Screening from e_g states and antiferromagnetic correlations in $d^{(1,2,3)}$ perovskites: A GW +EDMFT investigation

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We perform a systematic *ab initio* study of the electronic structure of $\text{Sr}(\text{V},\text{Mo},\text{Mn})\text{O}_3$ perovskites, using the parameter-free GW +EDMFT method. This approach self-consistently calculates effective interaction parameters, taking into account screening effects due to nonlocal charge fluctuations. Comparing the results of a three-band (t_{2g}) description to those of a five-band ($t_{2g} + e_g$) model, it is shown that the e_g states have little effect on the low-energy properties and the plasmonic features for the first two compounds but play a more active role in SrMnO_3 . In the case of SrMnO_3 , paramagnetic GW +EDMFT yields a metallic low-temperature solution on the verge of a Mott transition, while antiferromagnetic GW +EDMFT produces an insulating solution with the correct gap size. We discuss the possible implications of this result for the nature of the insulating state above the Néel temperature, and the reliability of the GW +EDMFT scheme.

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I. INTRODUCTION

Transition-metal oxides represent an important test ground for new theoretical and computational schemes aimed at a quantitative description of electron-electron correlations. In this class of compounds, methods based on a single-particle description of the solid do not provide a satisfactory description due to the strong many-body interactions within the partially filled and narrow d or f bands. Emergent properties, such as high-temperature superconductivity and other electronic ordering phenomena, are the result of subtle competitions between different interactions and require an accurate estimation of free energies. Even in the absence of symmetry breaking, basic properties of the solids, such as the metallic or insulating nature of the ground state, cannot be easily predicted [1]. Theoretical models capturing the essential physics need to be developed, solved, and the results compared to experiments.

A widely used approach is the combination of density functional theory (DFT) and dynamical mean-field theory (DMFT) [2,3]. This scheme reduces the problem to a multiorbital Hubbard model with hopping parameters derived using Wannier basis functions. In many of these calculations, the value of the local Coulomb interaction is chosen to reproduce experimentally observed properties, such as mass enhancements or positions of Hubbard bands. With suitably chosen parameters, many properties of correlated materials

can be described by DFT+DMFT [4]. What is not captured by this approach are collective long-range charge fluctuations and the resulting dynamical screening effects. These dynamical long-range correlations are, on the other hand, well described by the GW approximation [5], one of the most successful methods for the study of excited-state properties of weakly correlated compounds, such as semiconductors [6]. After more than a decade of development [7–12], the power of these two methods has been combined into a multitier GW +EDMFT formalism, which is applicable to moderately and strongly correlated materials [12,13]. EDMFT is the extended version of DMFT [14], which allows to treat the effect of long-range interactions. Multitier refers to the fact that different degrees of freedom are treated with different physically motivated approximations: the highest-energy bands with single-shot GW , an intermediate energy window within self-consistent GW , and only the most strongly correlated bands near the Fermi energy within GW +EDMFT. This separation makes the scheme computationally feasible and can be implemented without any double countings. Apart from the choice of these energy windows, multitier GW +EDMFT is free of adjustable parameters, and thus a true *ab initio* method.

To assess the accuracy and predictive power of this approach, it is important to test it on a broad range of compounds, and with different choices of energy windows. Here, we continue the effort started in Refs. [12,13] and present a systematic study of three prototypical perovskite compounds, namely, SrVO_3 , SrMoO_3 , and SrMnO_3 . These materials exhibit different fillings of the t_{2g} orbitals, and hence different correlation effects. SrVO_3 and SrMoO_3 are experimentally found to be paramagnetic metals [15–18], while SrMnO_3 is an insulator with a local moment which orders antiferromagnetically below 260 K [19–22]. DFT+DMFT based modeling of

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these materials often only considered the t_{2g} shell [23–26], but here we also explore the effect of including the almost empty e_g states. This provides a consistency check for the multitier scheme since the three-band and five-band treatments should produce consistent results for the low-energy electronic structure.

As far as the methodology is concerned, detailed descriptions can be found in Ref. [13]. One extension in this work is the implementation of a self-consistency loop with two sublattices, which allows to stabilize solutions with antiferromagnetic order.

The paper is organized as follows: In Sec. II we briefly summarize the method, the steps of the self-consistency loop, and the rationale behind the multitier subdivision of the orbital space. In Sec. III we compare the results of the five- and three-band models for SrVO₃, SrMoO₃, and SrMnO₃. In Sec. IV we present our conclusions.

II. METHOD

In this section we give a brief overview of the multitier GW +EDMFT method developed in Refs. [12,13] and explain its extension to antiferromagnetically ordered systems.

A. GW +EDMFT

By defining a set of localized wave functions $w_{i\mathbf{R}}(\mathbf{r})$, where i is an orbital index and $\mathbf{R}$ is a site index, the self-energy Σ and polarization Π can be divided into the local (onsite) components Σ^{loc} , Π^{loc} , and the remaining nonlocal components

$$\Sigma_{i,j}(\mathbf{k}, i\nu) = \Sigma_{i,j}^{\text{loc}}(i\nu) + \Sigma_{i,j}^{\text{nonloc}}(\mathbf{k}, i\nu), \quad (1)$$

$$\Pi_{\alpha,\beta}(\mathbf{k}, i\omega) = \Pi_{\alpha,\beta}^{\text{loc}}(i\omega) + \Pi_{\alpha,\beta}^{\text{nonloc}}(\mathbf{k}, i\omega). \quad (2)$$

The Greek indices α, β denote a product basis, $\alpha = \{i, j\}$, necessary to expand the two-particle functions, and we have assumed that two basis functions localized on different sites do not overlap. The self-energy and polarization are related to the Green's function G and screened interaction W through the Dyson equations

$$G = G^0 + G^0 \Sigma G, \quad (3)$$

$$W = v + v \Pi W, \quad (4)$$

where G^0 is the bare propagator and v the bare Coulomb interaction. The key approximation in EDMFT is that the nonlocal components of Σ and Π are negligible, $\Sigma = \Sigma^{\text{loc}}$ and $\Pi = \Pi^{\text{loc}}$. With these approximations, the full lattice problem can be mapped to an effective local impurity problem with a dynamical bare propagator $\mathcal{G}(i\nu)$ and a dynamical bare impurity interaction $\mathcal{U}(i\omega)$. These so-called Weiss fields are determined self-consistently such that the impurity Green's function reproduces the local lattice Green's function $G^{\text{imp}} = G^{\text{loc}}$ and correspondingly for the screened interaction $W^{\text{imp}} = W^{\text{loc}}$.

GW +EDMFT can be regarded as an extension of EDMFT, where the nonlocal components are accounted for within the GW approximation:

$$\begin{aligned} \Sigma_{ik}^{\text{nonloc}}(\mathbf{q}, \tau) = & - \sum_{\mathbf{k}j\mathbf{l}} G_{j\mathbf{l}}(\mathbf{k}, \tau) W_{ijkl}(\mathbf{q} - \mathbf{k}, \tau) \\ & + \sum_{j\mathbf{l}} G_{j\mathbf{l}}^{\text{loc}}(\tau) W_{ijkl}^{\text{loc}}(\tau), \end{aligned} \quad (5)$$

$$\begin{aligned} \Pi_{mm'nn'}^{\text{nonloc}}(\mathbf{q}, \tau) = & \sum_{\mathbf{k}} G_{mn}(\mathbf{k}, \tau) G_{n'm'}(\mathbf{k} - \mathbf{q}, -\tau) \\ & - G_{mn}^{\text{loc}}(\tau) G_{n'm'}^{\text{loc}}(-\tau). \end{aligned} \quad (6)$$

The matrix elements of the screened interaction are defined as

$$\begin{aligned} W_{ijkl}(\mathbf{q}, i\omega) = & \sum_{\mathbf{R}, \mathbf{R}'} e^{i\mathbf{q}(\mathbf{R}-\mathbf{R}')} \int d\mathbf{r} d\mathbf{r}' w_{i\mathbf{R}}^*(\mathbf{r}) w_{j\mathbf{R}}(\mathbf{r}) \\ & \times W(\mathbf{r}, \mathbf{r}', i\omega) w_{k\mathbf{R}'}(\mathbf{r}') w_{l\mathbf{R}'}^*(\mathbf{r}'), \end{aligned} \quad (7)$$

where we once again have assumed that two basis functions localized on different sites have zero overlap.

The GW +EDMFT self-consistency cycle contains the following steps:

- (1) Start with an initial guess for Σ^{imp} , Π^{imp} , and $G_{\mathbf{k}}$. $\Sigma^{\text{loc}} = \Sigma^{\text{imp}}$ and $\Pi^{\text{loc}} = \Pi^{\text{imp}}$ (EDMFT approximations).
- (2) Compute Σ^{nonloc} and Π^{nonloc} according to Eqs. (5) and (6).
- (3) Define $\Sigma_{\mathbf{k}} = \Sigma^{\text{imp}} + \Sigma_{\mathbf{k}}^{\text{nonloc}}$ and $\Pi_{\mathbf{q}} = \Pi^{\text{imp}} + \Pi_{\mathbf{q}}^{\text{nonloc}}$ (GW +EDMFT approximations).
- (4) Calculate $G_{\mathbf{k}} = [(G_{\mathbf{k}}^{(0)})^{-1} - \Sigma_{\mathbf{k}}]^{-1}$ and $W_{\mathbf{q}} = v_{\mathbf{q}}(\mathbb{1} - \Pi_{\mathbf{q}} v_{\mathbf{q}})^{-1}$.
- (5) Using $G^{\text{loc}} = \frac{1}{N} \sum_{\mathbf{k}} G_{\mathbf{k}}$ and $W^{\text{loc}} = \frac{1}{N} \sum_{\mathbf{q}} W_{\mathbf{q}}$ calculate the fermionic Weiss field

$$\mathcal{G} = [\Sigma^{\text{imp}} + (G^{\text{loc}})^{-1}]^{-1} \quad (8)$$

and the effective impurity interaction

$$\mathcal{U} = W^{\text{loc}}(\mathbb{1} + \Pi^{\text{imp}} W^{\text{loc}})^{-1}. \quad (9)$$

- (6) Numerically solve the impurity problem to obtain G^{imp} and the impurity charge susceptibility $\chi^{\text{imp}} = \langle \hat{n}(\tau) \hat{n}(0) \rangle$.

- (7) Use the current $\mathcal{G}$ and $\mathcal{U}$ to calculate $\Sigma^{\text{imp}} = \mathcal{G}^{-1} - (G^{\text{imp}})^{-1}$, $\Pi^{\text{imp}} = \chi^{\text{imp}}(\mathcal{U} \chi^{\text{imp}} - \mathbb{1})^{-1}$, and $W^{\text{imp}} = \mathcal{U} - \mathcal{U} \chi^{\text{imp}} \mathcal{U}$.

- (8) If the self-consistency conditions $G^{\text{imp}} = G^{\text{loc}}$ and $W^{\text{imp}} = W^{\text{loc}}$ are not fulfilled within a given tolerance, go back to step 2.

B. Multitier approach

If the self-consistency cycle is performed in the complete Hilbert space, the GW +EDMFT formalism is derivable from a free-energy functional Ψ and is hence conserving [7]. However, in practice this is not feasible. To overcome this problem, a multitier implementation was developed in Refs. [12,13]. In this approach, the complete Hilbert space is divided into three different subspaces, each treated at a different level of approximation. Correspondingly the calculations are divided into three tiers which refer to the different subspaces. The multitier approach is a systematic downfolding procedure from the complete Hilbert space to a smaller subspace and

includes well-defined double-counting corrections at each step:

(1) Tier III: First, a DFT calculation is performed using the FLAPW code FLEUR [27]. Based on the DFT band structure we compute the one-shot GW self-energy $\Sigma^{G^0W^0}$ using the SPEX code [27,28]. Then, an intermediate- or low-energy subspace I , which includes $O(10)$ bands around the Fermi energy, is defined using maximally localized Wannier functions as implemented in the WANNIER90library [29–32]. The effective Coulomb interaction U on the intermediate subspace is computed within the constrained random-phase approximation (cRPA) [33] using the SPEXcode. The G^0W^0 self-energy contribution from within the intermediate subspace is removed from $\Sigma^{G^0W^0}$ to define an effective bare propagator G_k^0 for the intermediate subspace.

(2) Tier II: In the intermediate subspace the self-energy is calculated using a custom self-consistent GW implementation (see Ref. [13]). A correlated subspace C , which can be smaller or equal to the intermediate subspace, is defined. The local part of the GW self-energy and polarization from within the correlated subspace is subtracted to define the effective bare propagator and effective bare interaction for C .

(3) Tier I: At each step of the self-consistency cycle, local corrections to the self-energy and polarization in the correlated subspace C are computed using EDMFT. The effective impurity problem is solved using the CT-Hyb [34–36] quantum Monte Carlo algorithm implemented in ALPS [37–39], while the self-consistency equations make use of the TRIQS framework [40]. The EDMFT calculation provides local corrections to the self-energy and polarization within the correlated subspace.

The complete expressions for the Green's function and screened interactions are as follows:

$$G_k^{-1} = \overbrace{i\omega_n + \mu - \varepsilon_k^{\text{DFT}} + V_{\text{XC},k}}^{\text{TIER III, } G_{I,k}^0{}^{-1}} - \underbrace{(\Sigma_k^{G^0W^0} - \Sigma_k^{G^0W^0}|_I)}_{-\Sigma_{r,k}} - \underbrace{(\Sigma_k^{GW}|_I - \Sigma_k^{GW}|_{C,\text{loc}} + \Delta V_H|_I)}_{\text{TIER II}} - \underbrace{\Sigma^{\text{EDMFT}}|_{C,\text{loc}}}_{\text{TIER I}}, \quad (10)$$

$$W_q^{-1} = \overbrace{v_q^{-1} - (\Pi_q^{G^0G^0} - \Pi_q^{G^0G^0}|_I)}^{\text{TIER III, } U_{I,q}^{-1}} - \underbrace{\Pi_{r,q}}_{-\Pi_{r,q}} - \underbrace{(\Pi_q^{GG}|_I - \Pi_q^{GG}|_{C,\text{loc}})}_{\text{TIER II}} - \underbrace{\Pi^{\text{EDMFT}}|_{C,\text{loc}}}_{\text{TIER I}}. \quad (11)$$

The self-energies in Eq. (10) only contain the exchange and correlation parts, while $\Delta V_H|_I$ represents the change of the Hartree potential within the intermediate subspace (see Ref. [13] for a detailed description). $V_{\text{XC},k}$ is the exchange-correlation potential from the DFT calculation. The notation $A|_I$ means that all internal sums when evaluating A are restricted to the subspace I . In all the setups presented in this study, the intermediate and correlated subspaces are the same.

C. Antiferromagnetic extension

Strongly correlated multiorbital systems at certain integer fillings tend to develop long-range magnetic ordering. In particular, on a bipartite lattice, there is a strong tendency to antiferromagnetic order at half-filling. At sufficiently low temperature, we then expect the appearance of a solution with a local spin polarization. A staggered magnetization on a bipartite lattice can be easily treated in DMFT [2] by considering two sublattices A and B and imposing the following relation between the self-energies:

$$\Sigma_{\uparrow}^A = \Sigma_{\downarrow}^B, \quad \Sigma_{\downarrow}^A = \Sigma_{\uparrow}^B. \quad (12)$$

This allows to reduce the EDMFT calculation to the solution of a single impurity problem, while the unit cell used in the lattice self-consistency has to be doubled. We extended the multitier formalism in order to include this kind of long-range spin ordering by doubling the unit cell of the GW calculations in tier III and tier II, which doubles the size of the lattice Green's function G_k and screened interaction W_q . The calculation in tier III (which provides the input for tier II) is kept paramagnetic, but we allow for a spin symmetry breaking at the EDMFT level in tier I, which feeds back into tier II. Hence, in tier I, we introduce spin-dependent self-energies

$$\Sigma^{\text{EDMFT}}|_{C,\text{loc}} \longrightarrow \Sigma^{\text{EDMFT}}|_{\uparrow,\downarrow}|_{C,\text{loc}}. \quad (13)$$

We do not need to apply any seed since the Monte Carlo errors enable a symmetry breaking in the self-consistent calculation. The spin-dependent local self-energies are then associated with the two sites of the lattice Green's function. Since the decoupling of the long-range interaction is in the charge channel, implying that the local vertex $\Pi^{\text{EDMFT}}|_{C,\text{loc}}$ is computed via the local charge susceptibility, all the two-particle fields remain symmetric with respect to the spin index.

We will apply this extension only to SrMnO_3 , which meets the requirements for antiferromagnetic order in terms of filling and interaction strength, and which is experimentally found to be in an antiferromagnetic phase at low temperatures.

III. RESULTS

In the following, we will investigate the three perovskite compounds SrVO_3 , SrMoO_3 , and SrMnO_3 using two different low-energy models. In the $t_{2g} + e_g$ model the correlated subspace (tier I) contains all five d orbitals, while in the t_{2g} model, it is restricted to the t_{2g} subshell. The calculations for SrVO_3 and SrMoO_3 are performed at inverse temperature $\beta = 10 \text{ eV}^{-1}$, corresponding to $T = 1160 \text{ K}$, while in the case of SrMnO_3 we use $\beta = 40 \text{ eV}^{-1}$ corresponding to $T = 290 \text{ K}$. The latter value is close to $T_{\text{Néel}} \sim 260 \text{ K}$ for SrMnO_3 [19–22], whose magnetic moments order antiferromagnetically in all directions (G-type ordering).

A. SrVO_3 and SrMoO_3

SrVO_3 is one of the simplest and most extensively studied correlated compounds due to its undistorted cubic lattice structure [8,10,15–17,23–26,41]. The conduction band

FIG. 1. Local and $\mathbf{k}$ -resolved spectral function of SrVO₃ (left) and SrMoO₃ (right) in the three- (top) and five-band (bottom) descriptions. Thin black lines represent the LDA band structure.

is formed by vanadium $3d$ states of t_{2g} character which are populated by one electron per unit cell. Within the local density approximation (LDA), the conduction band is isolated with a bandwidth of roughly 2.2 eV. The 3.8-eV-wide conduction band in SrMoO₃ originates from the t_{2g} states of the molybdenum cations which are in a $4d^2$ configuration. In both systems, the DFT calculation predicts empty e_g states which start at about 1 eV above the Fermi level. The main difference between the two perovskites is thus the filling and the width of the t_{2g} band. The experimental photoemission (PES) and inverse photoemission (IPES) spectra of SrVO₃ display a renormalized quasiparticle peak, corresponding to an effective mass enhancement of approximately 2, a pronounced upper satellite feature at roughly 3 eV, and a very weak lower satellite feature at around -1.5 eV [15–17,26]. SrMoO₃, on the other hand, exhibits a very weakly renormalized quasiparticle peak and a pronounced shoulder structure in the PES [18]. The satellite features in the local spectral function of both systems clearly indicate correlation effects beyond the LDA. For SrMoO₃ it was shown in Ref. [18] that the satellite features cannot be described as Hubbard bands. A later publication, using the same GW+EDMFT multitier formalism as employed in this paper, showed that the satellites in this compound should instead be interpreted as plasmon satellites originating from long-range charge fluctuations [13]. This is consistent with the conclusions of Ref. [18], and hence there is a relative consensus in the literature on the nature of the satellite features in the spectral function of SrMoO₃. This is not the case for SrVO₃, where the origin of the satellite features is still under debate. For this compound the $3d$ valence states [and hence also the maximally localized Wannier functions (MLWFs) constructed from the t_{2g} bands] are relatively localized around the V ion. In cRPA this yields an effective Coulomb interaction with a static value of 3.4 eV. LDA+DMFT calculations in which the value of U was chosen to reproduce the experimental effective mass enhancement [23–26], as well as *ab initio* one-shot combinations of GW and DMFT [8,10,41] can roughly reproduce the band narrowing

and the lower satellite, but place the upper satellite observed in IPES too close to the Fermi energy. In Ref. [10] it was instead speculated that the upper satellite could originate from the e_g states. Common to all the above-mentioned calculations is that the lower satellite in the SrVO₃ spectral function was interpreted as a Hubbard band because of the strong local correlations between t_{2g} electrons on the same V site, while the upper satellite was either interpreted as originating from the e_g states or left unexplained.

The interpretation of the satellites as Hubbard bands may be related to the fact that DFT+DMFT calculations only include local correlations in the solution of the low-energy model. On the other hand, the satellite structures of SrVO₃ are well described by the cumulant expansion [42], which is an expansion of the Green's function that is based on the GW approximation of the self-energy [43,44]. Because the GW method does not capture the strong local correlations that give rise to Hubbard bands, these satellite features should be interpreted as plasmons.

Multitier GW+EDMFT considers both types of correlations and therefore is a good scheme to investigate the origin of the satellites. In Refs. [12,13] it was shown that the GW+EDMFT multitier technique yields high-energy satellites which are most naturally explained in terms of plasmonic excitations when the intermediate and correlated subspaces include only the t_{2g} orbitals. This conclusion is supported by the relatively small value of the self-consistently computed local interaction $\mathcal{U}(0)$, which cannot explain those structures as Hubbard bands, while reproducing the experimental mass enhancement relatively well (the band renormalization is slightly too small). In Fig. 1 we show the local and $\mathbf{k}$ -resolved spectral functions for SrVO₃ and SrMoO₃ obtained from the three- and five-band calculations. Focusing on the spectral function associated with the t_{2g} states, we see that the inclusion of the e_g orbitals has no significant effect on the partial t_{2g} spectral function. In particular, the position and strength of the satellite features is similar in the three- and five-band models. The fact that the satellites at 3 eV in the

FIG. 2. Frequency-dependent cRPA interaction $U_{\text{cRPA}}^{t_{2g}}(i\omega_n)$, Hubbard $U^{t_{2g}}(i\omega_n)$, and Hund's $J^{t_{2g}}(i\omega_n)$ component (dashed) of the local effective interaction and screened interactions $W_{\text{loc}}^{t_{2g}}(i\omega_n)$ for SrVO₃ (top) and SrMoO₃ (bottom) in the three- [(a) and (c)] and five-band [(b) and (d)] descriptions.

local spectral function follow the dispersion of the unoccupied part of the quasiparticle bands is consistent with the plasmon scenario. This interpretation is furthermore supported by the fact that the emerging high-energy structures are not particle-hole symmetric, while for Hubbard bands this asymmetry is expected to be less pronounced.

We find that the crystal-field splitting between the two manifolds is significantly enhanced by correlation effects in the case of SrMoO₃, while for SrVO₃ the e_g states remain at the same position as in the LDA band structure.

In Fig. 2 we show the frequency-dependent interaction along the Matsubara axis. The results of the three- and five-band calculations for SrMoO₃ do not show any significant difference, probably as a consequence of the correlation enhanced crystal-field splitting, which decouples the t_{2g} and e_g bands. In the case of SrVO₃, the screening effects on the local t_{2g} interaction $U^{t_{2g}}(0)$ coming from the inclusion of the e_g orbitals are more pronounced. Here, the five-band system is characterized by $U^{t_{2g}}(0) = 2.4$ eV versus $U^{t_{2g}}(0) = 2.1$ eV in the three-band case. This difference is bigger than the difference in the screened cRPA interaction provided as input (see red line). The relatively large difference in $U^{t_{2g}}(0)$ may be related to the pole at low frequency discussed in Ref. [13]. A small shift of this pole to lower frequencies can lead to substantial changes in the static value of the interaction (the results for $U^{t_{2g}}$ above the pole are very similar in both models). In spite of this difference in the local interaction strength, long-range charge fluctuations lead to an almost complete screening of the interaction, i.e., an almost complete vanishing of $W_{\text{loc}}^{t_{2g}}(0)$.

The similarity between the three- and five-band results is also seen in the broad pole structure in $-\text{Im}W_{\text{loc}}(\omega)$ (Fig. 3) which provides a consistent explanation of the satellites in the

FIG. 3. Thick lines represent the imaginary part of the fully screened interaction on the real frequency axis $-\text{Im}W_{\text{loc}}^{t_{2g}}(\omega)$ for SrVO₃ and SrMoO₃ while thin lines in the same color code indicate the imaginary part of the initial cRPA interaction $-\text{Im}U_{\text{cRPA}}^{t_{2g}}(\omega)$.

spectral function in terms of long-range charge fluctuations. Also, in agreement with our previous studies on the same compounds [12,13], the plasmon peak is higher in SrVO₃ indicating stronger screening effects compared to SrMoO₃.

We conclude from this analysis that the three- and five-band calculations yield consistent interpretations of the satellite features in these compounds as plasmons rather than Hubbard bands. The presented data also provide a convincing check for the validity of the downfolding procedure in the multitier approach.

B. SrMnO₃

1. Results for paramagnetic SrMnO₃

There is a substantial level of agreement on the importance of electronic correlations in SrMnO₃, while their role in determining the experimentally observed insulating ground state is still debated. Previous studies on this material [45–47] employed DFT for the structural properties or DFT+DMFT to incorporate the effect of Hubbard-type interactions. A common aspect of all these studies has been the *ad hoc* choice of the Hubbard interaction U and Hund coupling J_H , which were chosen with the goal of reproducing experimental observations like the band gap or the magnetic moment.

In the following, we apply the fully self-consistent and parameter-free GW+EDMFT approach to the three- and five-band models of SrMnO₃ which, at first glance, appear

FIG. 4. Frequency-dependent cRPA interaction $U_{\text{cRPA}}^{t_{2g}}(i\omega_n)$, Hubbard $U^{t_{2g}}(i\omega_n)$, and Hund's $J^{t_{2g}}(i\omega_n)$ (dotted blue line) components of the local effective interaction and screened interaction $W_{\text{loc}}^{t_{2g}}(i\omega_n)$ for SrMnO₃ in the three- (a) and five-band (b) cases. Panel (c) shows the local spectral function for the different correlated subspaces. In panel (d) the imaginary parts of the cRPA interaction (thin lines) and the fully screened (thick lines) interactions are shown on the real-frequency axis.

quite similar to the models described above. In SrMnO₃, three electrons populate the t_{2g} states, hosted by the manganese cation, which results in a 2.5-eV-wide band. In contrast to the previous systems, already at the DFT level, the e_g manifold crosses the Fermi level, which puts a question mark behind the validity of a three-band description for this compound.

This observation also suggests that the e_g states provide non-negligible screening channels. Indeed, the static values of the cRPA interaction reported in Fig. 4 are quite different for the two models, namely, $U_{\text{cRPA}}(0) = 1.9$ eV in the three-band model and $U_{\text{cRPA}}(0) = 2.7$ eV in the five-band model. We also notice that, even though the effective local interaction U is similar for the two models, the fully screened interaction $W_{\text{loc}}^{t_{2g}}$ is substantially smaller in the five-band model. This indicates a more metallic behavior of the five-band model due to enhanced screening within the low-energy subspace.

Similar conclusions can be reached from the lower right panel of the same figure showing $\text{Im}W_{\text{loc}}^{t_{2g}}$ along the real frequency axis. $\text{Im}W_{\text{loc}}^{t_{2g}}$ closely follows the cRPA U for low frequencies (<1 eV), which indicates that the metallic screening within the t_{2g} subspace is weak in the three-band model. In this case, the dominant low-energy screening is in the t_{2g} - e_g channel which is incorporated into the cRPA interaction. On the other hand, in the five-band model the low-energy screening is stronger even though the e_g states are pushed up in energy [see Fig. 4(c)] and therefore should not contribute significantly to screening channels below 1 eV. Hence, in the five-band model, the screening within the t_{2g} subspace is enhanced. This can be understood from the larger weight of the quasiparticle peak in the five-band case (see Fig. 5 and the discussion below) which corresponds to an increased t_{2g} spectral weight around the Fermi energy. Again, this indicates that an effective model containing only the t_{2g} states might be insufficient in describing the screening effects in paramagnetic SrMnO₃.

In Fig. 5 we show the local and $\mathbf{k}$ -resolved spectral functions of SrMnO₃ in the paramagnetic phase obtained by the GW+EDMFT approach and compare it with the result from a single-shot GW calculation. The latter yields a dispersion similar to SrVO₃ and SrMoO₃ (see Ref. [13]) with a smaller

FIG. 5. Local and $\mathbf{k}$ -resolved spectral function of SrMnO₃ obtained using the GW+EDMFT method (left) and with single-shot GW (right) in the three- (top) and five-band (bottom) descriptions. Thin black lines represent the LDA band structure.

bandwidth and a plasmonic broadening occurring mainly in the proximity of the Γ and X points. The inclusion of local vertex corrections beyond GW , however, has striking effects on SrMnO_3 : the near Mottness of the compound, in both the three- and five-band models alike, manifests itself with the formation of broad structures centered at ± 1 eV and with an extremely narrow peak at the Fermi level. We identify these features as Hubbard bands considering that their separation agrees with the magnitude of the static local effective interaction $U^{t_{2g}}(0) = 1.8$ eV. In addition, especially in the three-band model, within each of the three main structures (the two Hubbard bands at $\sim \pm 1$ eV and the narrow quasiparticle band) it is possible to recognize renormalized and/or broadened replicas of the noninteracting dispersion. This behavior is typical of the Mott transition scenario [2]. The asymmetry between the occupied and unoccupied parts of the spectra appears to be a consequence of the GW -derived $\mathbf{k}$ -dependent self-energy, which is known to produce such effects [48]. It is also worth noting that these strong correlation effects occur even though the ratio between $U^{t_{2g}}(0)$ and the bandwidth is similar to the previous two compounds. This is a Hund coupling effect, which leads to a suppression of the kinetic energy at half-filling. As a result, the critical interaction for the Mott transition in a multiorbital system with $J_H > 0$ is lowest at half-filling [49].

The low-energy structures in $\text{Im}W_{\text{loc}}(\omega)$ will give rise to weak satellites (or broad tails) on the high-energy side of the Hubbard bands, a feature seen in the local t_{2g} spectral function shown in Fig. 5. Similar physics was investigated for model systems in Ref. [11], where it was shown that a Mott gap in the fermionic spectral function is associated with a peak in $\text{Im}\mathcal{U}(\omega)$ at ω corresponding to the characteristic energy for charge excitations across this gap. In the presence of a quasiparticle band, there are also screening modes associated with transitions between the quasiparticle band and the Hubbard bands. The situation for real materials is, as discussed above, more complicated since there are multiple screening channels giving rise to different peaks in $\text{Im}W(\omega)$ and a careful analysis of the different screening channels is needed to clarify the origin of the satellite features.

In contrast to the results in previous LDA and LDA+DMFT studies with *ad hoc* parameters [45,46], both our models of SrMnO_3 remain conducting in the paramagnetic phase. The metallicity is due to a very narrow quasiparticle band pinned at the chemical potential. The quasiparticle peak in the five-band case is a bit larger [see Fig. 4(c)] and, as a result, the screened interaction $W_{\text{loc}}^{t_{2g}}$ is smaller [Fig. 4(d)]. Similarly to the case of SrMoO_3 , the e_g center of mass is shifted to higher energies compared to LDA. However, a low amount of e_g spectral weight remains in the occupied part of the spectral function and is responsible for the self-doping effect on the t_{2g} states, as one can infer from the peak at the Fermi level not being at the center of the gap. The self-doping from the e_g states alters the partial t_{2g} filling slightly away from half-filling which tends to reduce the local correlations, as discussed above. Hence, it is this self-doping that is responsible for the larger quasiparticle peak in the five-band model. Finally, we observe that also the increase of the crystal-field splitting, if compared with the previous compounds, can be explained by the proximity of SrMnO_3 to the Mott state, which would

FIG. 6. $\mathbf{k}$ -resolved spectral function of a three-band model of SrMnO_3 with long-range antiferromagnetic order.

require an empty e_g manifold. The same explanation would hold also for the five-band models of $\text{Sr}(\text{V},\text{Mo})\text{O}_3$, which are relatively far from the Mott state as far as it concerns the combination of filling and interaction strength. From these observations we conclude that, if restricted to the paramagnetic case, the GW +EDMFT approach applied to the three- and five-band models of SrMnO_3 yields a metal on the verge of a Mott transition, in which the e_g bands play an active role in determining the overall physics of the system.

2. Antiferromagnetic phase of SrMnO_3

By construction, the paramagnetic GW +EDMFT calculation cannot account for the magnetic ground state which is experimentally observed in the cubic phase of SrMnO_3 [19–22]. Measurements of the magnetic moment report a value of $2.6 \pm 0.2 \mu_B$ and previous DFT+DMFT calculations yield compatible results [45]. The low-temperature behavior has been reported to be well described by the ordering of $S = \frac{3}{2}$ local moments with a $T_{\text{Néel}}$ between 233 and 260 K. The variations in the ordering temperature can be accounted for if oxygen defects are considered.

To describe antiferromagnetic ordering we extended the GW +EDMFT multiter approach to a bipartite lattice as described in Sec. II C. At $\beta = 40 \text{ eV}^{-1}$, the solution with G-type antiferromagnetism self-consistently emerges in our parameter-free simulation. The local and $\mathbf{k}$ -resolved spectral functions of the three-band model are reported in Fig. 6 and exhibit a gap of about 0.5 eV, as well as pronounced features at ± 2 eV. The position of the lower satellite is in good agreement with the PES measurements of Refs. [21,22], which were however taken above $T_{\text{Néel}}$. There are spectral weight tails up to very high energy, consistent with plasmonic sidebands. From the imbalance in the spin population on a given site we compute the magnetic moment as

$$m = \sum_{\alpha} (n_{\alpha\uparrow} - n_{\alpha\downarrow}) \mu_B, \quad (14)$$

where α is the orbital index. The result is $2.05 \mu_B$ on both sublattices. This is in reasonably close agreement with the above-quoted $2.6 \mu_B$ which has been experimentally found at a much lower temperature [19]. Due to numerical stability issues, we could not go below 290 K in our calculations, which means that our simulation results are a bit above the experimental $T_{\text{Néel}}$. However, in mean-field-based approaches

FIG. 7. (a) Frequency-dependent cRPA interaction $U_{\text{cRPA}}^{t_{2g}}(i\omega_n)$, Hubbard $U^{t_{2g}}(i\omega_n)$, and Hund's $J^{t_{2g}}(i\omega_n)$ (dotted blue line) components of the local effective interaction and screened interaction $W_{\text{loc}}^{t_{2g}}(i\omega_n)$ for the symmetry-broken phase of SrMnO_3 in the three-band model. (b) Imaginary part of the fully screened interaction $-\text{Im}W_{\text{loc}}^{t_{2g}}(\omega)$ (thick lines) and the initial cRPA interaction $-\text{Im}U_{\text{cRPA}}^{t_{2g}}(\omega)$ (thin lines) on the real frequency axis.

such as DMFT, $T_{\text{Néel}}$ is expected to be substantially overestimated since long-range fluctuation are neglected. In the $\text{GW}+\text{EDMFT}$ approach used in this work, long-range spin fluctuation are not included, as the long-range interaction is decoupled in the charge channel. We can thus not expect a significant improvement in the description of magnetic ordering temperatures. The value of m is also reduced compared to the low-temperature value because our simulation temperature is close to $T_{\text{Néel}}$. The magnetic moment should increase as the temperature is lowered toward the experimental value. Due to the lack of adjustable parameters, this kind of temperature-dependent analysis would be an interesting topic for future studies.

Because of the heavy numerical cost of the two-sublattice calculation, we analyze the symmetry-broken phase only for the three-band model of SrMnO_3 . The insulating nature of the solution should result in small screening effects within the correlated subspace. Indeed, as shown in Fig. 7(a), the effective local interaction is essentially equal to the cRPA result and the fully screened interaction $W_{\text{loc}}^{t_{2g}}$ is only slightly smaller. The bosonic spectrum $\text{Im}W_{\text{loc}}^{t_{2g}}(\omega)$, shown in Fig. 7(b), is also similar to $\text{Im}U_{\text{cRPA}}^{t_{2g}}(\omega)$, but shifted to slightly higher energy (due to the gap) and without a prominent feature near 4 eV. The gap in the spectrum suppresses the low-frequency screening and introduces a screening channel corresponding to transitions across the gap which modify the low-energy peak in $\text{Im}W_{\text{loc}}(\omega)$. On the other hand, the local effective interaction on the real axis, shown in Fig. 8, features a broad pole centered at $\omega = 3$ eV, which is inherited from the corresponding peak in $\text{Im}U_{\text{cRPA}}^{t_{2g}}(\omega)$.

IV. CONCLUSIONS

We used the recently developed multitier $\text{GW}+\text{EDMFT}$ approach to perform a systematic analysis of the electronic properties in a family of transition metal perovskites. This self-consistent computational scheme captures both local Hubbard physics and long-range charge fluctuation, and does not rely on any choice of parameters, apart from the energy windows defining the model subspaces (tiers). Access to

FIG. 8. Imaginary part of the local effective interaction on the real frequency axis $-\text{Im}U^{t_{2g}}(\omega)$ for the three-band model of SrMnO_3 in the paramagnetic (red line) and antiferromagnetic (green line) phases. The peaks mainly originate from excitations from outside the t_{2g} subspace, which are transferred from U_{cRPA} , but also include contributions from excitations within the quasiparticle band (paramagnetic case) and excitations across the gap (antiferromagnetic case), respectively.

the frequency dependence of the self-consistently determined interactions allows to discriminate spectral features originating from local physics and from plasmonic excitations. The latter are collective charge fluctuations that screen the local effective interaction below a frequency that depends on the details of the correlated electronic structure. Both effects are self-consistently accounted for on equal footing, making $\text{GW}+\text{EDMFT}$ a fully *ab initio* approach.

The three perovskites considered, namely, SrVO_3 , SrMoO_3 , and SrMnO_3 , contain 1, 2, and 3 electrons in the t_{2g} shell, respectively, while the e_g shell is essentially empty. Due to the effects of filling and Hund coupling, the correlations in these three materials are qualitatively different. For example, the first two materials are correlated metals, while SrMnO_3 is an antiferromagnetic insulator. Reproducing these basic properties is a first important test for a parameter-free *ab initio* scheme. The comparison between the three- and five-band descriptions serves as an additional consistency check for the multitier $\text{GW}+\text{EDMFT}$ approach, which should produce consistent results for the low-energy electronic structure, independent of the choice of subspace for the self-consistency cycle. In the case of SrVO_3 we obtained a quasiparticle weight in the t_{2g} shell which is slightly larger than the $Z^{t_{2g}} \approx 0.5$ determined in photoemission studies [17] (see Table I). A precise estimate of Z from the local

TABLE I. Screened effective interaction parameters and quasiparticle weight Z in the t_{2g} subshell for the different low-energy models considered.

	SrVO_3		SrMoO_3		SrMnO_3	
	t_{2g}	$t_{2g} + e_g$	t_{2g}	$t_{2g} + e_g$	t_{2g}	$t_{2g} + e_g$
$U^{t_{2g}}(0)$	2.13	2.38	2.78	2.76	1.88	2.01
$J^{t_{2g}}(0)$	0.38	0.41	0.24	0.24	0.32	0.31
$Z^{t_{2g}}$	0.62	0.62	0.7	0.7	0.07	0.08

spectral function would be heavily influenced by the analytic continuation. We therefore considered the $\omega \rightarrow 0$ derivative of the local EDMFT contribution to the total self-energy. The corresponding renormalization is defined with respect to a Green function in which the GW self-energy is included. As a consequence, the band narrowing compared to experiment is actually underestimated. Both in the three- and five-band descriptions, the satellite features appear to have a plasmonic origin since the self-consistently computed static interaction is too small to produce Hubbard bands. Hence, within GW +EDMFT, SrVO_3 is described as a correlated metal with strong nonlocal screening effects within the t_{2g} subspace. The results for SrMoO_3 indicate an even more weakly correlated metal. In both these metallic systems, the inclusion of the e_g states has little effect on the t_{2g} states, which makes the interpretation of the satellite structures in terms of plasmonic excitations resilient against the choice of the low-energy window.

A qualitatively different situation is encountered in the case of SrMnO_3 which is experimentally found to be insulating both above [21,22] and below $T_{\text{Néel}} \approx 290$ K. If restricted to paramagnetic solutions, GW +EDMFT predicts a strongly correlated metal in proximity to a Mott transition, in both the three- and five-band models. The Hubbard bands of this strongly correlated metal are at too low energy compared to experiment. On the other hand, the extension of the method to states with broken spin symmetry produces an antiferromagnetic solution with spectral features and a magnetic moment in good agreement with experiments. There are two possible conclusions one can draw from this observation:

(i) The (short-range) antiferromagnetic spin correlations at $T \approx 300$ K may still be so strong that the material is more accurately described by the antiferromagnetic solution than by the paramagnetic solution, which ignores nonlocal correlations completely. In this case, SrMnO_3 would not be a pure Mott insulator above $T_{\text{Néel}}$, but a material strongly influenced by short-range magnetic correlations. These short-range antiferromagnetic fluctuations are, in the paramagnetic phase, described by the nonlocal vertex which is not included in the present calculations. On the other hand, below the Néel temperature, antiferromagnetic long-range order can be incorporated without the need to include the nonlocal vertex, as described in Sec. II C. Hence, the lack of a nonlocal vertex likely explains why the GW +EDMFT method only yields a correct description of SrMnO_3 in the antiferromagnetic phase.

(ii) The second possibility is that the interaction parameters which are self-consistently computed in GW +EDMFT are too small. Recent model studies have shown that cRPA can strongly overestimate the screening from bands which are relatively close to the Fermi level [50], which may lead to

an underestimation of U_{cRPA} , and hence of the effective bare interactions in tiers II and I. However, we have to note that both the five- and three-band models yield metallic solutions, and that the five-band model actually is more metallic. This indicates that the problem with U_{cRPA} , if it exists at all, is not related to screening from e_g states.

If the first scenario turns out to be correct, it implies that SrMnO_3 , similar to SrVO_3 [12], is a material whose physics has been incorrectly described by standard DFT+DMFT treatments. In these calculations, interaction parameters are chosen *ad hoc* to reproduce spectral features (e.g., Hubbard bands) based on preconceived notions about the nature of the material. The second scenario can be checked by systematically enlarging the low-energy space (tier I). As we mentioned, the inclusion of the e_g states in SrMnO_3 significantly alters the fermionic and bosonic Weiss fields. It is then conceivable that states outside our correlated subspace, but located at a similar energy separation, require a treatment beyond U_{cRPA} . In particular, it would be interesting to include the oxygen p orbitals in tier I since these may produce a significant screening. Indeed, in Ref. [47] it was shown that these states lie not far from the Fermi level in SrMnO_3 , while the authors of Ref. [51] even argue that they must be included in the low-energy models of all the perovskites.

While this study showed that the GW +EDMFT approach can handle different correlation strengths and magnetic phases in material-specific setups, and produces consistent results for different choices of low-energy models, it will be important to perform additional tests on experimentally well-characterized compounds. The great strength of GW +EDMFT is that it is free from *ad hoc* parameters and capable of treating weakly and strongly correlated systems. However, the treatment of nonlocal correlations is limited to charge fluctuations whereas spin fluctuations are not included. Furthermore, the initial downfolding in tier III relies on cRPA. Additional studies are needed to establish for which class of compounds the method has predictive power.

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