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 $\gamma = 26.94 \, \text{mJ} \, \text{K}^{-2} \, \text{mol}^{-1}$ 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.

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 $33.9 \, \text{mJ} \, \text{K}^{-2} \, \text{mol}^{-1}$, 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].

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

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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 $\gamma_{\mathrm{exp}} = 33.9\,\mathrm{mJ\,K^{-2}\,mol^{-1}}$ obtained in the experiment (as compared to that derived from DFT) [9]. Our DFT calculations yield the electronic specific heat coefficient $\gamma_{\mathrm{DFT}} = 9.67\,\mathrm{mJ\,K^{-2}\,mol^{-1}}$, 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 $\gamma_{\mathrm{DMFT}} = 26.94\,\mathrm{mJ\,K^{-2}\,mol^{-1}}$, evaluated using the Fermi-liquid formula, $\gamma_{\mathrm{DMFT}} = \frac{1}{3}\pi^2 k_{\mathrm{B}}^2 \sum_\alpha A_\alpha(\omega = 0)(m^*/m)_\alpha$ 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_{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 $\chi_\alpha(\tau) = \langle \hat{s}_\alpha^z(\tau)\hat{s}_\alpha^z(0)\rangle$. On the imaginary time interval $\tau \in (0, \beta/2)$ ($\beta = 1/k_{\mathrm{B}}T$) 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.

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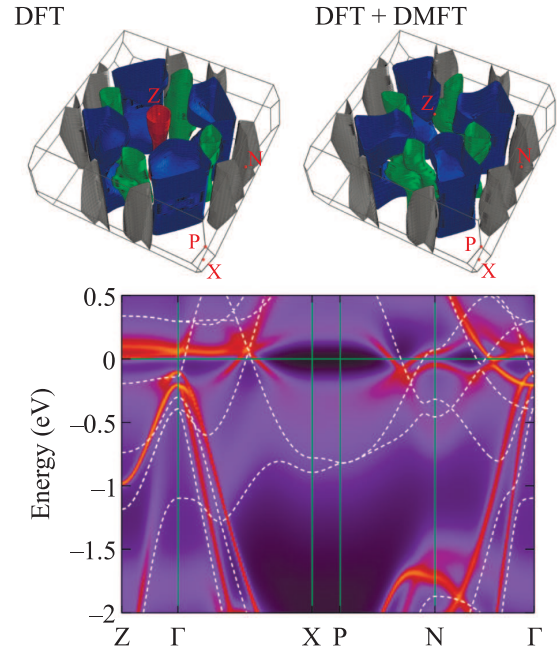

Fig. 1. (Color online) Top: Fermi surface of V_2Te_2O as computed by non-magnetic DFT (left) and paramagnetic DFT + DMFT (right) at $T = 290\,\mathrm{K}$. Bottom: spectral function $A(\mathbf{k},\omega)$ of paramagnetic V_2Te_2O computed by DFT + DMFT at $T = 290\,\mathrm{K}$ (contours) in comparison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

1. S. Z. Butler, S. M. Hollen, L. Cao et al. (Collaboration), ACS Nano **7**, 2898 (2013).

2. K. S. Novoselov, V. I. Falko, L. Colombo, P. R. Gellert, M. G. Schwab, and K. Kim, Nature **490**, 192 (2012).

3. R. M. Fernandes, A. I. Coldea, H. Ding, I. R. Fisher, P. J. Hirschfeld, and G. Kotliar, Nature **601**, 35 (2022).

4. J. A. Wilson and A. D. Yoffe, Adv. Phys. **18**, 193 (1969).

5. T. F. Smith, R. N. Shelton, and R. E. Schwall, J. Phys. F: Met. Phys. **5**, 1713 (1975).

6. C. B. Scruby, P. M. Williams, and G. S. Parry, Philos. Mag. **31**, 255 (1975).

7. I. Guillamón, H. Suderow, S. Vieira, L. Cario, P. Diener, and P. Rodiére, Phys. Rev. Lett. **101**, 166407 (2008).

8. S. Koley, N. Mohanta, and A. Taraphder, Eur. Phys. J. B **93**, 77 (2020).

9. A. Ablimit, Y.-L. Sun, E.-J. Cheng, Ya-B. Liu, S.-Q. Wu, H. Jiang, Z. Ren, S. Li, and G.-H. Cao, Inorg. Chem. **57**, 14617 (2018).

10. H. Lin, J. Si, X. Zhu, K. Cai, H. Li, L. Kong, X. Yu, and H.-H. Wen, Phys. Rev. B **98**, 075132 (2018).

11. S. Manzeli, D. Ovchinnikov, D. Pasquier, O. V. Yazyev, and A. Kis, Nat. Rev. Mater. **2**, 17033 (2017).

12. G. H. Han, D. L. Duong, D. H. Keum, S. J. Yun, and Y. H. Lee, Chem. Rev. **118**, 6297 (2018).

13. M. Valldor, P. Merz, Y. Prots, and W. Schnelle, Eur. J. Inorg. Chem. **2016**, 23 (2016).

14. A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, Rev. Mod. Phys. **68**, 13 (1996).

15. H. Park, A. J. Millis, and C. A. Marianetti, Phys. Rev. B **90**, 235103 (2014).

16. M. V. Sadovskii, Phys.-Uspekhi **51**, 1201 (2008).

17. M. Aichhorn, L. Pourovskii, V. Vildosola, M. Ferrero, O. Parcollet, T. Miyake, A. Georges, and S. Biermann, Phys. Rev. B **80**, 085101 (2009).

18. S. L. Skornyakov, V. I. Anisimov, and D. Vollhardt, Phys. Rev. B **86**, 125124 (2012).

19. S. L. Skornyakov and I. Leonov, Phys. Rev. B **100**, 235123 (2019).

20. L. V. Pourovskii, G. Kotliar, M. I. Katsnelson, and A. I. Lichtenstein, Phys. Rev. B **75**, 235107 (2007).

21. S. L. Skornyakov, N. A. Skorikov, A. V. Lukoyanov, A. O. Shorikov, and V. I. Anisimov, Phys. Rev. B **81**, 174522 (2010).

22. S. L. Skornyakov, D. Y. Novoselov, T. Gürel, and V. I. Anisimov, JETP Lett. **96**, 118 (2012).