



Coulomb and electron–phonon interactions and doping effects in KNi_2Se_2 crystal

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Abstract.

Using density functional theory combined with dynamical mean-field theory (DFT+DMFT), we explore the correlated electronic structure of the KNi_2Se_2 superconductor and analyze how its paramagnetic normal state spectra is modified by electron–electron interactions and electron/hole doping. We demonstrate the emergence of orbital selectivity, characterized by the coexistence of Mott-localized states, pseudogapped spectral features, and narrow Kondo-like quasiparticle excitations. In addition, we investigate the interplay between on-site Coulomb interactions and electron–phonon coupling in both pristine and doped KNi_2Se_2 . Our results reveal that the combination of these interactions gives rise to low-energy electron bound states. These findings provide microscopic insight into the heavy-electron normal state of KNi_2Se_2 and contribute to a deeper understanding of the coupled roles of dynamical electronic correlations, electron–phonon interactions, and band filling in shaping the normal state properties of unconventional multi-orbital superconductors.

1 Introduction

Since the seminal discovery of unconventional superconductivity in an Fe-containing material [1], substantial progress has been made in understanding the diverse magnetic, nematic, and electronic phases of tetragonal layered materials. A central issue in this field is whether the unconventional superconducting state emerges from a pairing instability of a Fermi-liquid (FL) metal or from a non-Fermi-liquid (nFL) state in proximity to orbital-selective Mottness [2]. One effective way to address this question is to systematically investigate different members of the family of tetragonal transition-metal superconductors. Based on numerous studies, it has become increasingly clear that many of these unconventional superconductors display bad- or strange-metal behavior in their normal paramagnetic state. In such regimes, the electrical resistivity deviates strongly from the FL T^2 behavior as $T \rightarrow 0$, as commonly observed in conventional metals [3]. In correlated multi-orbital (MO) electron systems, the resistivity $\rho(T)$ often follows a power-law behavior, $\rho(T) \propto T^n$, with $1 < n < 2$ in the bad-metal regime and $n = 1$ in the strange-metal regime. This behavior reflects the formation of a strongly renormalized normal electronic state driven by intrinsic MO electronic correlations.

In this context, the discovery of a heavy-electron normal state [4,5], followed by unconventional superconductivity at low temperatures (T) in the tetragonal 122 Ni chalcogenides [4–7] (see Fig. 1), is particularly remarkable. Of special interest here is KNi_2Se_2 , which is non-magnetic, but exhibits charge-density-wave (CDW) instabilities [4,8,9]. It has been argued that, in the stoichiometric composition, the effective valence of the Ni ions in these 122 compounds is close to 1.5+. This mixed-valence $\text{Ni}^{1.5+}$ electronic state [5,8,9] is believed to be responsible for the heavy effective electron masses, $m^* = (6 - 20)m_e$ [5,7], observed in these Ni-based superconducting systems. This interpretation is supported by angle-resolved photoemission spectroscopy studies, which suggest that both TiNi_2Se_2 [10] and KNi_2Se_2 [11] are not strongly correlated materials. In particular, the camelback-shaped band at the Z point has been shown to generate a van Hove singularity near the Fermi energy (E_F) in TiNi_2Se_2 [10], thereby providing a one-band explanation for the heavy-electron behavior in a system governed by relatively weak Coulomb interactions [11]. Within this picture, the role of electron–electron correlation effects in 122 Ni-containing superconductors [12,13] has received comparatively little attention, despite the presence of heavy quasiparticles with considerably enhanced effective masses [7,14,15]. Motivated by this unresolved issue, and following the strategy adopted in Ref. [12], we employ density functional theory combined with

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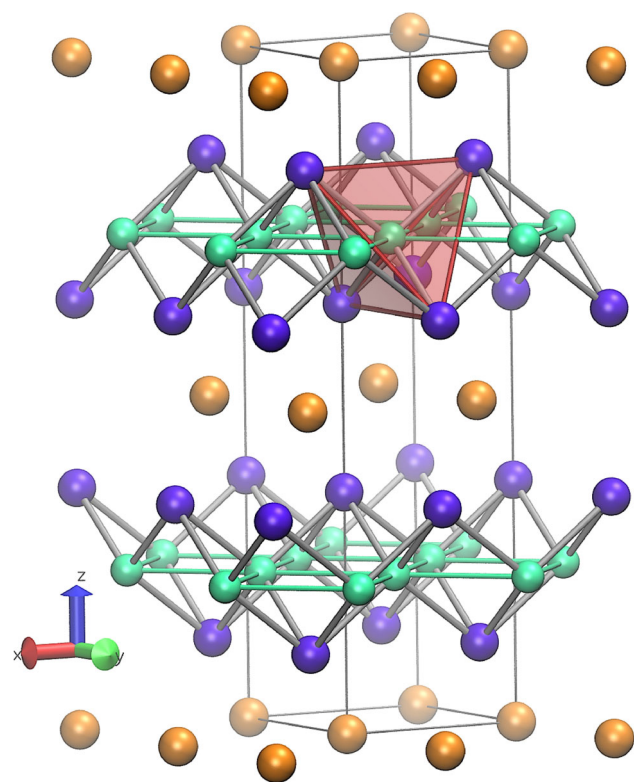


Fig. 1 Illustration of the crystal structure of KNi_2Se_2 and 122 Ni-containing analogs (Ni green, K orange, Se violet). The tetragonal coordination of Ni by Se is represented as a red tetrahedron. The tetragonal unit cell is shown in gray

dynamical mean-field theory (DFT+DMFT) to investigate the role of dynamical MO electron–electron interactions in shaping the correlated electronic structure of the Ni 3*d* shell in pure and doped KNi_2Se_2 . As shown in our earlier studies of MOI Fe-based superconductors, all five Ni 3*d* orbitals must be taken into account to properly resolve the correlated electronic state associated with heavy electron formation in KNi_2Se_2 . In MO systems close to orbital-selective Mottness, changes in many-particle interactions and band filling can generate nontrivial physical effects arising from dynamical electronic correlations, which are captured here within the DFT+DMFT framework. As shown here, MO degrees of freedom can drive the system into a correlated regime in which the chemical potential lies within an energy window of nearly vanishing density of states (DOS), leading to the coexistence of metallic, insulating, and pseudogapped electronic states. To investigate the origin of these unconventional phenomena, we employ the MO DFT+DMFT [16] framework to explore the non-CDW normal state of the KNi_2Se_2 superconductor. In addition, we discuss the emergence of orbital selectivity upon electron and hole doping of the parent compound, as well as its evolution under coupled electron–electron and electron–phonon interactions [17], providing specific predictions that could be tested in future studies of pure and doped bulk KNi_2Se_2 crystals.

2 Results and discussion

The one-electron electronic structure of KNi_2Se_2 was calculated within density-functional theory using the plane-wave pseudopotential implementation of Quantum ESPRESSO, similar to Ref. [12]. The exchange–correlation energy was treated within the PBE generalized-gradient approximation. Pseudopotentials from the SSSP Library (v. 1.1.2, precision branch) were used [18]. The plane-wave and charge-density cutoffs were set to 90 Ry and 750 Ry. The crystal structure data obtained from experiments (see Fig. 1) was used as input [19]. The primitive cell contained five atoms, with two Ni, two Se, and one K atom, with fractional coordinates Ni (0.5, 0.75, 0.25) and (0.5, 0.25, 0.75), Se (0, 0.64612, 0.64612) and (0, 0.35388, 0.35388), and K (0, 0, 0). Brillouin-zone integrations in the self-consistent calculation were performed using an $8 \times 8 \times 8$ Monkhorst–Pack mesh, corresponding to 59 irreducible *k* points. Marzari–Vanderbilt smearing with a width of 0.01 Ry was used. The electronic self-consistency threshold was 1.0×10^{-8} Ry, and 35 Kohn–Sham bands were included. A subsequent non-self-consistent calculation was performed on a uniform $10 \times 10 \times 10$ grid containing 1000 *k* points, using the converged charge density from the self-consistent calculation. These Bloch states were used to construct a low-energy tight-binding representation with Wannier90 [20] (v3.1). The Wannier basis consisted of 10 maximally localized Wannier functions generated from Ni 3*d* trial projections on the two Ni sites. The Wannierization used 35 input Bloch bands, with a disentanglement window from 2.50 to 17.0 eV and a frozen window from 5.7 to 9.9 eV in the Quantum ESPRESSO energy reference. The choice of those energy ranges guarantees Ni localized, atomic-like Wannier functions, with the correct site symmetry and inter-site symmetry-equivalence. The Fermi energy used for Wannier interpolation was 8.6940 eV. Disentanglement was carried out with a mixing ratio of 0.69, and a convergence threshold of 1.0×10^{-14} over a three-iteration convergence window. The final spread minimisation used a 1.0×10^{-12} tolerance over a 10-iteration convergence window. The final total spread of the 10 Wannier functions was 22.6285 Å². The resulting Wannier Hamiltonian was used as the one-electron input for the MO DFT+DMFT treatment of the Ni-3*d* shell. The obtained orbital resolved DFT DOS is displayed in Fig. 2. Consistent with earlier band structure calculations for tetragonal Ni-based superconductors [12, 13, 21] the lineshape of the bare DOS is clearly orbital dependent and strongly particle-hole asymmetric, showing MO metallicity within the Ni-*d*⁸ oxidation of KNi_2Se_2 parent compound.

The one-electron part of the Ni-3*d* model Hamiltonian relevant to the normal paramagnetic state of KNi_2Se_2 is $H_0 = \sum_{\mathbf{k}, a, \sigma} \varepsilon_a(\mathbf{k}) c_{\mathbf{k}, a, \sigma}^\dagger c_{\mathbf{k}, a, \sigma} + \sum_{i, a, \sigma} (\varepsilon_a^0 - \mu) n_{i, a, \sigma}$, where $a = x^2 - y^2, 3z^2 - r^2, xz, yz, xy$ denotes its 3*d* orbitals and $\varepsilon_a(\mathbf{k})$ is the corresponding band dispersion, which encodes details of the DFT band struc-

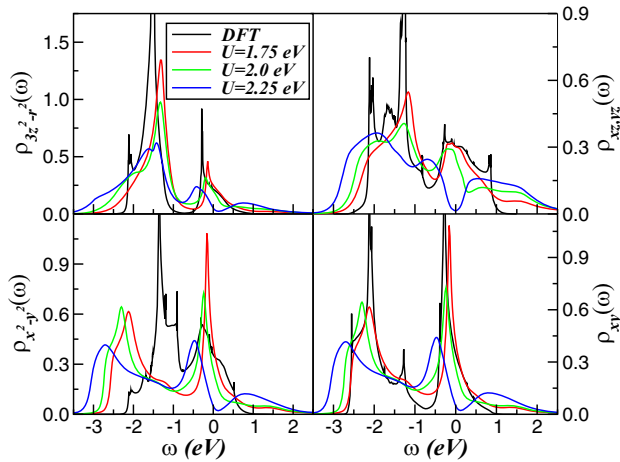


Fig. 2 DFT and DFT+DMFT orbital-resolved density of states (DOS) of the tetragonal KNi_2Se_2 superconductor, illustrating a pronounced spectral weight transfer upon increasing the on-site Coulomb interaction U . While all orbitals remain metallic for U values up to 2.0 eV, a strong electronic reconstruction sets in at $U = 2.25$ eV, leading to the emergence of a pseudogapped electronic state. Also noteworthy are the van Hove singularities appearing at different energies in the DFT DOS, which are progressively suppressed as a consequence of multi-orbital (MO) dynamical correlations

ture. In H_0 above, $\varepsilon_{a,\sigma}^0 \equiv \varepsilon_a^0 - U(n_{a,\bar{\sigma}} - \frac{1}{2}) + \frac{J_H}{2}(n_{a,\sigma} - 1)$, with ε_a^0 being the on-site energies of a given 3d orbital, whose bare values are read off from the DFT spectral functions, and the other terms are subtracted therefrom to avoid double-counting of interactions already treated on average by DFT [22–24], and μ is the chemical potential. These five Ni-3d orbitals are the relevant one-particle inputs for MO-DMFT, which generates a strongly renormalized electronic state as shown below. Moreover, the correlated many-body Hamiltonian considered to KNi_2Se_2 reads $H_{\text{int}} = U \sum_{i,a} n_{i,a,\uparrow} n_{i,a,\downarrow} + U' \sum_{i,a \neq b} n_{i,a} n_{i,b} - J_H \sum_{i,a \neq b} \mathbf{S}_{i,a} \cdot \mathbf{S}_{i,b}$, where U and $U' = U - 2J_H$ are, respectively, the intra- and inter-orbital Coulomb interaction term and J_H is the Hund's coupling, inter-orbital Coulomb interaction and the Hund's coupling. We evaluate the retarded one-particle Green's functions $G_a(\mathbf{k}, \omega) = [\omega - \Sigma_a(\omega) - \varepsilon_a(\mathbf{k})]^{-1}$, with $\Sigma_a(\omega)$ being the Ni-3d self-energy of orbital a of the many-particle problem of KNi_2Se_2 using the MO iterated perturbation theory (MO-IPT) as an impurity solver to DMFT. This interpolative *ansatz* is known to capture the correct low- and high-energy behavior of the one-particle spectral functions and self-energies of MO Hubbard-like models in the infinite-dimensional limit (DMFT). In particular, it reproduces the Mott-Hubbard metal-insulator transition from a correlated metallic state to a Mott-insulating phase as a function of the on-site Coulomb interaction U . The MO-IPT scheme is computationally efficient and provides real-frequency spectra directly at zero temperature, thereby enabling the investigation of electronic structure recon-

struction in real materials with competing phase instabilities. The full set of equations for the MO case can be found, for example, in Ref. [25]; therefore, they are not repeated here.

Since the role of electronic correlations in the excitation spectrum of KNi_2Se_2 and related compounds remains not fully understood, in Fig. 2 we compare the DFT orbital-resolved DOS with the corresponding renormalized spectral functions. This allows us to highlight the evolution of the electronic structure with increasing on-site Coulomb interaction U at fixed $J_H = 0.7$ eV, as used in Ref. [12]: Here, all DFT+DMFT calculations were performed at $T = 0$, using a real-frequency mesh of 0.02 eV, a numerical convergence criteria for the DMFT self-consistency cycle of 0.0075 eV, and a lifetime broadening $i\eta = 0.01$ eV for the retarded Green's functions, the latter being consistent with the values used in Refs. [26]. As shown in Fig. 2, upon inclusion of MO dynamical correlations, the sharp and well-defined van Hove-like singularities present in the DFT DOS are smeared across all Ni-3d orbitals. Many-particle MO self-energy corrections, shown in Fig. 3, induced by an on-site Coulomb interaction of $U = 1.75$ eV lead to the emergence of lower and upper Hubbard bands (LHB and UHB) in the valence- and conduction-band regions, respectively, at different energies for all orbitals of the d^8 electronic configuration of paramagnetic KNi_2Se_2 . While some sharp van Hove features remain visible in the $3z^2 - r^2$, $x^2 - y^2$, and xy spectral functions, the overall spectra are considerably broadened compared with the orbital-resolved DFT DOS. Interestingly, at $U = 2.0$ eV, the $x^2 - y^2$ and xy orbitals reveal FL quasiparticles [27] at low energies near $E_F = \omega = 0.0$ eV. However, at $U = 2.25$ eV, this MO FL metallic state evolves into a pseudogapped electronic state, characterized by a strongly depleted DOS near E_F . Taken together, the results for $U = 2.25$ eV in Fig. 2 reveal the coexistence of pseudogapped states and incoherent Hubbard bands across all Ni-3d orbitals of KNi_2Se_2 . Future polarized X-ray emission and absorption spectroscopy studies would be valuable to test our prediction of the orbital-resolved electronic spectrum of the normal, paramagnetic electronic state of tetragonal KNi_2Se_2 single crystals.

To further characterize the strongly correlated nature of the paramagnetic normal state of KNi_2Se_2 , Fig. 3 displays the U dependence of the imaginary part of the self-energy associated with the active Ni 3d orbitals. Compared with our previous results for the TiNi_2Se_2 superconductor [12], similar qualitative features are observed, although the two systems exhibit distinct fingerprints of Coulomb correlations. In particular, $\text{Im}\Sigma_a(\omega)$ displays a pronounced particle-hole asymmetry, which becomes increasingly enhanced with increasing U . This frequency dependence suggests that the proximity to Mottness in the Ni-3d valence state of KNi_2Se_2 is governed by the dynamical structure of the self-energy, where a sharp pole develops above E_F . In the orbital-selective metallic regime, $\text{Im}\Sigma_a(\omega)$ exhibits the expected quadratic frequency dependence characteristic of a FL metal. By contrast, in the pseu-

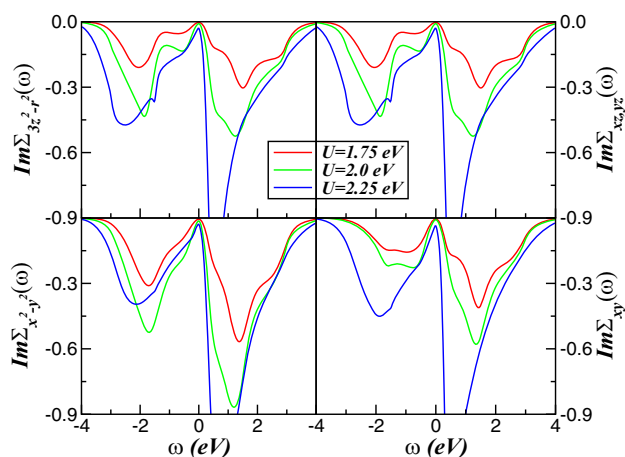


Fig. 3 Orbital-resolved imaginary parts of the self-energy for undoped ($n = 8.0$) KNi_2Se_2 , showing their evolution with increasing U at fixed $J_H = 0.7$ eV. A notable feature is the Fermi liquid (FL) behavior characteristic of a correlated good metal for U values up to 2.0 eV, followed by a clear deviation at $U = 2.25$ eV. This deviation is responsible for the emergence of a pseudogapped electronic state across all Ni-3d orbitals in the nominal Ni-3d⁸ electronic configuration

dogapped phase at $U = 2.25$ eV, $\text{Im}\Sigma_a(\omega)$ remains finite at low energies, rather than following the ω^2 behavior near E_F expected for a conventional FL [27]. As discussed in our earlier studies of tetragonal Fe-based superconductors [28, 29], the energy dependence of $\text{Im}\Sigma_a(\omega)$ shown in Fig. 3 is reminiscent of that found in Mott-Dirac liquids, where massive Dirac-like fermions display deviations from the quadratic FL form at small ω . This behavior is also consistent with the sublinear frequency dependence expected in marginal FLs [30]. A similar sub-linear self-energy behavior was also reported for TiNi_2Se_2 [12], suggesting the presence of hidden marginal FL tendencies near the border of the Mott metal-insulator transition in KNi_2Se_2 . Finally, since the self-energy in DMFT is local, i.e., momentum independent, the quasiparticle residue of orbital a can be obtained directly from the low-energy slope of the real part of the self-energy, i.e., $\frac{m_a^*}{m_e} = \frac{1}{Z_a} = \left(1 - \frac{\partial \text{Re}\Sigma_a(\omega)}{\partial \omega}\right)_{\omega=0}$, where m_e is the free electron mass. Thus, from the slope of the self-energy real part we obtain for $U = 2.25$ eV and $n = 8.0$ ($\frac{m_{3z^2-r^2}^*}{m_e}, \frac{m_{x^2-y^2}^*}{m_e}, \frac{m_{xz,yz}^*}{m_e}, \frac{m_{xy}^*}{m_e}$) = (2.8, 2.7, 20.0, 3.3). Although the $3z^2 - r^2, x^2 - y^2, xy$ have considerably smaller electron mass enhancement as compared with the xz, yz orbitals, the latter are, however, consistent with the heavy effective electron mass $m^* = (24)m_e$ and $m^* = (6 - 18)m_e$ reported, respectively in Refs. [4] and [5].

We shall mention here that LiFeAs is generally regarded as a moderately to weakly correlated FL metal, for which an on-site Coulomb interaction of approximately $U \simeq 2.5$ eV provides a reasonable description of its correlated electronic structure [31]. In contrast, our results for TiNi_2Se_2 in Ref. [12] reveal

a substantially stronger sensitivity to local electronic interactions. In its nominal 3d⁸ (Ni^{2+}) valence configuration, TiNi_2Se_2 undergoes a first-order metal-insulator transition within a remarkably narrow critical interaction range, $U_c \simeq 2.4 - 2.5$ eV. Thus, although comparable values of the bare Coulomb interaction are relevant for the two materials, their many-body responses are qualitatively different: LiFeAs remains on the itinerant FL side, whereas TiNi_2Se_2 is located much closer to a correlation-induced MO Mott insulating instability. This contrast emphasizes that the effective degree of correlation cannot be inferred from the magnitude of U alone, but is controlled by its interplay with the orbital-dependent bandwidths and band fillings, crystal-field splittings, among other degrees of freedom. Thus, our results shown here for U between 2.0 and 2.25 eV in Figs. 2 and 3 contribute to the ongoing discussion [32, 33] concerning the relationship between MO electronic correlations, proximity to pseudogapness and Mottness, and unconventional superconductivity. In particular, they suggest that proximity to a correlation-driven localization threshold may provide an important fundamental principle for understanding the markedly different normal-state electronic properties from which superconductivity emerges in Fe- and Ni-based materials.

To gain further insight into the reconstruction of the normal-state electronic structure of KNi_2Se_2 , Fig. 4 shows the effect of electron and hole doping of the 3d⁸ parent compound. This analysis is motivated by earlier studies [5, 9], which suggested that the Ni ion exhibits a mixed-valence electronic state. It is also motivated by Refs. [19, 34], which reported deintercalation reactions in KNi_2Se_2 and investigated the physical properties of $\text{K}_x\text{Ni}_{2-y}\text{Se}_2$ single crystals, respectively. These effects are all expected to modify the total band filling n of the Ni 3d shell in the KNi_2Se_2 parent compound. Therefore, understanding the role of electron and hole doping is crucial. Despite these earlier studies, the possible emergence of novel electronic states and their instability toward orbital selectivity, as observed in a wide range of correlated electron systems, highlights the relevance of this question. Here, we build on the strengths of MO DFT+DMFT calculations to analyze the effect of carrier (electron/hole) doping in KNi_2Se_2 . In particular, based on self-consistent calculations, we present a set of predictions for the normal state that could be tested in future experiments.

Figure 4 displays the evolution of the correlated electronic structure of KNi_2Se_2 upon electron and hole doping ($n = 8.0 \pm \delta$), calculated for $U = 2.25$ eV and $J_H = 0.7$ eV. An intriguing observation is that orbital selectivity does not occur for weak electron doping, i.e., $\delta = +0.25$, indicating the robustness of the pseudogapped regime against small electron addition. By contrast, when the total 3d filling is reduced, an orbital-selective electronic state develops in hole-doped KNi_2Se_2 . According to our results, the orbital-selective state in the $n = 7.5$ regime is characterized by insulating xz, yz orbitals, a pseudogapped $x^2 - y^2$ orbital, and metallic $3z^2 - r^2$ and xy spectral functions. The origin of

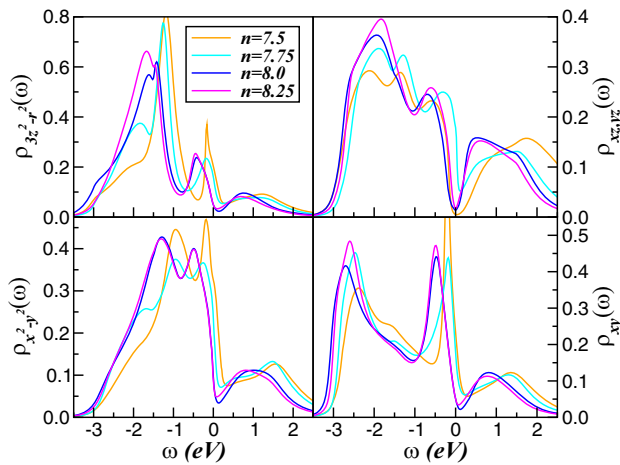


Fig. 4 Effect of electron/hole doping ($n = 8.0 \pm \delta$) on the orbital-resolved DOS of KNi_2Se_2 computed using $U = 2.25$ eV. Particularly interesting features are the robustness of the pseudogap metal for the electron doped case and the emergence of a narrow peak in the $3z^2 - r^2, x^2 - y^2, xy$ for $n = 7.5$, while the xz, yz orbital remain Mott localized in this hole doping regime

these orbital-selective features can be traced to scattering processes between carriers occupying orbital states that are split by the crystal-field environment and shaped by the bare DFT lineshape. This has two important implications. First, the $3d$ bands undergo orbital-dependent shifts relative to one another due to static Hartree contributions arising from the static, Hartree parts of the orbital-dependent self-energies. Second, strong dynamical correlations associated with sizable U , U' , and J_H induce substantial spectral-weight transfer over large energy scales upon electron or hole doping. This latter effect leads to an orbital-selective reconstruction of the spectral functions, as shown in Fig. 4. Thus, in doped KNi_2Se_2 , coexisting Mott-insulating, pseudogapped, and narrow quasiparticle-like components are predicted to emerge at low energies. The hidden orbital selectivity, together with the large-scale dynamical transfer of spectral weight, provides a stringent test of our proposal, and its experimental verification would place this scenario on firmer ground.

We now turn our attention to the combined effects of MO electron–electron and electron–phonon interactions in pure as well as electron- and hole-doped KNi_2Se_2 . To address this issue, we follow the strategy reported in Ref. [17], where the interplay between Coulomb and electron–phonon interactions was investigated in NbS_2 monolayers. Within their formulation, the electron–phonon coupling g , which couples the local charge density to a given phonon mode, is integrated out. This procedure leads to a purely electronic model characterized by an effective dynamical local interaction. In the single-mode approximation, the phonon-mediated contribution to the interaction can be written as $U \rightarrow U_{eff}(\omega) = U - \frac{2g^2\omega_{ep}}{\omega_{ep}^2 - \omega^2}$, with ω_{ep} being the phonon frequency. We shall also mention that within

our MO framework the effective inter-orbital Coulomb interaction now reads $U'_{eff}(\omega) = U_{eff}(\omega) - 2J_H$. However, before looking at the phonon contribution to the reconstructed electronic state of KNi_2Se_2 crystal, we shall mention here that in the local Holstein model the electronic density is coupled to an optical phonon through $H_{ep} = \omega_{ep} \sum_i b_i b_i^\dagger + g \sum_i (b_i + b_i^\dagger) n_{i,\sigma}$ [17, 35], where g is the coupling constant and $b_i^\dagger$ creates a phonon with an oscillator frequency ω_{ep} [35]. Importantly, g is related to the dimensionless coupling λ [37] through the corresponding model dependent relation of the form $\lambda \simeq \frac{2\rho^0(E_F)g^2}{\omega_{ep}}$ [35], with $\rho^0(E_F)$ being the electronic bare DOS at the Fermi level. Importantly, Ref. [36] reported a dimensionless coupling constant $\lambda = 2.1$ which was estimated from heat capacity measurements, indicating that KNi_2Se_2 is a strong-coupling superconductor [35]. Thus, by inverting the dimensionless relation of λ above and using total DFT DOS at E_F for each spin channel of KNi_2Se_2 (not shown) we obtain $g = 0.65$ eV for $\lambda = 2.1$, however, since the precise value of λ or g remains to be confirmed in future studies, here we use $g = 0.49$ eV as a model parameter for the DFT+DMFT calculations. With this mind, and motivated by Ref. [36] reporting the effect of cation-vacancy on the phonon dynamics of pure and doped KNi_2Se_2 , in Fig. 5 we show the reconstructed electronic state obtained using $U = 2.25$ eV, $J_H = 0.7$ eV and the different doping values considered in Fig. 4. A striking feature in Fig. 5 is the large-scale reconstruction of the electronic structure at finite electron–phonon coupling g . As shown, both the low-energy electronic line shapes and the Hubbard bands are strongly renormalized by dynamical spectral-weight transfer induced by the effective interactions U and U' . Equally important is the formation of an insulating state together with the emergence of narrow bound states driven by local dynamical fluctuations, which further reshapes the strongly correlated electronic state. For the doping values considered in Fig. 5, all electron–phonon-induced bound states are substantially modified, suggesting that these states can be tuned by external perturbations, such as chemical substitution. Another notable feature is the loss of spectral weight from the upper Hubbard bands, which is dynamically transferred to electronic bound states located slightly above E_F . Taken together, the interplay between orbital selectivity and strong spectral-weight redistribution represents a robust fingerprint of the correlated electron–phonon-driven electronic state in this tetragonal lattice system, warranting future experimental verification.

As seen in Fig. 5, phonon peaks [39] or the electron–phonon bound states are deeply interconnected also in strongly correlated fermion systems. The combined effect of U and g modifies the band dispersion, splitting conduction- and valence-band edges into distinct branches. As shown in Figs. 5, this reconstruction can confine electronic states near E_F , giving rise to low-energy bound states. Such states are not mere artifacts of electronic structure, but signatures of strong interactions between electron–phonon effects and electron

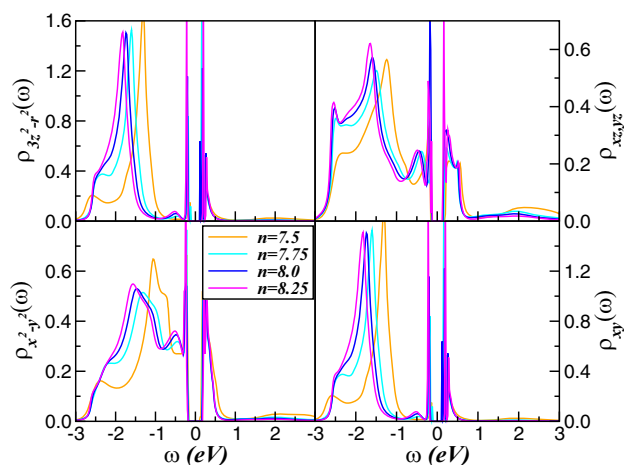


Fig. 5 Effect of combined electron–electron and electron–phonon interactions on the orbital resolved spectral functions of pure and electron/hole doped KNi_2Se_2 . Important features to be seen are the low-energy bound states near E_F and the evolution of upper Hubbard bands at high energies obtained using different total band filling n and a fixed coupling constant $g = 0.49$ eV and $U = 2.3$ eV

correlations. The appearance of very localized, narrow electronic features is expected to provide a microscopic understanding for this emergent electronic behavior in unconventional superconductors. Thus, carrier’s self-trapping [38] which might be induced in a system with sizable electron–phonon interactions, can be taken as a manifestation of the strong coupling between orbital and lattice degrees of freedom in three dimensional materials. As shown in Figs. 5, the electron–phonon interaction can increase the localization tendency by splitting orbital or band states and generating bound states near E_F , thereby reducing the mobility of itinerant carriers. Here, local electronic fluctuations act as a local scattering channel that disrupts coherent motion, reinforcing the confinement of charge carriers into self-trapped states.

3 Conclusion

In summary, we employed a five-orbital Hubbard model within the DFT+DMFT framework to investigate the correlation-induced electronic reconstruction of the normal paramagnetic state of tetragonal KNi_2Se_2 . Our results show that multi-orbital dynamical correlations drive a crossover from a metallic Fermi-liquid state to a pseudogapped electronic state. Upon electron or hole doping the parent compound, the pseudogapped phase undergoes an orbital-selective reconstruction, characterized by the coexistence of insulating, pseudogapped, and metallic orbitals with narrow quasiparticle features. This orbital-selective behavior is particularly relevant to experiments on heavy-electron states with large effective masses, which may be linked to multiband superconductivity at low temperatures [7]. We fur-

ther demonstrate that electron–phonon and electron–electron correlations cooperate to generate bound states and enhance orbital selectivity. A key outcome of this study is the identification of a transition from a metallic state to an insulating regime, accompanied by sharp low-energy phonon-induced features emerging below and above the Fermi level and by a substantial reshaping of the spectral functions [39]. These findings provide a microscopic description of the orbital-selective reconstruction in the normal state of KNi_2Se_2 , positioning this compound as a model system for exploring the interplay between multi-orbital electron–electron interactions, electron–phonon coupling, and doping effects in transition-metal systems.

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Author contributions

S.L. has performed the DFT calculation. L.C. designed and carried out the DFT+DMFT study. The authors have approved the final version of the manuscript.

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