Normal-state correlated electronic structure of the tetragonal TiNi$_2$Se$_2$ superconductor

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On the basis of density functional plus dynamical mean-field theory calculations we explore the electronic structure of the TiNi$_2$Se$_2$ superconductor, showing how the normal-state spectra are reshaped by dynamical many-particle interactions. Upon electron-doping the 3$d^9$ Fermi liquid metal, hidden orbital selectivity with coexisting Mott localized, pseudogapped, and narrow Kondo-quasiparticle electronic excitations is predicted for TiNi$_2$Se$_2$. By specifically focusing on the energy spectrum that is relevant to the mixed-valence heavy-electron normal state of unconventional superconductors, our findings have the potential to enhance our microscopic understanding of the intricate interplay between many-particle physics and electron band filling, paving the way for further advancements.

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I. INTRODUCTION

Since its discovery in 2008 [1], multiorbital (MO) unconventional superconductivity in tetragonal layered materials has become a prominent area of research in condensed matter and materials science. Significant progress has been made in understanding the origins and interdependence of various phases in this category of correlated electron systems. The ongoing discussions surrounding the transition from the normal state to unconventional superconductivity have had a significant impact on the debate about correlated electronic structure reconstruction and potential phase instabilities. A key point of contention has been determining whether superconductivity arises from a pairing instability of a Fermi liquid (FL) metal or as a non-Fermi liquid near an orbital-selective Mott metal-insulator transition [2].

One effective approach to answering this fundamental question is to conduct a systematic investigation of different members of the family of MO tetragonal transition-metal superconductors. It has accordingly become evident that these materials exhibit bad-metal behavior in their normal, paramagnetic state, where the electrical resistivity deviates considerably from the canonical FL $T^2$ behavior as $T \to 0$ [3]. In strongly correlated electron systems, $\rho(T)$ typically follows a power-law behavior with $1 < n < 2$ in the bad-metal state, reflecting the development of a highly renormalized lattice coherence scale resulting from the growing relevance of dynamic MO electronic correlations at temperature decreases.

In this context, the discovery of multiband [4] and multigap nodeless [5] superconductivity, along with $\rho(T) = \rho_0 + AT^2$ behavior observed from 25 K down to temperatures near $T_c = 3.7$ K in the 122-type Ni chalcogenide TiNi$_2$Se$_2$ (see Fig. 1), is noteworthy. This finding adds another member to the tetragonal superconducting family, where superconductivity emerges in the parent compound under ambient pressure. Similar to the LiFeAs superconductor and in contrast to most Fe-based superconductors [6], TiNi$_2$Se$_2$ has no magnetic or structural phase instabilities. Compared with Fe-based superconductors, Ni-based systems have lower superconducting critical temperatures, which are usually below 5.0 K even upon doping [7]. To date, there is no evidence for ordered or even fluctuating magnetism associated with Ni in the normal state. Although under debate [8–10], for KNi$_2$X$_3$ (X = Se, S) [11,12] and TiNi$_2$Se$_2$ [4], the weakly correlated electronic state that gives rise to pseudogap formation in optics [13] and unconventional superconductivity [10] seems to involve heavy-electron [4,11] behavior below a FL coherent temperature that is driven by mixed valence.

In contrast to KFe$_2$Se$_2$ [14], the Ni-based 122-type compounds are homogeneous without Ni vacancy or phase separation [4,11]. It has been argued that the effective valence of 122-type Ni compounds is 1.5+. The mixed valency of Ni$^{1.5+}$ is believed to be responsible for the heavy-electron effective mass $m^* = (6-20)m_e$ [4,11] inferred from extant electronic specific heat studies. This view is supported by angle-resolved photoemission spectroscopy studies, suggesting that neither TiNi$_2$Se$_2$ [9] nor KNi$_2$Se$_2$ [8] is a strongly correlated material and that the camelback-shaped band at the $Z$ point gives rise to a Van Hove singularity near the Fermi energy $E_F$ in TiNi$_2$Se$_2$ [9], providing a one-band explanation for the heavy-electron behavior in a system governed by weak Coulomb correlations [8]. However, the consequences of orbital selectivity and proximity to electronic localization in tetragonal 122-type Ni superconductors have not received enough attention, in spite of hidden heavy electrons with considerably large effective masses [4,11,12,15].

LiFeAs is considered a weakly correlated Fermi liquid metal with an on-site Coulomb interaction $U$ of around 2.5 eV [16]. In contrast, our findings indicate that the 3$d^8$ (Ni$^{2+}$) valent state of TiNi$_2$Se$_2$ is considerably more correlated than that of LiFeAs, displaying a first-order metal-insulator transition at a critical $U$ value ranging between 2.4 and 2.5 eV. This insight helps to further the ongoing discussion [17]...
about the connection between the level of MO electronic correlations and the proximity to Mottness unconventional superconductors.

In this paper, we investigate the role of dynamical MO electronic correlations in the electronic structure reconstruction in the Ni-3d shell of both self-hole-doped (Ni$^{2+}$, see our discussion below) and mixed-valence (Ni$^{1.5+}$) TINi$_2$Se$_2$. As shown in our earlier studies on MO Fe-based superconductors [18–20], all five 3d bands need to be considered in order to satisfactorily resolve the correlated electronic state composed of heavy particles in stoichiometric ($d^{8.5}$) TINi$_2$Se$_2$. In MO systems near selective Mott localization, changes in many-particle interactions and electron band filling can lead to interesting physical effects driven by electronic correlations. These tunable degrees of freedom can create an exotic regime where the chemical potential is found in an energy region with almost zero density of states (DOS), resulting in a coexistence of metallic, insulating, and pseudogapped electronic states. To study these phenomena, we employ the density functional plus dynamical mean-field theory (DFT+DMFT) framework and examine the correlated normal state of the TINi$_2$Se$_2$ superconductor. Additionally, we discuss the influence of FL metallicity and selective Mottness and provide specific predictions that can be tested in future studies.

II. RESULTS AND DISCUSSION

MO physics is inherently complex due to orbital and spin degrees of freedom. These coupled correlations have been hampering fully realistic theoretical studies. In recent years, however, the DFT+DMFT approximation [21] has established itself as a reference computational framework that allows for a systematic approach to the treatment of MO electronic correlations in real crystals. Here, we used DFT to calculate Bloch electronic states in the Perdew-Burke-Ernzerhof (PBE) approximation using Vanderbilt ultrasoft standard solid state pseudopotentials (SSSP) [22]. The plane-wave expansion of valence electron wave functions and charge density used kinetic energy cutoffs of 75 and 650 Ry, respectively. Calculations were performed using the PW.x code (v6.7) of the QUANTUM ESPRESSO (QE) package. The crystal structure data obtained from experiments (see Fig. 1) were used as input [23]. Self-consistent-field (SCF) calculations were run on a k grid of $8 \times 8 \times 8$ data points. A tight-binding band structure model was obtained from interpolating maximally localized Wannier functions (MLWFs), calculated with the WANNIER90 package (v3.1) [24]. As the initial guess for the Wannier functions, 3d orbitals were used for Ni (ten MLWFs in total). Iterative spread minimization provided real-valued, maximally localized Wannier functions with the correct site symmetry and intersite symmetry equivalence (Fig. 2). The space group symmetry compliance of the Wannier Hamiltonian after spread minimization was checked with WANNIERTOOLS [25]. The quality of the Wannier fit is shown in Fig. 3. The resulting orbital-resolved DFT DOS is displayed in Fig. 4. Consistent with earlier band structure calculations for tetragonal Ni-based superconductors [26], the line shape of the bare DOS is strongly orbital dependent, showing MO all-electron metallicity within the Ni-d$^8$ oxidation state in contrast to the KFe$_2$Se$_2$ superconductor [19], due to there being more 3d electrons in TINi$_2$Se$_2$ than in KFe$_2$Se$_2$.

Similar to Fe-based superconductors [18,19] as well as to infinite-layer Ni superconductors [27], the one-electron part of the Ni-3d model Hamiltonian [28–30] relevant to the normal paramagnetic state of TINi$_2$Se$_2$ is

$$H_0 = \sum_{\mathbf{k},\mathbf{a},\sigma} \epsilon_{\mathbf{a}}(\mathbf{k}) c_{\mathbf{a},\sigma}^\dagger c_{\mathbf{a},\sigma} + \sum_{i,a,\sigma} \left( \epsilon_{i,a,\sigma} - \mu \right) n_{i,a,\sigma},$$

where $a = x, y, z$, $3\mathbf{r}^2 - \mathbf{r}_z^2$, $\mathbf{x}_z$, $\mathbf{y}_z$, $\mathbf{x}_y\mathbf{y}$ denotes the 3d orbitals of TINi$_2$Se$_2$. Here, $\epsilon_{\mathbf{a}}(\mathbf{k})$ is the corresponding band dispersion, which encodes details of the one-electron (DFT) band structure, and $\epsilon_{i,a,\sigma}^{(0)} \equiv \epsilon_{i,a,\sigma} - U (n_{i,a,\uparrow} - \frac{1}{2})^+ + \frac{1}{2} (n_{i,a,\uparrow} - 1)$. $\epsilon_{i,a}$ is the on-site energy of a given 3d orbital, and the other terms are subtracted therefrom to avoid double-counting of interactions already treated on average by DFT [31]; $J_H$ is the Hund’s coupling. Finally, $\mu$ is the chemical potential of the system. These five Ni-3d orbitals are the relevant one-particle inputs for MO-DMFT, which generates a renormalized electronic state as shown below. Moreover, consistent with our earlier studies, the correlated many-body Hamiltonian considered for TINi$_2$Se$_2$ reads $H_{\text{int}} = U \sum_{i,a,\uparrow} n_{i,a,\uparrow} n_{i,a,\downarrow} + U' \sum_{i,a\neq b} n_{i,a} n_{b} - J_H \sum_{i,a\neq b} S_i.a \cdot S_{i,b}$. Here, $U' = U - 2J_H$ is the interorbital Coulomb interaction term. Given the complexity of the MO problem, with diagonal and off-diagonal lattice Green’s functions and self-energies [32], here we work in the basis that diagonalizes the one-particle density matrix. In this basis, interorbital one-electron overlap is zero, and so in the paramagnetic phase we have $G_{a,b,\sigma}(\omega) = \delta_{a,b} G_{a,\sigma}(\omega)$ [31]. In this regime, electrons among different orbitals interact only via the interorbital Coulomb interaction and the
Hund’s coupling. We evaluate the retarded Green’s functions $G_a(k, \omega) = \frac{1}{\omega - \Sigma_a(\omega) - E_k}$, with $\Sigma_a(\omega)$ being the Ni-3$d$ self-energy of orbital $a$ of the many-particle problem of TlNi$_2$Se$_2$ using the multiorbital iterated perturbation theory (MO-IPT) as impurity solver within DMFT. This interpolative ansatz is known to account for the correct low- and high-energy behavior of the one-particle spectral function and self-energies of MO Hubbard-like models in the infinite-dimension limit (DMFT). It ensures the Mott-Hubbard metal-insulator transition from a correlated metal to a Mott insulator as a function of the Coulomb interaction $U$. The MO-IPT scheme is computationally very efficient, with real-frequency output at zero temperature, enabling the study of electronic structure reconstruction of real materials with different phase instabilities. The full set of equations for the MO case can be found, for example, in Ref. [33]; therefore we do not repeat the equations here. This real-frequency perturbative scheme has a proven record of good semiquantitative agreement with experiment for a range of correlated materials, and it gives results in qualitative accord with numerically exact continuous-time quantum Monte Carlo (CT-QMC) calculations for the tetragonal FeSe superconductor [34], in spite of the fact that fully charged self-consistent DFT+DMFT calculations are presently unreachable within our scheme.

FIG. 2. Wannier functions for the two symmetry-equivalent Ni sites in TlNi$_2$Se$_2$, in the Wannier projections (a) $3z^2-r^2$, (b) $xz$, (c) $yz$, (d) $x^2-y^2$, and (e) $xy$. Isosurfaces are interpolated at $\pm 3.53$ (red and blue lobes). Ni, gray; Se, yellow. The distorted tetrahedral Se coordination around Ni is shown. Wannier functions display ($D_{2d}$) close-to-atomic character.

FIG. 3. Band structure of TlNi$_2$Se$_2$, calculated along $\Gamma$-X-$S$-$Y$-$T$-$R$-$U$-$Z$-$\Gamma$. The Wannier band structure model (orange dashed lines) is displayed for the ten 3$d$ bands (black dashed lines) around the Fermi level. Further occupied (gray dashed lines) and empty bands (green) are shown.

FIG. 4. DFT and DFT+DMFT orbital-resolved density of states (DOS) of the tetragonal TlNi$_2$Se$_2$ superconductor, showing spectral weight redistribution over large energy scales with increasing on-site Coulomb interaction $U$ within the Ni-3$d^8$ electronic configuration. While all orbitals are metallic at $U = 2.0$ eV, Mott localization is obtained for $U = 3.0$ eV. Here, all DFT+DMFT(MO-IPT) spectral functions are computed at zero temperature.
The IPT approach is an interpolative ansatz that connects the two exactly soluble limits of the one-band Hubbard model [35], namely, the uncorrelated \((U=0)\) and the atomic \(\epsilon(k)=0\) limits. It accounts for the correct low- and high-energy behavior of the one-particle spectra, and the metallic FL behavior in the large-\(D\) limit (DMFT) [36]. It ensures the Mott-Hubbard metal-insulator transition from a correlated FL metal to a Mott-Hubbard insulator as a function of the Coulomb interaction \(U\). As shown below, the DFT+DMFT(MO-IPT) solution for \(\text{TlNi}_2\text{Se}_2\) introduces nontrivial effects stemming from the dynamical nature of strong electronic correlations. These processes lead to transfer of spectral weight across large energy scales in response to changes in the on-site Coulomb repulsion, a characteristic lying at the heart of the anomalous responses of correlated electron systems. We recall that a similar perturbative scheme to that used here for the MO Hamiltonian of \(\text{TlNi}_2\text{Se}_2\) has been proposed in Ref. [37], where electron correlation effects in local-orbital electronic structure calculations were applied to Si bulk crystals and H\(_2\)O molecules. It is also worth noting that direct comparisons between MO-IPT results with numerically exact methods, such as the CT-QMC, have been performed in recent years [38], showing good qualitative agreement between the two impurity solvers. Hence, in view of this and our previous studies on correlated electron systems showing good theory-experiment agreement [31,34], we are confident in using MO-IPT to explore the normal-state electronic structure reconstruction of the \(\text{TlNi}_2\text{Se}_2\) superconductor.

Since the effect of electronic correlations in the excitation spectrum of \(\text{TlNi}_2\text{Se}_2\) and analogs is not fully understood yet, in Fig. 4 we compare the DFT orbital-resolved DOS with the corresponding renormalized spectral functions to highlight the electronic structure evolution which emerges with increasing the on-site Coulomb interaction \(J_H\) and fixed \(J_H=0.7\) eV [29]. To obtain the total orbital occupations, the DFT+DMFT equations are constrained by the sum rule \(n_{\text{total}}=\int_{-\infty}^{\infty} \rho_{\text{total}}(\omega)\text{d}\omega\equiv n\). The chemical potential \(\mu\) is then determined self-consistently within the DFT+DMFT loop at each stage using \(\rho_{\text{total}}(\omega) = \sum_{\alpha} \rho_\alpha(\omega)\) computed in each iteration. As seen in Fig. 4, upon consideration of MO dynamical correlations, the sharp and well-defined singular features of the bare electronic structure are smeared on all Ni-3d orbitals. Many-particle MO self-energy corrections induced using an on-site Coulomb interaction \(U=2.0\) eV lead to the emergence of lower and upper Hubbard bands (LHB and UHB, respectively) in the valence and conduction bands at energies close to ±1.0 eV on all orbitals for the 3d\(^{8}\) electronic configuration of self-hole-doped \(\text{TlNi}_2\text{Se}_2\). While some sharp features remain visible in the \(xz, yz\), and \(xy\) spectral functions at high binding energies, the overall spectra are considerably broadened compared with the bare spectral functions. Interestingly, at \(U=2.0\) eV all orbitals reveal emergent FL quasiparticles [36] at low energies. However, at \(U=3.0\) eV all electron localization is induced in tetragonal \(\text{TlNi}_2\text{Se}_2\), which is characterized by a pronounced Mott-Hubbard gap near \(E_F = \omega = 0\). A similar insulating state could possibly be seen in future studies on strained \(\text{TlNi}_2\text{Se}_2\) crystals, due to an enhanced \(U/W\) ratio via strain-induced one-particle band narrowing [40]. Taken together, our results in Fig. 4 reveal the coexistence of Kondo quasiparticles and incoherent electronic excitations on all Ni-3d orbitals of 3d\(^{8}\)\(\text{TlNi}_2\text{Se}_2\). Future polarized x-ray emission and absorption spectroscopy studies are called for to confirm our prediction for the orbital-resolved electronic spectrum of the unstrained \(\text{TlNi}_2\text{Se}_2\) superconductor.

We shall notice here that while incoherent, bad metals are characterized by pseudogapped electronic excitations near \(E_F\), coherent FL metals show, on the other hand, narrow Kondo-quasiparticle resonances. The Kondo quasiparticle, also referred to as the Abrikosov-Suhl resonance [36], is a key feature of the FL metal, in which the spin-flip scattering process leads to the formation of a many-body bound state known as the Kondo resonance. The Kondo resonance is a quasiparticle excitation that appears as a sharp peak in the one-particle spectral function at low temperatures. The fingerprint of the Kondo quasiparticle can be observed in the low-energy spectral function, which in systems away from half filling displays a characteristic asymmetry in the peak shape. The observation of the Kondo quasiparticle in the one-particle spectral function provides a direct probe of Kondo-like (multiorbital or not) physics, and it has been used to study a wide range of coherent FL metals and heavy-fermion materials, among other quantum many-particle systems.

For a more detailed analysis of dynamical MO correlation effects, in Fig. 5 we display the abrupt changes in the spectral functions across the first-order metal-to-insulator transition, which could possibly be seen in future studies on strained \(\text{TlNi}_2\text{Se}_2\). As seen in this figure, MO electronic correlations

![Graph](image_url)
lead to abrupt modifications of the correlated spectra. Below the critical value of the Mott localized phase ($U_c = 2.5$ eV) the many-body spectra describe a coherent, FL metal with orbital-dependent low-energy Kondo-quasiparticle features. Noteworthy, however, is the suppression of the quasiparticle coherence at low energies at $U_c$ and the emergence of a well-defined $V$-shaped gap near $E_F$. Similar Dirac-like electronic dispersion has been reported for BaFe$_2$As$_2$ [41], suggesting that 3$d^8$ TiNi$_2$Se$_2$ may share a universal band structure with TlNi$_2$Se$_2$, in spite of a chemical potential shift due to the steric hindrance of Tl$_2$O$_2$ in TlNi$_2$Se$_2$. Comparing our results with results for ThFeAsS [4], it is worth noting that we only observe effective electron mass enhancements consistent with previous observations in the 3$d^8$ Mott-Dirac regime. This suggests that there is a complex interplay between MO electronic correlations and electron band filling as the 122-type Ni-based superconductors approach the mixed-valence state [11]. The results presented below illustrate this relationship in more detail.

To rationalize the overall correlated electronic behavior of TiNi$_2$Se$_2$, in Fig. 7 we compare our orbital-resolved DFT+DMFT result for $U = 2.4$ eV and $n = 8.0$, ($\frac{m^*}{m_e} = \frac{m^*}{m_e} = \frac{m^*}{m_e} = \frac{m^*}{m_e} = 2.6, 4.0, 3.3, 2.7$), with the experimental curves at $E_F$, which is considerably smaller as compared with the heavy-electron effective mass $m^* = (14$–$20)m_e$ inferred from electronic specific measurements [4]. It is worth noting that the quasiparticle residue $Z_a$ of an orbital $a$, which defines the renormalized Fermi energy, directly yields the effective electron mass enhancement: $m^* = \frac{1}{Z_a} = (1 - \frac{\partial \Re \Sigma_a(\omega)}{\partial \omega})_{\omega=0}$, where $m_e$ is the free electron mass. Thus, from the slope of the quasiparticle real energy part we obtain, for $U = 2.4$ eV and $n = 8.0$, ($\frac{m^*}{m_e} = \frac{m^*}{m_e} = \frac{m^*}{m_e} = \frac{m^*}{m_e} = 2.6, 4.0, 3.3, 2.7$), which is somehow reminiscent of marginal, Mott-Dirac liquids where the valence one-electron band structure of TlNi$_2$Se$_2$ and its link to Kondo-quasiparticle and incoherent components with distinct orbital and k-resolved electronic line shape should be visible at low to moderate binding energies, in spite of large-scale spectral weight transfer induced by sizable electron-electron interactions. Interesting in this context is the crossing point seen at the experimental curves at $-0.41$ eV, a fingerprint of dynamical transfer of spectral weight. Given the intrinsic 3$d^8$ nominal state of TiNi$_2$Se$_2$, future studies are called for to corroborate our prediction of hidden self-hole-doping behavior possibly due to charge disproportionation of Tl$^{5+}$ to Tl$^{4+}$ and Tl$^{3+}$ [11] in the TiNi$_2$Se$_2$ superconductor or to valence...
the distribution curves (EDCs) taken from Ref. [9]. The theory curve for the peak position of the incoherent one-particle excitation close to theory-experiment agreement at high binding energies, particularly coincide with experiment at low energies. Notice the good qualitative of the Kondo-quasiparticle resonance in the preserve the total number of 3 electrons (i.e., eight electrons for Ni).

To gain further insight into the normal-state electronic structure reconstruction of TlNi2Se2, in Fig. 8 we show the effect of increasing the total electron band filling n of the 3d shell. This is motivated by the fact that similar to KNi2Se2 [8] in an ionic picture the Ni orbital configuration is 3d8.5 for TlNi2Se2. Interestingly, this Ni-3d8.5 electronic configuration has been proposed [11] to induce the effective band mass enhancement seen in experiment. Therefore understanding the effect of electron band filling is crucial. Despite existing spectroscopy and transport data, the generic appearance of novel states and the instabilities of such states to pseudogap formation [13], and, in particular, to unconventional superconductivity [10], in a wide variety of other correlated electron systems highlight the importance of this question. Our aim here is to build upon the strengths of MO correlated electronic structure modeling to analyze the effect of electron band filling in TlNi2Se2. In particular, on the basis of explicit calculations, we will present a set of predictions which could be tested in future experiments in the normal state.

fluctuating instability similar to that reported for infinite-layer nickelates [45]. Additionally, as discussed for perovskite nickelates [46], the self-hole-doped regime we have found here for TlNi2Se2 might also come from a hidden charge self-regulation mechanism [47] in which the electronic density around transition-metal cations reorganizes itself in order to preserve the total number of 3 electrons (i.e., eight electrons for Ni).

In Fig. 8 we show the changes in the correlated electronic structure (for U = 2.4 eV and JH = 0.7 eV) upon increasing the total electron concentration (n = 8.0 + δ, with δ > 0) of self-hole-doped TlNi2Se2. An intriguing observation is that orbital selectivity does not occur at small δ (i.e., δ ≲ 0.2), showing the robustness of the FL regime to weak electron addition. However, as δ increases to about 0.3, an orbital-selective state develops in stoichiometric TlNi2Se2. According to our results, orbital selectivity in the n = 8.5 mixed-valence state is characterized by the presence of insulating (x2 − y2), pseudogapped (3z2 − r2), and metallic (xz, yz, xy) spectral functions. The narrow Kondo quasiparticles, responsible for the emergent heavy electrons observed in experiments (see our results in Fig. 8), are particularly intriguing. The origin of these orbital-selective features can be attributed to scattering between different carriers in orbital states split due to specific crystal fields and bare DFT line shape. The latter has two implications: orbital-dependent shifts of the 3d bands relative to each other via static Hartree contributions (from the static part of the orbital-dependent self-energies), and strong dynamical correlations due to sizable U, U′, and JH, which cause appreciable spectral weight transfer over large energy scales upon carrier doping. This second feature leads to an orbital-selective modification of the spectral functions, as shown in Fig. 8. Thus, for mixed-valence TlNi2Se2, coexisting Mott insulating, pseudogapped,
and narrow Kondo components are predicted to emerge at low energies. The hidden orbital selectivity and large-scale dynamical transfer of spectral weight are stringent tests of our proposal for the mixed-valence state, and experimental verification should place it on solid ground.

Lastly, motivated by the emergence of the superconducting state observed in 122-type Ni superconductors from a mixed-valence-driven heavy-electron behavior [11], in Fig. 9 we display the renormalized electron mass as function of total band filling n, computed using the DFT+DMFT orbital-resolved spectral functions of ThNi2Se2 for U = 2.4 eV and JH = 0.7 eV. Our results suggest strong orbital differentiation between the εg (xy, xz, yz) and eg (x2 − y2, 3z2 − r2) orbitals and enhanced mass renormalization within the εg electronic channel with increasing n. This response is characteristic of strongly correlated materials, where the orbital differentiation is linked to charge and orbital excitations [48] in the reconstructed electronic state at low temperatures.

III. CONCLUSION

We used a five-orbital Hubbard model treated within the DFT+DMFT framework to investigate the correlation-induced electronic reconstruction in tetragonal TiNi2Se2. Our results show that the metallic Fermi liquid nature of the compound arises from multiorbital dynamical correlations. Upon electron-doping the self-hole-doped Ni2+ valence state, we observe a first-order phase transition from a Fermi liquid metal to an orbital-selective phase with insulting, pseudogapped, and metallic orbitals with narrow Kondo quasiparticles. This transition occurs upon approaching the heavy-electron mixed-valence regime and is accompanied by significant changes in dynamical spectral weight transfer. Our finding of orbital-selective Kondo quasiparticle behavior is relevant to experiments involving heavy electrons with large effective masses which may give rise to multiband superconductivity [4] at low temperatures.

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