Dynamical screening in correlated metals: Spectral properties of SrVO₃ in the *GW* approximation and beyond

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SrVO₃ is a prototypical strongly correlated metal. Here we interpret the signatures of electronic correlation measured in photoemission spectroscopy by combining the many-body *GW* approximation of the self-energy with an exponential representation of the Green's function. We explain those correlation effects as the consequence of the dynamical screening of the Coulomb interaction and the coupling between elementary excitations in the solid. Moreover we address the issue of satellites for empty states and discuss the possibility of plasmon-satellite series above the Fermi energy E_F . In good agreement with experiment, we obtain from first principles the renormalization of the V *3d* quasiparticle bands and the satellites close to E_F that so far have been interpreted on the basis of the Hubbard model. Finally, we identify incoherent features due to dynamical correlation also at the bottom of the O *2p* bands, beyond the traditional three-band Hubbard-model description.

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I. INTRODUCTION

Understanding electronic correlations from first principles represents a great challenge for condensed-matter physics. At the same time, strongly correlated materials like transition-metal oxides display a rich variety of physical properties, such as metal-insulator transitions, high-temperature superconductivity, or colossal magnetoresistance, which makes these materials very attractive for a wide range of applications. Strontium vanadate (SrVO₃) has a cubic crystal structure and a simple d^1 electronic configuration, with three V t_{2g} bands crossing the Fermi level that are isolated from the other bands. Thus SrVO₃ has been identified as a benchmark material to study the effects of electronic correlation in metals and as the starting point to understand more complex materials.^{1–25}

A prominent tool for the investigation of electronic excitations in solids is photoemission spectroscopy (PES). In SrVO₃ direct^{1–11} and inverse¹² PES measurements have revealed a three-peak structure of the V t_{2g} states around the Fermi energy E_F . Occupied t_{2g} states are characterized by a main peak at E_F , ~ 0.7 eV wide, and a broad satellite centered at about 1.6 eV binding energy (BE). Moreover, inverse PES shows a further peak at 2.5–3 eV (which actually also overlaps with unoccupied V e_g states²⁶). Their interpretation based on the Hubbard model is today well established. Besides the main quasiparticle peak, the two side peaks are interpreted as the metallic remnants of the lower and upper Hubbard bands. This is the key result obtained also by several dynamical mean-field theory (DMFT) calculations.^{13–25} Therefore, it is usually concluded that SrVO₃ is a strongly correlated metal.

The Hubbard model is a powerful tool to describe features due to electronic correlation in solids and has provided much insight. Within the model, electronic properties are the result of the competition between the tendency for the electrons to localize on a lattice site due to the on-site Coulomb repulsion U and the opposite tendency due to the hopping t to spread over other sites giving rise to a finite bandwidth. However,

the description of correlation based on the Hubbard model is limited to a few electronic degrees of freedom, typically d or f electrons, whereas all the other electrons are treated as uncorrelated, and is restricted to on-site interactions only, neglecting nonlocality effects. Moreover, the screening of the Coulomb interaction is most often taken into account only in its static limit, neglecting dynamical effects in the Hubbard U ,^{25,27–29} which in practice is treated as an adjustable parameter.

In the present work we make use of parameter-free methods of many-body perturbation theory (MBPT) without resorting to the Hubbard model. We reproduce the satellites measured in PES and explain them as a genuine many-body effect due to dynamical screening and to the coupling between different electronic elementary excitations. Moreover, we address the issue of describing and understanding satellites for *empty* states. For the *occupied* valence band in fact it has been shown that the many-body *GW* approximation (GWA)³⁰ can suffer from the presence of spurious plasmaron satellites.^{31–33} Here we analyze what happens also above the Fermi energy, an issue that has been much less studied. To this end we extend also to unoccupied states the exponential representation, also known as cumulant (C) expansion,^{34–40} of the Green's function, recently derived in Ref. 33 from a decoupling approximation. Within the *GW* + C approximation we find a series of plasmon satellites and solve the plasmaron problem also for empty states. Finally, our analysis allows us to put forward additional signatures of dynamical correlation for the occupied O *2p* states at higher binding energies that cannot be described by the traditional three-band Hubbard model in SrVO₃. These results highlight, also for correlated materials, the very promising potentialities of the *ab initio* MBPT approaches that treat all electrons on equal footing.

II. METHODS

The standard model of electronic structure calculations is mostly based on Kohn-Sham (KS) density-functional theory

(DFT) in the local density approximation (LDA).⁴¹ However, KS-DFT is a ground-state theory and is not supposed to yield the electronic excitations measured by PES. Instead, MBPT approaches based on the one-particle Green's function G are in principle designed to obtain the spectral function $A(\omega) = 1/\pi |\text{Im}G(\omega)|$, and hence the measured spectra. To calculate G we make use of the GWA, in which the self-energy $\Sigma(\omega)$ is given by the convolution between $G(\omega)$ and the screened Coulomb potential $W(\omega)$. In this framework the dielectric function $\epsilon(\omega)$ plays a prominent role, by providing the dynamical screening of the static bare Coulomb interaction v : $W(\omega) = \epsilon^{-1}(\omega)v$. In the GWA $W(\omega)$ is calculated in the random phase approximation (RPA).

In the standard G_0W_0 implementation, excitation energies in the GWA are obtained as a first-order perturbative correction to KS LDA. Its applications have been focused mainly on quasiparticle (QP) band-structure calculations of sp semiconductors and metals.⁴² Results on the full spectral function $A(\omega)$ within the GWA are more scarce, due to the computational complexity and the discrepancies with experiments concerning both the position and the intensity of satellite features obtained in the G_0W_0 scheme.^{33–37,43} Advances beyond this standard G_0W_0 implementation have been recently developed, notably the quasiparticle self-consistent GW (QPscGW) scheme.^{44–46} This has allowed the successful application of the GWA also in correlated materials,^{44,45,47–49} where the KS LDA orbitals are often not a good approximation to the QP wave functions and thus the perturbative G_0W_0 is *a priori* not justified.

We performed LDA calculations within a plane-wave scheme (with 100 Hartree energy cutoff) with norm-conserving pseudopotentials.⁵⁰ On top of LDA results we did G_0W_0 and QPscGW calculations using a $8 \times 8 \times 8$ mesh of $\mathbf{k}$ points. In both kinds of GW calculations we have taken into account the dynamical dependence of self-energy by an accurate contour-deformation (CD) integration technique.⁵¹ $W(\omega)$ was sampled with 60 frequencies on the real axis up to 1.5 Hartree and with 18 frequencies along the imaginary axis. Convergence was achieved with 140 and 200 bands for W and Σ , respectively, a 36 Hartree cutoff for the wave functions and 6 Hartree for ϵ^{-1} . In QPscGW 20 bands around E_F were calculated self-consistently.

III. RESULTS

In Fig. 1(a) we compare KS-LDA and G_0W_0 QP band structures. QPscGW results are similar [see cyan lines in Fig. 1(c)], validating the G_0W_0 scheme for the band structure of SrVO_3 . In agreement with experiment, GW induces a renormalization of the t_{2g} bands with a reduction of their LDA bandwidth of ~ 0.5 eV [see Figs. 1(b) and 1(c)]. At the same time, GW also corrects the position of the O $2p$ states below -3 eV, with an increase of the gap with the t_{2g} states [see Fig. 1(a)].

It is known since Hedin's work^{30,52} that dynamical screening in metals tends to reduce the bandwidth. Screening is the result of the polarization of the system, through the formation of electron-hole pairs and collective neutral excitations (e.g., plasmons). It gives rise to spectral structures in the loss function $L(\mathbf{q}, \omega) = -\text{Im}\epsilon^{-1}(\mathbf{q}, \omega)$, which can be

FIG. 1. (Color online) (a) Band structure plot in KS-LDA and G_0W_0 . (b) Zoom on the t_{2g} states. Comparison between LDA, G_0W_0 , QPscGW, and G_0W_0 with several plasmon-pole models (PPM).

measured by inelastic x-ray scattering. The calculated RPA loss function of SrVO_3 is shown in Fig. 2 at a small momentum transfer $\mathbf{q}$.⁵³

FIG. 2. (Color online) Calculated loss function for $\mathbf{q} = (0.16, 0.00, 0.00)$. Arrows: position of the different PPM poles used to calculate the t_{2g} band structure in Figs. 1(b)–1(c). $d-d$ transitions give rise to the peak at $\omega_{dd} = 1.8$ eV.

Following Hedin,³⁸ we can write the correlation part Σ_c of the G_0W_0 self-energy as

$$\Sigma_c(\mathbf{r}, \mathbf{r}', \omega) = \sum_{i,s \neq 0} \frac{\phi_i(\mathbf{r}) \phi_i^*(\mathbf{r}') W_s(\mathbf{r}, \mathbf{r}')}{\omega - \epsilon_i + \omega_s \text{sgn}(\mu - \epsilon_i)}, \quad (1)$$

where ϵ_i and ϕ_i are Kohn-Sham energies and orbitals, respectively. Moreover, ω_s are the neutral energies that correspond to peaks in L (electron-hole transitions or plasmons) and, in turn, to poles in W . W_s are the corresponding amplitudes. In general, the GW self-energy thus contains the coupling with all the possible neutral excitation ω_s in W (which gives rise to the *dynamical screening*). This is also the result of the full-frequency CD integration. Plasmon-pole models (PPM) are instead obtained when the sum over s in Eq. (1) is restricted to a limited number of poles. Therefore, by comparing the full-frequency (CD) and the single-plasmon-pole model (PPM) GW calculations, we can infer the origin of the main contributions to the renormalization of the t_{2g} states.⁵⁴ When the plasmon-pole model energy is $\omega_p = 13$ or 29 eV, representing the main energy loss, the GW PPM results [gray or orange lines in Fig. 1(b)] are intermediate between LDA and the GW CD bands (red and blue lines, respectively). Instead, when $\omega_p = 5$ eV, i.e., the coupling with the low-energy excitations is explicitly accounted for, the GW CD results for t_{2g} bands are retrieved also in the GW PPM calculation [green lines in Fig. 1(c)]. Thus we can conclude that the renormalization of the t_{2g} bands is mainly given by the dynamical screening produced by these low-energy excitations.

To understand the origin of incoherent features measured by PES, a band-structure description, either in LDA or GW , is by definition insufficient. Therefore, we have determined the full spectral function. To this end QPscGW calculations are followed by an additional GW step, in which the complexity and frequency dependence of the GW self-energy are taken into account. The result for the total $A(\omega) = \sum_{\mathbf{n}\mathbf{k}} A_{\mathbf{n}\mathbf{k}}(\omega)$, where

$$A_{\mathbf{n}\mathbf{k}}(\omega) = \frac{1}{\pi} \frac{|\text{Im}\Sigma_{\mathbf{n}\mathbf{k}}(\omega)|}{[\omega - \epsilon_{\mathbf{n}\mathbf{k}}^H - \text{Re}\Sigma_{\mathbf{n}\mathbf{k}}(\omega)]^2 + [\text{Im}\Sigma_{\mathbf{n}\mathbf{k}}(\omega)]^2}, \quad (2)$$

is reported in Fig. 3 (red lines) (here $\epsilon_{\mathbf{n}\mathbf{k}}^H = \epsilon_{\mathbf{n}\mathbf{k}} - V_{\mathbf{n}\mathbf{k}}^{xc}$). We first focus on the satellites of t_{2g} states [see Fig. 3(a)]. Besides the main QP peaks around E_F (see gray line), two side peaks are present, centered at ~ -2 eV and $\sim +3.5$ eV, which we call LS and USP, respectively. In addition, a weak shoulder (US1) at $\sim +2.2$ eV is also visible.

Concerning occupied t_{2g} states, in the GW spectral function $A_{\mathbf{n}\mathbf{k}}(\omega)$ at Γ [red line in Fig. 4(a)], we recognize the same LS satellite as in the total $A(\omega)$ in Fig. 3(a), whereas the corresponding satellite calculated in G_0W_0 is much weaker (orange dashed line). This points out the deficiency of the G_0W_0 scheme for satellites and the need to obtain the self-energy within a scheme where the QP energies entering $\Sigma(\omega)$ are calculated self-consistently.^{38,55} Moreover, we find that the LS satellite occurs in close correspondence to structures in the GW $\text{Im}\Sigma_{\mathbf{n}\mathbf{k}}(\omega)$ (green line), the deviation stemming from $\text{Re}\Sigma_{\mathbf{n}\mathbf{k}}(\omega)$ (cyan line) not being locally a constant function of ω . In turn, the peak in $\text{Im}\Sigma_{\mathbf{n}\mathbf{k}}(\omega)$ derives from the peak in the loss function $L(\omega)$ at $\omega_{dd} = 1.8$ eV (see Fig. 2) that

FIG. 3. (Color online) (a) Spectral function $A(\omega)$ calculated in GW and $GW + C$ (a) for $V t_{2g}$ states and (b) over a wide energy range. In (a) $A(\omega)$ with only QP contributions (with renormalization factors set to 1) is also shown for comparison. In the inset to (b), experimental data from photoemission spectroscopy: PES (1) with $h\nu = 60$ eV photon energy from Ref. 12 and PES (2) with $h\nu = 900$ eV photon energy from Ref. 4, and from bremsstrahlung isochromat spectroscopy (BIS) from Ref. 12.

is due to t_{2g} $d-d$ transitions. The LS satellite is thus the signature of the coupling between the photohole and these neutral $d-d$ excitations. In LDA + DMFT calculations²⁵ in which a dynamical Hubbard $U(\omega)$ is obtained in a constrained RPA approach²⁸ instead, the same $d-d$ transitions are explicitly removed from the screening of Coulomb interaction and the resulting “low-energy model” is solved with a local interaction only.

We now turn to satellites for unoccupied t_{2g} states. By analyzing the spectral function $A_{\mathbf{n}\mathbf{k}}(\omega)$ at R [see Fig. 4(b)], we find that the USP satellite above E_F occurs in correspondence to a minimum of $|\text{Im}\Sigma_{\mathbf{n}\mathbf{k}}(\omega)|$ and is actually due to $|\omega - \epsilon_{\mathbf{n}\mathbf{k}} - \text{Re}\Sigma_{\mathbf{n}\mathbf{k}}(\omega)|$ approaching 0. We thus recognize it as a plasmaron satellite.^{31–33} However, analogous GW results for occupied valence bands call for inquiring whether such a plasmaron satellite above E_F is actually an artifact of the GWA. To this end, we have extended the $GW + C$ approach of Ref. 33 also to empty states. In a single PPM, where $W_{\mathbf{n}\mathbf{k}}(\tau) = -i\lambda_{\mathbf{n}\mathbf{k}}\{e^{-i\omega_{\mathbf{n}\mathbf{k}}\tau}\theta(\tau) + e^{i\omega_{\mathbf{n}\mathbf{k}}\tau}\theta(-\tau)\}$ for each matrix element of W , using a Taylor expansion of the exponential we

FIG. 4. (Color online) Real and imaginary contributions from $\Sigma_{nk}(\omega)$ and corresponding $A_{nk}(\omega)$ (multiplied by 10) in GW (a) for the lowest occupied t_{2g} state at Γ and (b) for the highest empty t_{2g} state at R . At Γ also the perturbative G_0W_0 $A_{nk}(\omega)$ is shown for comparison.

find for $\omega > E_F$:

$$A_{nk}(\omega) = \left| \frac{e^{-a_{nk}}}{\pi} \sum_{m=0}^{\infty} \frac{a_{nk}^m}{m!} \frac{\Gamma_{nk}}{(\omega - E_{nk} - m \tilde{\omega}_{nk})^2 + \Gamma_{nk}^2} \right|, \quad (3)$$

where E_{nk} and Γ_{nk} are the real and imaginary parts of the GW QP energies, respectively, and $a_{nk} = \lambda_{nk}/\tilde{\omega}_{nk}^2$. We thus obtain a series of satellites at energies *higher* than the QP peak, from which they are distant a multiple m of the plasmon-pole energy. This is opposed to core and valence states, where plasmon satellites are located at energies *lower* than the main QP peak.^{34,38–40}

The results of the $GW + C$ calculations, where we go beyond the single PPM of Eq. (3),⁵⁶ are shown in Fig. 3 (blue lines). They compare very well with the GW spectral function below E_F , including the LS satellite. The position of the LS satellite is slightly overestimated with respect to PES, similar to some LDA + DMFT results (also at -2 eV, see, e.g., Ref. 4). However, the comparison with LDA + DMFT depends on the choice of the value of the U parameter used in the Hubbard model. The present calculations are parameter free. The main variations of the $GW + C$ with respect to GW concern the unoccupied t_{2g} states. We observe: (i) the elimination of the spurious GW plasmaron satellite (USP), (ii) the strong

enhancement of the feature US1 at $\sim +2.7$ eV that in GW was only a weak shoulder, and (iii) the appearance at $\sim +4.5$ eV of a weaker replica (US2) of the US1 satellite; US1 and US2 are separated from the QP peak by ω_{dd} and $2\omega_{dd}$, respectively.

By calculating the spectral function over a wide energy range (from -14 to $+9$ eV) [see Fig. 3(b)], we benefit from the great advantage of treating all electrons on equal footing, as done here in GW and $GW + C$. The O $2p$ states in GW are in fact shifted to higher BE with respect to LDA. Thus the t_{2g} LS satellite remains visible also in the total spectral function, in agreement with the experiment. Instead, in other many-body approaches (like LDA + DMFT) that focus only on t_{2g} states and describe all the others at the LDA level, the LS satellite is hidden by O $2p$ states, which remain at their LDA location (see, e.g., Refs. 21 and 24). Moreover, here we find that the satellite US1 associated to the t_{2g} states overlaps in energy with the unoccupied e_g states, and this explains the nature of the peak seen in inverse PES.^{12,26} Finally, taking into account possible correlation effects also among O $2p$ states leads us to discover an incoherent tail (T) at the bottom of the O $2p$ band below -8 eV, visible already in the GWA and further enhanced in the $GW + C$ spectral function. This feature cannot be predicted by the three-band Hubbard model that treats O $2p$ electrons as uncorrelated (thus not even using a dynamical $U(\omega)$ in LDA + DMFT²⁵). In fact, by repeating the same analysis done for the t_{2g} states, we can conclude that this incoherent feature T is due to the coupling of the O $2p$ states with the same neutral $d-d$ excitations that give rise to the satellite close to E_F .

IV. CONCLUSIONS

In summary, our results point towards a general trend for transition-metal oxides in their metallic phases. So far exponential expansions of G have generally been used for occupied states only and for coupling with high-energy plasmons.^{33,34,37–40} Our results demonstrate that the $GW + C$ approach is able to describe also a coupling with low-energy excitations and that also above E_F it is possible to find a series of plasmon satellites. The dynamical screening of the Coulomb interaction is the unifying physical effect that gives rise to the different correlation features seen in the photoemission spectra. In fact, the renormalization of the t_{2g} bands, the satellites in the spectral function around E_F , and the incoherent tail at the bottom of the O $2p$ bands have been all coherently explained in terms of the coupling of the photohole (or the photoelectron in inverse photoemission) with neutral low-energy $d-d$ excitations.

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