with that of the TBM identified at moderate temperature, we observe that correlation effects induce electron transfer from d_{z^2} and $d_{x^2-y^2}$ to d_{yz} orbitals, thereby reducing the overestimated orbital polarization in DFT calculations. As shown in Table III, during the nematic transition from high to moderate temperature, the occupation of the d_{xy} orbital decreases significantly, while electrons populate the d_{xz} and d_{yz} orbitals. This coincides with orbital polarization between d_{xz} and d_{yz} orbitals, likely driven by orbital-selective correlations. At the moderate-temperature phase, this polarization is saturated and remains nearly constant on cooling to the low-temperature phase. Between the moderate- and low-temperature phases, however, the anisotropy of nearest-neighbor intraorbital d_{xy} hoppings increases progressively (Table IV), ultimately lifting one electron pocket above the Fermi level to form the missing-pocket phenomenon, consistent with previous theory [59].

The description of nematic band structure requires the introduction of symmetry-breaking order parameters (Φ_1 – Φ_3). However, these parameters fail to fully account for both the ^{57}Fe NMR anomalies [85] and the missing pocket phenomenon. To resolve this inconsistency, a nonlocal d_{xy} nematic order parameter (Φ_4) is involved, derived from d -wave nematic interactions in the d_{xy} channel [60,86]. While Φ_4 successfully reconstructs the missing pocket [59], it concomitantly induces an undesirable bandwidth renormalization of the d_{xy} orbital. Compensation through a Hartree-like energy shift of the d_{xy} orbital, probably due to repopulation of electrons among orbitals, becomes essential to maintain the van Hove singularity near the M point at experimentally observed energies [55,59]. This adjustment, however, introduces a discrepancy between the calculated and measured d_{xy} band energies at the Γ point. Such unphysical outcomes may originate from the neglect of temperature-dependent hopping corrections governed by the second term in H_{cor}^T . The incorporation of this term achieves dual critical advancements by reproducing the essential experimental signatures of the single electron pocket with a reduced Φ_4 [59] and simultaneously enabling a more precise characterization of the band dispersion.

V. CONCLUSION

Our LDA + U calculations reveal that as U increases toward 3.0 eV, the magnetic ground state of FeSe under ambient pressure undergoes a transition from the PAFM order to CAFM order, accompanied by the breaking of Hund's rules. The competition between this striped spin order and orbital polarization further induces a transition to a nonmagnetic purely orbitally polarized state between $U = 4.0$ and 4.5 eV. At $U = 4.5$ eV, this competition explains both the absence of magnetism under ambient pressure and the emergence of CAFM order under moderate pressure, thereby establishing the effective on-site Coulomb repulsion in FeSe to be approximately 4.5 eV.

Our band structure calculations reveal that neither static spin order nor static orbital polarization can describe the experimental band structure, indicating the critical role of quantum fluctuations. By incorporating electronic correlations into a tight-binding model through renormalization of hopping parameters, we successfully reconstruct the nematic

FeSe and reproduce the band structures of FeSe at different temperatures. Comparative analysis between the TBM and DFT-derived ground states reveals that correlation effects drive electron redistribution from d_{z^2} and $d_{x^2-y^2}$ to d_{yz} orbitals, thereby suppressing orbital polarization. This further highlights the essential role of electronic correlations in FeSe.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

APPENDIX

We calculate the intensity of ARPES signal at MT following [87]

$$I(\mathbf{k}, \omega) \propto \sum_n |M_{f,i}^k|^2 \delta(\omega - \tilde{E}_n(\mathbf{k})) f(\tilde{E}_n(\mathbf{k}), T). \quad (\text{A1})$$

Here, $\delta(\omega - \tilde{E}_n(\mathbf{k})) = \Gamma/\pi \cdot [(\omega - \tilde{E}_n(\mathbf{k}))^2 + \Gamma^2]^{-1}$, where $\tilde{E}_n(\mathbf{k})$ is the n th eigenvalue at $\mathbf{k}$ calculated from our fitted tight-binding model and we set broadening factor of $\Gamma = 10$ meV. And $f(\tilde{E}_n(\mathbf{k}), T) = [1 + e^{\tilde{E}_n(\mathbf{k})/(k_B T)}]^{-1}$ is the Fermi distribution function. Within the free electron final state approximation, one-electron matrix element follows

$$M_{f,i}^k \propto S \langle \mathbf{k} | \phi \rangle, \quad (\text{A2})$$

where $|\mathbf{k}\rangle$ is the free electron final state with wave vector $\mathbf{k}$ and $|\phi\rangle$ is the initial state expanded in Wannier basis. Here, $S = 1$ or 0 depending on whether the parity of the incident light matches that of ϕ , which directly illustrates the selection rule. In our tight-binding model,

$$\begin{aligned} \phi &= \sum_{\alpha} (C_{\alpha,k}^{\text{Fe1}} \phi_{\alpha}^{\text{Fe1}} + C_{\alpha,k}^{\text{Fe2}} \phi_{\alpha}^{\text{Fe2}}) \\ &= \sum_{\alpha} (C_{\alpha,k}^{\text{Fe1}} e^{-ik \cdot \mathbf{R}_{\text{Fe1}}} + \mathcal{G}_{\alpha} C_{\alpha,k}^{\text{Fe2}} e^{-ik \cdot \mathbf{R}_{\text{Fe2}}}) \tilde{\phi}_{\alpha}. \end{aligned} \quad (\text{A3})$$

Here, $\phi_{\alpha}^{\text{Fe1}}$ and $\phi_{\alpha}^{\text{Fe2}}$ represent the local 3d orbitals at $\mathbf{R}_{\text{Fe1}}$ and $\mathbf{R}_{\text{Fe2}}$ sites, respectively. And $C_{\alpha,k}^{\text{Fe1}}$ and $C_{\alpha,k}^{\text{Fe2}}$ represent the contribution of $\phi_{\alpha}^{\text{Fe1}}$ and $\phi_{\alpha}^{\text{Fe2}}$, respectively. $\mathcal{G}_{\alpha}$ arises from the glide symmetry when the Wannier functions $\phi_{\alpha}^{\text{Fe1}}$ and $\phi_{\alpha}^{\text{Fe2}}$ are shifted to the unit cell center to form $\tilde{\phi}_{\alpha}$. $\mathcal{G}_{\alpha} = -1$ for d_{xz} and d_{yz} orbitals, and 1 for the other 3d orbitals, respectively. Then, the matrix element can be written as

$$M_{f,i}^k \propto \sum_{\alpha} S_{\alpha} (C_{\alpha,k}^{\text{Fe1}} e^{-ik \cdot \mathbf{R}_{\text{Fe1}}} + \mathcal{G}_{\alpha} C_{\alpha,k}^{\text{Fe2}} e^{-ik \cdot \mathbf{R}_{\text{Fe2}}}) \cdot \langle \mathbf{k} | \tilde{\phi}_{\alpha} \rangle. \quad (\text{A4})$$

The calculation of $\langle \mathbf{k} | \tilde{\phi}_{\alpha} \rangle$ and the effect of light polarization on the ARPES intensity are discussed in Ref. [87].

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