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Coulomb correlations and the electronic structure of bulk V_2Te_2O

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The effect of Coulomb correlations on the electronic structure of bulk van der Waals material V_2Te_2O is studied by the charge self-consistent density functional theory and dynamical mean-field theory method. Our results show a significant correlation-induced renormalization of the spectral functions in the vicinity of the Fermi energy which is not accompanied by a transfer of the spectral weight to Hubbard bands. The computed quasiparticle effective mass enhancement m^*/m for the V $3d$ states varies from 1.31 to 3.32 indicating an orbital-dependent nature of correlation effects and suggests an orbital-selective formation of local moments in the V $3d$ shell. We demonstrate that taking into account of Coulomb interaction between the V $3d$ electrons yields the electronic specific heat coefficient $\gamma = 26.94\,\mathrm{mJ\,K^{-2}\,mol^{-1}}$ in reasonable agreement with the experiment. We show that the strength of Coulomb correlations is sufficient to trigger a band shift along the $Z - \Gamma - X$ path of the Brillouin zone leading to a collapse of the electronic Fermi surface pocket centered on the $\Gamma - Z$ direction predicted by density functional theory.

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Low-dimensional materials exhibiting a planar structural motif have always attracted a considerable amount of attention due to their exceptional physical properties [1]. The discovery of graphene and pnictide (chalcogenide) iron-based unconventional superconductors have stimulated an intense search of new (quasi) two-dimensional systems [2, 3]. Transition metal dichalcogenides are historically one of the most studied compounds of the class. From the 1960s these materials have been subject of ongoing research on superconductivity, spin- and charge-density waves [4–8].

Recently, a new family of two-dimensional van der Waals materials (TDWM) V_2Ch_2O (Ch = Se, Te) has been synthesized from their Cs and Rb precursors [9, 10]. In contrast to known metallic TDWM the electronic properties of V_2Se_2O and V_2Te_2O as well as the potential precursor of S-based TDWM CsV_2S_2O bear the fingerprints of strong electronic correlations [9–13]. In particular, the electronic specific heat coefficient of metallic V_2Te_2O is $33.9\,\mathrm{mJ\,K^{-2}\,mol^{-1}}$, almost four times of that obtained by density functional theory (DFT) calculations indicative of strong renormalization of the electron mass [9]. Concurrently, the isostructural isoelectronic system V_2Se_2O is a paramagnetic (PM) semiconductor and has large local magnetic moments. In addition, the resistivity of V_2Se_2O exhibits an anomalous $\log(1/T)$ behavior reminiscent of that in underdoped cuprates [10].

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In this Letter, we employ a combination of DFT with dynamical mean-field theory (DMFT) referred to DFT + DMFT to study the importance of Coulomb correlation effects in bulk PM V_2Te_2O [14, 15]. To this end, we compute and compare the DFT and DFT + DMFT Fermi surface, momentum-integrated and momentum-resolved spectral functions of V_2Te_2O. To quantify the strength of correlation effects we analyze the enhancement of quasiparticle mass and the local spin susceptibility of the V $3d$ states. In addition, we compute the electronic specific heat coefficient by DFT and DFT + DMFT and compare its value to the experiment.

Our results for the spectral properties show that the total spectral function in the vicinity of the Fermi energy (E_F) is dominated by the V $3d$ states. The shape of the V $3d$ orbitally-resolved spectral functions $A_\alpha(\omega)$ ($\alpha = 3z^2 - r^2, xz, yz, xy, x^2 - y^2$) computed by DFT + DMFT is similar to that obtained by DFT except a narrow region close to E_F. Specifically, we observe a strong renormalization of $A_\alpha(\omega)$ for the yz and $3z^2-r^2$ orbitals in the vicinity of E_F while the shape of those of the xz, xy, and x^2-y^2 states is less sensitive to correlation effects implying their orbital-selective nature. The overall transformation of the V $3d$ spectral function resembles that of the Fe $3d$ states in parent compounds of iron-based superconductors (FeSC) structurally related to oxychalcogenide TDWM [16, 17].

Next, we compute the orbitally-resolved quasiparticle mass enhancement m^*/m. Our results show that the largest m^*/m of 3.32 and 2.72 occurs in the $x^2 - y^2$ and xz states, respectively. The other V $3d$ states exhibit significantly smaller m^*/m ranging from ~ 1.3 ($3z^2 - r^2$ and yz) to 1.94 (xy). These values are by a factor of about 1.5 larger than those obtained by DFT + DMFT for moderately correlated parent compounds of FeSC [17–19].

The computed mass enhancement suggests a strongly correlated metallic ground state of V_2Te_2O in line with a large value of the Sommerfeld specific heat coefficient $\gamma_{\rm exp} = 33.9\,{\rm mJ\,K^{-2}\,mol^{-1}}$ obtained in the experiment (as compared to that derived from DFT) [9]. Our DFT calculations yield the electronic specific heat coefficient $\gamma_{\rm DFT} = 9.67\,{\rm mJ\,K^{-2}\,mol^{-1}}$, which is 3.5 times smaller the experimental estimate, in reasonable agreement with the result reported by Ablimit and coauthors [9]. By contrast, our DFT + DMFT calculations give $\gamma_{\rm DMFT} = 26.94\,{\rm mJ\,K^{-2}\,mol^{-1}}$, evaluated using the Fermi-liquid formula, $\gamma_{\rm DMFT} = \frac{1}{3}\pi^2 k_{\rm B}^2 \sum_\alpha A_\alpha(\omega = 0)(m^*/m)_\alpha$ reducing the deviation between theory and experiment to 20 % [20].

Our DFT + DMFT results for the momentum-resolved spectral function $A(\mathbf{k},\omega)$ (Fig. 1, bottom) show a coherent distribution of the spectral weight close to $E_{\rm F}$ corresponding to a well-defined Fermi surface (Fig. 1, top). We observe that the effect of electronic correlations on the band structure can be described by a scaling transformation of the energy bands obtained by DFT almost for the entire Brillouin zone in close similarity to parent compounds of FeSC [17, 21, 22]. We note however that unlike FeSC the shift of the bands for specific paths of the reciprocal space in V_2Te_2O is sufficient to induce a collapse of the electronic-like quasi two-dimensional pocket centered at the $\Gamma - Z$ direction.

Finally, we analyze the local spin susceptibility $\chi_\alpha(\tau) = \langle \hat{s}^z_\alpha(\tau)\hat{s}^z_\alpha(0)\rangle$. On the imaginary time interval $\tau \in (0, \beta/2)$ ($\beta = 1/k_{\rm B}T$) the computed $\chi_\alpha(\tau)$ exhibits a sharp decrease followed by an extended flat region close to zero for the $3z^2 - r^2$, xy, and yz states. By contrast, the imaginary time evolution of $\chi_\alpha(\tau)$ for the xz and $x^2 - y^2$ states is less pronounced. This behavior is indicative of localized spin moments in the xz and $x^2 - y^2$ states and more itinerant nature of the $3z^2 - r^2$, xy, and yz states.

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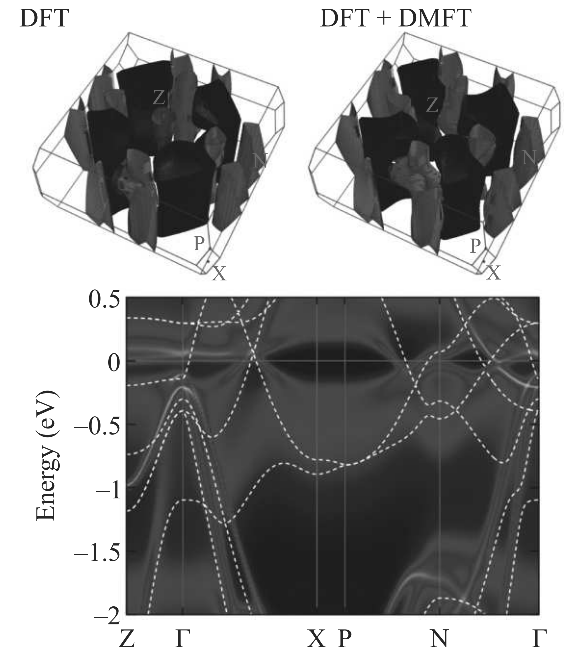

Fig. 1. (Color online) Top: Fermi surface of V_2Te_2O as computed by non-magnetic DFT (left) and paramagnetic DFT + DMFT (right) at $T = 290$ K. Bottom: spectral function $A(\mathbf{k},\omega)$ of paramagnetic V_2Te_2O computed by DFT + DMFT at $T = 290$ K (contours) in comparison with the band structure calculated by non-magnetic DFT (dashed lines)

Conflict of interest. The authors of this work declare that they have no conflicts of interest.

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