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Effect of Coulomb correlations on the electronic structure of bulk V_2Se_2O: a DFT + DMFT study

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We present results of density functional theory (DFT) plus dynamical mean-field theory (DFT + DMFT) calculations of the electronic structure of bulk paramagnetic V_2Se_2O. We show that local Coulomb correlations in the partially filled V $3d$ shells induce renormalizations of the DFT spectral functions close to the Fermi energy preserving their shape. These transformations are not accompanied by a spectral weight transfer to Hubbard bands, indicating a moderately correlated metallic state of bulk paramagnetic V_2Se_2O. The V $3d$ states exhibit a quasiparticle mass enhancement $m^*/m \sim 1.34 - 3.11$ comparable to that in the isostructural compound V_2Te_2O. We demonstrate that orbital selectivity of correlation effects in V_2Se_2O is less pronounced compared to V_2Te_2O as can be traced from the weaker differentiation of m^*/m and local spin correlation functions for different V $3d$ orbitals. The analysis of the temperature dependence of the self-energy allows us to speculate on possible deviations from the Fermi-liquid behavior of V_2Se_2O.

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In the last decades the intense search for novel quantum effects in solids has directed a significant attention of both theoretical and experimental communities toward layered compounds with unique structural and electronic properties [1]. In particular, the discovery of graphene and iron-based superconductors has placed quasi two-dimensional (quasi-2D) materials in the focus of modern condensed matter physics [2–4].

Recently, the class of quasi-2D compounds has been expanded by the new family of van der Waals vanadium oxychalcogenides V_2Ch_2O (Ch = S, Se, Te) [5, 6]. Theoretical research of V_2Ch_2O reported in the literature primarily focus on the case of an antiferromagnetically ordered monolayer and predict its intriguing properties important for spintronics and cathode materials applications emphasizing the importance of electron correlations [7, 8]. The role of correlation effects for explaining the properties of bulk V_2Ch_2O remains, however, much less investigated. Here, we employ a combined method of density functional theory DFT and dynamcal mean-field theory (DFT + DMFT) [9] to explore the effect of Coulomb correlations on the electronic structure of bulk V_2Te_2O.

We perform DFT + DMFT electronic structure calculations of paramagnetic V_2Se_2O using the experimental crystal structure [6] and Coulomb interaction parameters $U = 5$ eV and $J = 0.7$ eV, the same as those employed in [10]. Our DFT + DMFT results for the total spectral function $A(\omega)$ of V_2Se_2O show that Coulomb correlations trigger a notable modification of $A(\omega)$ only in a narrow vicinity of the Fermi energy E_F (in an energy interval $(-1, 1)$ eV relative to E_F). Most importantly, a comparison of $A(\omega)$ from DFT and DFT + DMFT does not reveal a significant reduction of the spectral weight closer to E_F and its localization in Hubbard bands.

Our results for the orbitally-resolved spectral functions $A_\alpha(\omega)$ ($\alpha = 3z^2 - r^2$, xy, xz, $x^2 - y^2$, and yz) show that the effect of Coulomb correlations is most pronounced for the xy, xz, and yz states. The most notable transformations exhibit the spectral functions of the xy and xz states which have the largest amplitude in vicinity of the Fermi energy due to the peaks located close to (in DFT) and almost right at (in DFT + DMFT) E_F. Analyzing the orbital contributions to the momentum-resolved spectral functions $A_\alpha(\mathbf{k}, \omega)$ we demonstrate that the peak of $A(\omega)$ of the xz states originates from two weakly dispersive bands located above and below E_F on the ΓXPNΓ path (Fig. 1). The xy states show the peak of $A(\omega)$ due to the spectral weight in the vicinity of the N point while $A(\omega)$ of the yz states features a precursor of the peak at ~ -0.25 eV. We also note that taking into account the on-site Coulomb interaction in V_2Se_2O yields a collapse of the Fermi surface along the Γ–Z direc-

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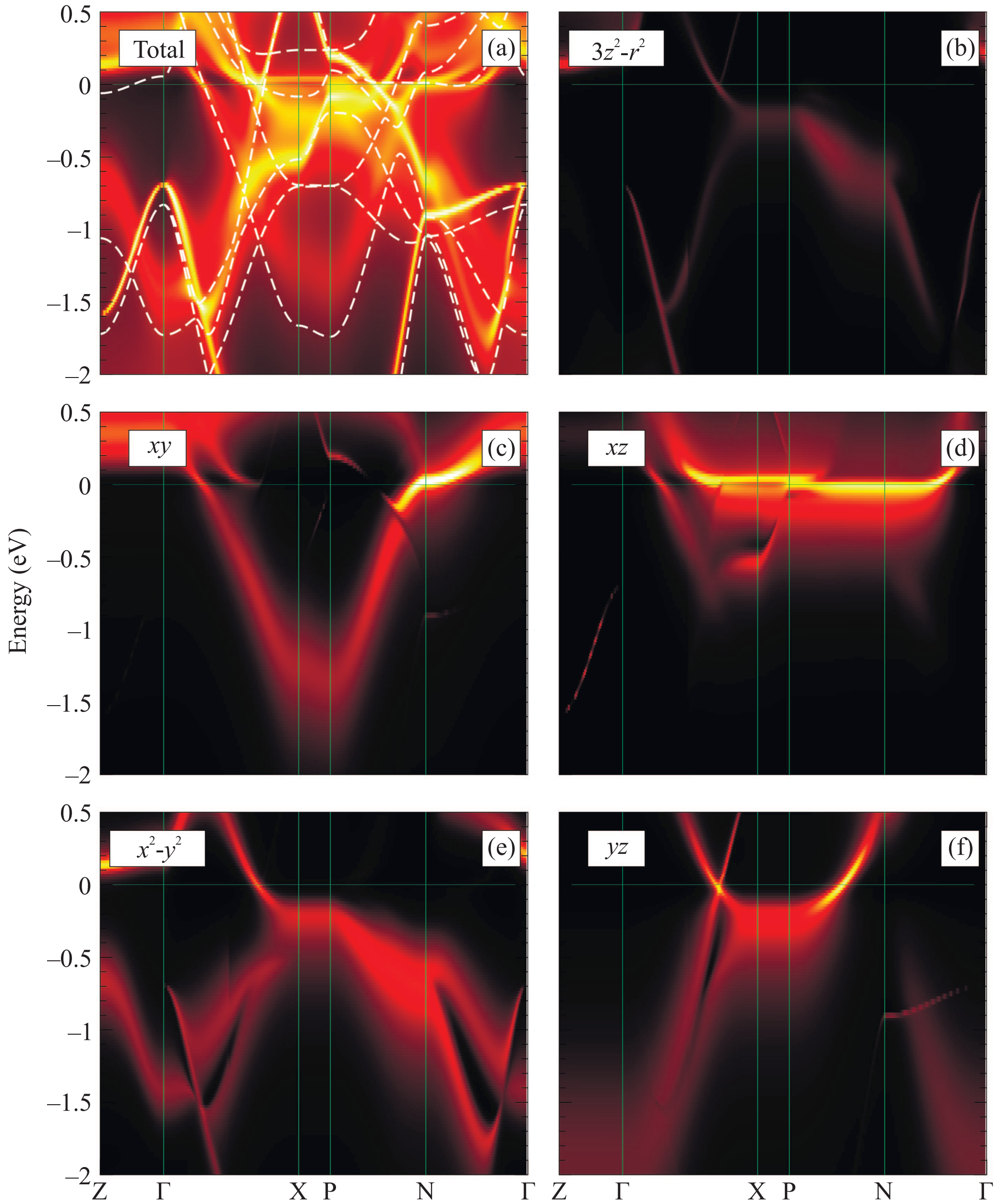

Fig. 1. (Color online) (a) – Total momentum-resolved spectral function $A(\mathbf{k},\omega)$ and (b)–(f) V $3d$ orbital contributions $A_\alpha(\mathbf{k},\omega)$ ($\alpha = 3z^2 - r^2$, xy, xz, $x^2 - y^2$, and yz) of paramagnetic V_2Se_2O computed by DFT + DMFT at $T = 290$ K along high-symmetry directions in the Brillouin zone. The dashed lines corresponds to the band structure of non-magnetic V_2Se_2O calculated by DFT. 0 eV corresponds to the Fermi energy

tion, in close similarity to the isostructural compound V_2Te_2O.

Next, we compute the enhancement of the quasiparticle mass $(m^*/m)_\alpha$ linked to diagonal matrix elements of the self-energy $\Sigma(\omega)$ as $(m^*/m)_\alpha = 1 - -d\mathrm{Re}\Sigma_{\alpha\alpha}(\omega)/d\omega|_{\omega\to 0}$. We obtain that the largest m^*/m of 3.11 corresponds to the xz states followed by the xy, yz, and $x^2 - y^2$ states with m^*/m equal to 2.66, 2.51, and 2.02, respectively. The $3z^2 - r^2$ states exhibit the smallest mass renormalization of 1.34. Computed m^*/m show a smaller variation over the $3d$ shell compared to that in V_2Te_2O signaling a reduction of orbital selectivity of correlation effects upon substitution of Te for Se in V_2Ch_2O. These large values coexist with the absence of spectral weight transfer from the Fermi energy to Hubbard bands and therefore V_2Se_2O should be classified a moderately correlated material.

Our results for the local spin correlator $\chi_\alpha(\tau) = \langle \hat{s}^z_\alpha(\tau)\hat{s}^z_\alpha(0)\rangle$ allow us to distinguish three orbital groups in accord with the different behavior of $\chi_\alpha(\tau)$. Namely, on the imaginary time interval $\tau \in (0, 1/2k_\mathrm{B}T)$ calculated $\chi_{3z^2-r^2}(\tau)$ shows a fast decrease and drops almost to zero, indicative of the itinerant electron behavior while $\chi_\alpha(\tau)$ of the x^2-y^2 and yz states exhibits a slightly weaker decrease and non-zero saturated values at $\tau = 1/2k_\mathrm{B}T$ implying weaker spin fluctuations. Magnetic moments due to the xz and xy states show even better spatial localization as can be traced from a less pronounced variation of their spin correlators along the $(0, 1/2k_\mathrm{B}T)$ interval and larger values at $\tau = 1/2k_\mathrm{B}T$.

To estimate the local moment lifetime we compute the Fourier transform $\chi_\alpha(i\omega_n)$ of $\chi_\alpha(\tau)$ and perform its analytical continuation to the real energies $\chi_\alpha(i\omega_n) \to \chi_\alpha(\omega)$. Our results for $\chi_\alpha(\omega)$ demonstrate that all V $3d$ orbitals except $3z^2-r^2$ exhibit peaks at $\omega = 0$ eV signaling the presence of local moments. The local moment lifetime $\Delta\tau$ can be evaluated from the half-width $\Delta\omega$ at half-height of these peaks via $\Delta\tau\Delta\omega \geq \hbar$. We obtain that the long-lived local moments with $\Delta\tau$ of 16 and 13 fs correspond to the xz and xy states, respectively. The local moment lifetime in the yz and x^2-y^2 states is almost two times shorter (9 and 8 fs) while $\Delta\tau$ for the $3z^2-r^2$ states is less than 1 fs.

Finally, we perform a qualitative analysis of the temperature dependence of the self-energy. We show that at low enough temperatures $Im\hat{\Sigma}_{\alpha\alpha}(i\pi k_\mathrm{B}T)$ is well represented by a linear function of temperature for both V_2Se_2O and V_2Te_2O. In case of V_2Te_2O an extrapolation of this dependence to $T = 0$ K gives zero. The same procedure for V_2Se_2O gives a non-zero value of $Im\hat{\Sigma}_{\alpha\alpha}(i\pi k_\mathrm{B}T)$ at $T \to 0$ for all V $3d$ orbitals except $3z^2-r^2$. According to the so-called "first Matsubara frequency rule" [11] such behavior might signal deviations from the Fermi-liquid regime in V_2Se_2O.

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

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