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 Sr(V,Mo,Mn)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 3-band ($t_{2g}$) description to those of a 5-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. 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). This scheme reduces the problem to a multi-orbital 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. 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, one of the most successful methods for the study of excited-state properties of weakly correlated compounds, such as semi-conductors. After more than a decade of development, the power of these two methods has been combined into a multitier GW+EDMFT formalism, which is applicable to moderately and strongly correlated materials. EDMFT is the extended version of DMFT 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. and 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 paramagnetic metals, while SrMnO$_3$ is an antiferromagnetic insulator. DFT+DMFT based modeling of these materials often only considered the $t_{2g}$ shell 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. One extension in the present work is the implementation of a self-consistency loop with two sublattices, which allows to stabilize solutions with antiferromagnetic order.
The manuscript 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 SrVO$_3$, SrMoO$_3$ and SrMnO$_3$. 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] and [13] and explain its extension to antiferromagnetically ordered systems.

A. GW$+$EDMFT

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

\[
\Sigma_{i,j}(k, \nu) = \Sigma^{\text{loc}}_{i,j}(k, \nu) + \Sigma^{\text{nonloc}}_{i,j}(k, \nu),
\]

\[
\Pi_{\alpha,\beta}(k, \nu) = \Pi^{\text{loc}}_{\alpha,\beta}(k, \nu) + \Pi^{\text{nonloc}}_{\alpha,\beta}(k, \nu).
\]

The Greek indices $\alpha, \beta$ 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

\[
G = G^0 + G^0 \Sigma G,
\]

\[
W = v + vGW,
\]

where $G^0$ is the bare propagator and $v$ the bare Coulomb interaction. The key approximation in EDMFT is that the nonlocal components of $\Sigma$ and $\Pi$ 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 $G^{\text{loc}}(\nu)$ and a dynamical bare impurity interaction $U(\nu)$. 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.

\[
\Pi^{\text{nonloc}}_{\alpha \nu, \beta \nu'}(q, \tau) = \sum_k G_{mn}(k, \tau)G_{n'm'}(k - q, -\tau) - G^{\text{loc}}_{mn}(\tau)G^{\text{loc}}_{n'm'}(-\tau),
\]

The matrix elements of the screened interaction are defined as

\[
W_{ijkl}(q, \nu) = \frac{e^{i(q-R)\tau}}{2\pi} \int dr dr' w_{iR}(r)w_{jR'}(r') \times W(r, r', \nu)w_{kR'}(r')w_{lR}(r),
\]

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 $\Sigma^{\text{imp}}, \Pi^{\text{imp}}$ and $G_k$.

2. Compute $\Sigma^{\text{loc}}$ and $\Pi^{\text{loc}}$ according to equations (1-2).

3. Define $\Sigma_k = \Sigma^{\text{imp}} + \Sigma^{\text{nonloc}}$ and $\Pi_q = \Pi^{\text{imp}} + \Pi^{\text{nonloc}}$ (GW$+$EDMFT approximations).

4. Calculate $G_k = ((G_k)^0)^{-1} - \Sigma_k)^{-1}$ and $W_q = v_q \left(1 - \Pi_q v_q\right)^{-1}$.

5. Using $G^{\text{loc}} = \frac{1}{N} \sum_k G_k$ and $W^{\text{loc}} = \frac{1}{N} \sum_q W_q$ calculate the fermionic Weiss field

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

and the effective impurity interaction

\[
\mathcal{U} = W^{\text{loc}} \left(1 + \Pi^{\text{imp}} W^{\text{loc}}\right)^{-1}.
\]

6. Numerically solve the impurity problem to obtain $G^{\text{imp}}$ 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 $\Sigma^{\text{imp}} = \mathcal{G}^{-1} - (G^{\text{imp}})^{-1}$, $\Pi^{\text{imp}} = \chi^{\text{imp}} (\mathcal{U} \chi^{\text{imp}} - 1)^{-1}$ and $W^{\text{imp}} = \mathcal{U} - \mathcal{U} \chi^{\text{imp}} \mathcal{U}$.

8. If the selfconsistency 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 $\Psi$ and is hence conserving. However, in practice this is not feasible. To overcome this problem a multitier implementation was
developed in Refs. [12] and [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 mult-tier 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[13]. Based on the DFT bandstructure we compute the one-shot $GW$ self-energy $\Sigma^G W^0$ using the SPEx code[14][15].

Then, an intermediate- or low-energy subspace, $I$, which includes up to 10 bands around the Fermi energy is defined using maximally localized Wannier functions as implemented in the Wannier90 library.[21][22] The effective Coulomb interaction, $U$, on the intermediate subspace is computed within the constrained random-phase approximation (cRPA)[23] using the SPEx code. The $G^0W^0$ self-energy contribution from within the intermediate subspace is removed from $\Sigma^G W^0$ to define an effective bare propagator $G^I_k$ 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[20][25] quantum Monte-Carlo algorithm implemented in ALPS[20][21], while the self-consistency equations make use of the TRIQS framework.[22] 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:

\[
G_{-1}^I I, q \rightarrow \sum_{I, q}^{-1} = -i \int \frac{d^3 q}{(2\pi)^3} 
\]

\[
W_{-1}^{-1} = v_{q}^{-1} - \left( \Pi_{q}^{G0} - \Pi_{q}^{G0} \right) \frac{-\Lambda_{q}}{q} - \left( \Pi_{q}^{GG} - \Pi_{q}^{GG} \right) \frac{-\Pi_{C, loc}^{EDMFT}}{C, loc}.
\]

(11)

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

### 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[23] by considering two sublattices $A$ and $B$ and imposing the following relation between the self-energies:

\[
\Sigma^A = \Sigma^B, \quad \Sigma^A = \Sigma^B.
\]

(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 multtier 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

\[
\Sigma^{EDMFT} \big|_{C, loc} \rightarrow \Sigma^{EDMFT}_{\uparrow \downarrow} \big|_{C, loc}.
\]

(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 $\Pi^{EDMFT} \big|_{C, loc}$ 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
term 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 $t_{2g}$ sub-shell. The calculations for SrVO$_3$ and SrMoO$_3$ are performed at inverse temperature $\beta = 10 \text{ eV}^{-1}$, corresponding to $T = 1160$ K, while in the case of SrMnO$_3$ we use $\beta = 40 \text{ eV}^{-1}$ corresponding to $T = 290$ K. The latter value is close to $T_{\text{Néel}} \approx 260$ K for SrMnO$_3$ whose magnetic moments order antiferromagnetically in all directions (G-type ordering).

Figure 1. Local and k-resolved spectral function of SrVO$_3$ (left) and SrMoO$_3$ (right) in the three- (top) and five- (bottom) band description. Thin black lines represent the LDA bandstructure.

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. The conduction band is formed by vanadium 3$d$ states of $t_{2g}$ character which are populated by one electron per unit cell. Within LDA the conduction band is isolated with a bandwidth of roughly 2.2 eV. The 3.8 eV wide conduction band in SrMoO$_3$ 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 photo-emission (PES) and inverse photo-emission (IPES) spectra of SrVO$_3$ display a renormalized quasi-particle 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. SrMoO$_3$, on the other hand, exhibits a very weakly renormalized quasi-particle peak and a pronounced shoulder structure in the PES. The satellite features in the local spectral function of both systems clearly indicate correlation effects beyond the LDA. For SrMoO$_3$ it was shown in Ref. 11 that the satellite features cannot be described as Hubbard bands. A later publication, using the same GW+EDMFT multitier formalism as employed in the current paper, showed that the satellites in this compound should instead be interpreted as plasmon satellites originating from long-range charge fluctuations. This is consistent with the conclusions of Ref. 11 and hence there is a relative consensus in the literature on the nature of the satellite features in the spectral function of SrMoO$_3$. This is not the case for SrVO$_3$, where the origin of the satellite features is still under debate. For this compound the 3$d$ valence states (and hence also the 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, as well as shot combinations of $GW$ and DMFT, 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 SrVO$_3$ 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 SrVO$_3$ are well described by the cumulant expansion which is an expansion of the Green’s function that is based on the $GW$-approximation of the self-energy. 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 and 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 $U(0)$, 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 k-resolved spectral functions for SrVO$_3$ and SrMoO$_3$ obtained from the three-band 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 local spectral function follow the dispersion of the unoccupied part of the quasi-particle bands is consistent with the plasmon scenario. We find that the crystal field splitting between the two manifolds is significantly enhanced by correlation effects in the case of SrMoO$_3$ while for SrVO$_3$ the $e_g$ states remain at the same position as in the LDA bandstructure.

In Fig. 2 we show the frequency dependent interaction $\mathcal{U}_{\text{cRPA}}(i\omega_n)$, Hubbard $\mathcal{U}^{t_{2g}}(i\omega_n)$ and Hund’s $\mathcal{J}^{t_{2g}}(i\omega_n)$ component (dashed) of the local effective interaction and screened interactions $W_{\text{loc}}^{t_{2g}}(i\omega_n)$ for SrVO$_3$ (top) and SrMoO$_3$ (bottom) in the three- (a,c) and five- (b,d) band description.
ing 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 $W_{loc}^{t_{2g}}(0)$.

The similarity between the three- and five-band results is also seen in the broad pole structure in $-\Im W_{loc}(\omega)$ (Fig. 3) which provides a consistent explanation of the satellites in the 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 SrVO$_3$ indicating stronger screening effects compared to SmO$_3$.

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. SrMnO$_3$**

1. Results for paramagnetic SrMnO$_3$

There is a substantial level of agreement on the importance of electronic correlations in SrMnO$_3$, while their role in determining the experimentally observed insulating ground state is still debated. Previous studies on this material [15,37] employed DFT for the structural properties or DFT+DMFT to incorporate the effect of Hubbard-like 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 SrMnO$_3$ which, at first glance, appear quite similar to the models described above. In SrMnO$_3$, 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 nonnegligible screening channels. Indeed, the static values of the cRPA interaction reported in Fig. 4 are quite different for the two models, namely $U_{cRPA}(0) = 1.9$ eV in the three-band model and $U_{cRPA}(0) = 2.7$ 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 $W_{loc}^{t_{2g}}$ 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 $3W_{loc}$ along the real frequency axis. $3W_{loc}^{t_{2g}}$ 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 panel (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. 3 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 SrMnO$_3$.

In Fig. 5 we show the local and $\mathbf{k}$-resolved spectral
functions of SrMnO$_3$ 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 SrVO$_3$ and SrMoO$_3$ (see Ref. [13] with a smaller bandwidth and a plasmonic broadening occurring mainly in the proximity of the $\Gamma$ 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 $\pm 1$ 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 quasi-particle band) it is possible to recognize renormalized and/or broadened replicas of the noninteracting dispersion. This behavior is typical of the Mott transition scenario. The asymmetry between the occupied and unoccupied parts of the spectra appears to be a consequence of the $GW$-derived $k$-dependent self-energy, which is known to produce such effects. 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 multi-orbital system with $J_H > 0$ is lowest at half-filling.

The low-energy structures in $\Im W_{\text{loc}}(\omega)$ 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 $\Im U(\omega)$ at $\omega$ corresponding to the characteristic energy for charge excitations across this gap. In the presence of a quasi-particle band, there are also screening modes associated with transitions between the quasi-particle 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 $\Im W(\omega)$ 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, both
our models of SrMnO$_3$ remain conducting in the paramagnetic phase. The metallicity is due to a very narrow quasi-particle band pinned at the chemical potential. The quasi-particle peak in the five-band case is a bit larger, see Fig. 4(c), and as a result, the screened interaction $W_{t_{2g}}^{\text{loc}}$ is smaller (panel (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 quasi-particle peak in the five-band model. 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$_3$. Measurements of the magnetic moment report a value of 2.6±0.2 $\mu_B$ and previous DFT+DMFT calculations yield compatible results. The low temperature behavior has been reported to be well described by the ordering of $S = 3/2$ local moments with a $T_{\text{Néel}}$ between 233 K and 260 K. The variations in the ordering temperature can be accounted for by oxygen defects are

To describe antiferromagnetic ordering we extended the GW+EDMFT multitier approach to a bipartite lattice as described in Sec. II C. At $\beta = 40$ eV$^{-1}$, the solution with G-type antiferromagnetism self-consistently emerges in our parameter-free simulation. The local and k-resolved spectral functions of the three-band model are reported in Fig. 7 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 Ref. 33 and 36, 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

$$m = \sum_\alpha (n_\alpha \uparrow - n_\alpha \downarrow) \mu_B,$$

where $\alpha$ 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. 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 such as DMFT, $T_{\text{Néel}}$ is expected to be substantially overestimated since long-range fluctuation are neglected. In the GW+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
Figure 8. (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 \( W^{t_2g}_{\text{loc}}(i\omega_n) \) for the symmetry broken phase of SrMnO\(_3\) in the three-band model. (b) Imaginary part of the fully screened interaction \( -\Im W^{t_2g}_{\text{loc}}(\omega) \) (thick lines) and the initial cRPA interaction \( -\Im U_{\text{cRPA}}^{t_2g}(\omega) \) (thin lines) on the real frequency axis.

The insulating nature of the solution should result in small screening effects within the correlated subspace. Indeed, as shown in Fig. 8(a), the effective local interaction is essentially equal to the cRPA result and the fully screened interaction \( W^{t_2g}_{\text{loc}} \) is only slightly smaller. The bosonic spectrum \( \Im W^{t_2g}_{\text{loc}}(\omega) \), shown in panel (b), is also similar to \( \Im U_{\text{cRPA}}^{t_2g}(\omega) \), 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 \( \Im W^{t_2g}_{\text{loc}}(\omega) \). On the other hand, the local effective interaction on the real axis, shown in Fig. 8(b) features a broad pole centered at \( \omega = 3 \) eV, which is inherited from the corresponding peak in \( \Im U_{\text{cRPA}}^{t_2g}(\omega) \).

IV. CONCLUSIONS

We used the recently developed multiband GW+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 the frequency dependence of the selfconsistently 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 GW+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-band and five-band description serves as an additional consistency check for the multiband GW+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 quasi-particle weight in the \( t_{2g} \) shell which is slightly larger than the \( Z^{t_{2g}} \approx 0.5 \) determined in photoemission studies. See Table I. Both in the three-band and five-band description, 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 \( T_{\text{Néel}} \approx 290 \) K and below \( T_{\text{Néel}} \approx 300 \) 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 Section IIC. Hence the lack of a nonlocal vertex is a likely reason for why the GW as described in Section IIC. Hence the lack of a 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 Section IIC. Hence the lack of a nonlocal vertex is a likely reason for 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, which may lead to an underestimation of $U_{\text{cRPA}}$, 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 $U_{\text{cRPA}}$, 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$_3$, 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) based on pre-conceived notions about the nature of 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 $U_{\text{cRPA}}$. 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 the current study showed that the GW+EDMFT approach can handle different correlations 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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