Frequency-dependent local interactions and low-energy effective models from electronic structure calculations

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We propose a systematic procedure for constructing effective models of strongly correlated materials. The parameters, in particular the on-site screened Coulomb interaction U, are calculated from first principles, using the random-phase approximation. We derive an expression for the frequency-dependent $U(\omega)$ and show, for the case of nickel, that its high-frequency part has significant influence on the spectral functions. We propose a scheme for taking into account the energy dependence of $U(\omega)$, so that a model with an energy-independent local interaction can still be used for low-energy properties.

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I. INTRODUCTION

Lattice fermion models such as the Hubbard model or the Anderson impurity model and their extensions have played a major role in studying electron correlations in systems with strong on-site correlations. Despite the widespread use of these models, little justification has been given in using them. The models are postulated on the basis of physical intuition. In particular, the models employ parameters, such as the famous Hubbard interaction U, which are normally adjusted to serve the given problem. Without judicious choice of parameters, the model may yield misleading results, or in the worst case, the model itself is not sufficient to describe the real system. One can define rigorously these concepts in the path-integral formulation of the many-body problem by performing a partial trace over the degrees of freedom that one wants to eliminate, and ignoring the retardation in the interactions generated by this procedure. However, this elimination of the degrees of freedom is very hard to perform for real materials. It is therefore very desirable to figure out a systematic way of constructing low-energy effective models with well-defined parameters calculated from first principles such that the model can quantitatively reproduce and predict physical properties of interest of the corresponding real system, especially when the correlation effects are crucial.

Another important issue that has not received sufficient attention is the role of energy dependence of the screened local Coulomb interaction U. Model studies investigating the importance of high-energy states in the Hubbard model can be found in Refs. 1–3. A dynamic Hubbard model has also been considered.[4] In most cases, however, U is assumed to be static, but on the other hand we know that at high energy the screening becomes weaker and eventually the interaction approaches the large bare Coulomb value, which is an order of magnitude larger than the static screened value. Of course, the high-energy part of the Coulomb interaction has in some way been downfolded into the Hubbard U but it is not clear how this downfolding is actually accomplished.

A number of authors have addressed the problem of determining the Hubbard U from first principles. One of the earliest works is the constrained local-density approximation (LDA) approach,[1,5] where the Hubbard U is calculated from the total-energy variation with respect to the occupation number of the localized orbitals. An approach based on the random-phase approximation (RPA) was later introduced,[6] which allows for the calculations of the matrix elements of the Hubbard U and its energy dependence. This was followed by a more refined approach for calculating U.[7] A yet different approach computes the matrix elements of the Coulomb interactions screened in real space and assumes a Yukawa form to extract the Hubbard U and the other interactions which determine the multiplet splittings.[8]

The purpose of the present work is to develop a precise formulation for a systematic construction of effective models where the parameters are obtained from realistic first-principles electronic structure calculations. In particular, we concentrate on the calculation of the Hubbard U and demonstrate the importance of its energy dependence. We show that a static Hubbard Hamiltonian, obtained from a construction in which this energy dependence is simply neglected, fails even at low energy. This static model can be appropriately modified, however, by taking into account the feedback of the high-energy part of U into the one-particle propagator. We illustrate our scheme in transition metals, concentrating on Ni as an example, since it is a prototype system consisting of a narrow 3d band embedded in a wide band. Furthermore, Ni is one of the most problematic case from the viewpoint of the LDA.

In summary, in Sec. II we propose a method for calculating a frequency-dependent Hubbard $U(\omega)$ and derive an effective model for electrons in a narrow band using $U(\omega)$ as

an effective interaction between the electrons. In Sec. III we investigate the influence of the frequency-dependent $U(\omega)$ on the self-energy and the spectral function within the *GW* approximation (GWA).[9,10] We then suggest a way of modifying the static Hubbard model so that it can still give reliable description of low-energy physics. We also propose a scheme of taking into account the frequency-dependent $U(\omega)$ in an approximate way.

II. THEORY

Let us suppose that the band structure of a given solid can be separated into a narrow band near the Fermi level and the rest, like, for example, in transition metals or $4f$ metals. Our aim is to construct an effective model which only includes the narrow $3d$ or $4f$ band. The effective interaction between the $3d$ electrons in the Hubbard model can be formally constructed as follows. We first divide the complete Hilbert space into the Hubbard space $\{\psi_d\}$, consisting of the $3d$ states or the localized states, and the rest. The bare Green's function G_d, spanning the d subspace, is given by

$$G_d(\mathbf{r},\mathbf{r}';\omega)=\sum_d^{occ}\frac{\psi_d(\mathbf{r})\psi_d^*(\mathbf{r}')}{\omega-\varepsilon_d-i0^+}+\sum_d^{unocc}\frac{\psi_d(\mathbf{r})\psi_d^*(\mathbf{r}')}{\omega-\varepsilon_d+i0^+}. \quad (1)$$

Let P be the total (bare) polarization, including the transitions between all bands,

$$P(\mathbf{r},\mathbf{r}';\omega)=\sum_i^{occ}\sum_j^{unocc}\psi_i(\mathbf{r})\psi_i^*(\mathbf{r}')\psi_j^*(\mathbf{r})\psi_j(\mathbf{r}')\times\left\{\frac{1}{\omega-\varepsilon_j+\varepsilon_i+i0^+}-\frac{1}{\omega+\varepsilon_j-\varepsilon_i-i0^+}\right\}. \quad (2)$$

P can be divided into $P=P_d+P_r$, in which P_d includes only $3d$ to $3d$ transitions (i.e, limiting the summations in Eq. (2) to $i,\ j\in\{\psi_d\}$), and P_r be the rest of the polarization. The screened interaction W on the RPA level is given by

$$\begin{aligned}W&=[1-vP]^{-1}v=[1-vP_r-vP_d]^{-1}v\\&=[(1-vP_r)\{1-(1-vP_r)^{-1}vP_d\}]^{-1}v\\&=\{1-(1-vP_r)^{-1}vP_d\}^{-1}(1-vP_r)^{-1}v\\&=[1-W_rP_d]^{-1}W_r\end{aligned} \quad (3)$$

where we have defined a screened interaction W_r that does not include the polarization from the $3d$-$3d$ transitions:

$$W_r(\omega)=[1-vP_r(\omega)]^{-1}v \quad (4)$$

(we have not explicitly indicated spatial coordinates in this equation). The *identity* in Eq. (3) explicitly shows that the interaction between the $3d$ electrons is given by a frequency-dependent interaction W_r. It fits well with the usual physical argument that the remaining screening channels in the Hubbard model associated with the $3d$ electrons, represented by the $3d$-$3d$ polarization P_d, further screen W_r to give the fully screened interaction W.

We now choose a basis of Wannier functions $\{\phi_{Rn}\}$, centered about atomic positions R, corresponding to the $3d$ Bloch functions $\{\psi_{kn}\}$, and consider the matrix elements of the (partially screened) frequency-dependent Coulomb interaction W_r:

$$U_{R_1nR_2n',R_3mR_4m'}(\tau-\tau')\doteq\int d^3rd^3r'\,\phi_{R_1n}^*(\mathbf{r})\phi_{R_2n'}(\mathbf{r})\times W_r(\mathbf{r},\mathbf{r}';\tau-\tau')\phi_{R_3m}^*(\mathbf{r}')\phi_{R_4m'}(\mathbf{r}'). \quad (5)$$

We would like to obtain an effective model for the $3d$ degrees of freedom. Because of the frequency dependence of the U's (corresponding to a retarded interaction), this effective theory will not take a Hamiltonian form. We can however, write such a representation in the functional-integral formalism[11] by considering the effective action for the $3d$ degrees of freedom given by

$$S=\int d\tau d\tau'\Big[-\sum d_{Rn}^\dagger(\tau)G^{-1}_{Rn,R'n'}(\tau-\tau')d_{Rn'}(\tau')+\frac{1}{2}\sum :d_{R_1n}^\dagger(\tau)d_{R_2n'}(\tau):\times U_{R_1nR_2n',R_3mR_4m'}(\tau-\tau'):d_{R_3m}^\dagger(\tau')d_{R_4m'}(\tau'):\Big], \quad (6)$$

where: $d^\dagger d$: denotes normal ordering, which accounts for the Hartree term, and the summation is over repeated indices. When using a Wannier transformation which does not mix the d subspace with other bands, the Green's function can be taken, to first approximation, to be the bare Green's function G^0_{dd} constructed from the Bloch eigenvalues and eigenfunctions. If, instead, an LMTO formalism[12] is used, one should in principle perform a downfolding procedure onto the d subspace, i.e, perform a partial trace over s, p degrees of freedom.

In the following, we retain only the local components of the effective interaction on the same atomic site. This is expected to be a reasonable approximation because the $3d$ states are rather localized. The formalism may be easily extended to include intersite Coulomb interactions if necessary. Hence, we consider the frequency-dependent Hubbard interactions:

$$U_{nn',mm'}(\tau-\tau')\doteq\int d^3rd^3r'\,\phi_n^*(\mathbf{r})\phi_{n'}(\mathbf{r})W_r(\mathbf{r},\mathbf{r}';\tau-\tau')\times\phi_m^*(\mathbf{r}')\phi_{m'}(\mathbf{r}'), \quad (7)$$

with ϕ_n being the Wannier orbital for $R=0$. In order to illustrate the procedure within the linear muffin-tin orbital (LMTO) basis set, we use instead of the Wannier orbital the normalized function head of the LMTO ϕ_H which is a solution to the radial Shrödinger equation matching to a Hankel function at zero energy at the atomic sphere boundary. The Wannier function of the d band is a substantial mixture of the original LMTO atomic d orbital with other s and p orbitals.

In this paper, we investigate the importance of the energy dependence of U. Therefore, we shall compare the results obtained from Eq. (6) with those of a Hamiltonian approach

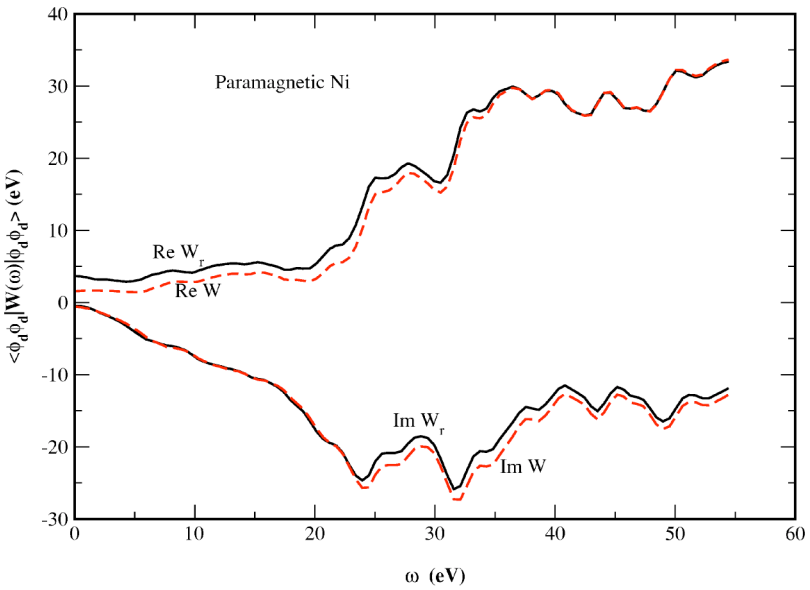

FIG. 1. The real and imaginary parts of the diagonal matrix elements of the screened interaction of paramagnetic nickel in the d orbital (e_g). The dashed curves correspond to the fully screened interaction and the solid curves to the screened interaction without the d to d transitions. The results for the t_{2g} orbitals are almost identical.

in which one would construct a Hubbard model with a *static* interaction U:

$$H = \sum_{Rn,R'n'} c^{\dagger}_{Rn} h_{Rn,R'n'} c_{R'n'} + \frac{1}{2} \sum_{R,nn',mm'} c^{\dagger}_{Rn} c_{Rn'} U_{nn',mm'} c^{\dagger}_{Rm} c_{Rm'}. \quad (8)$$

It seems natural to identify the static Hubbard U with the (partially) screened local interaction in the low-frequency limit $W_r(\omega=0)$. Note that the Hubbard model (8) has been constructed in the simplest manner, by simply taking the quadratic part to be the d block of the noninteracting Hamiltonian.

In order to compare the results obtained from the full dynamical U to those of the static Hubbard model, we need to solve Eqs. (6) and (8) within some consistent approximation scheme. In the following, the exact self-energy for the solid is assumed to be given by the GWA. Since we are considering a system with moderate correlation strength (U/bandwidth$\approx$unity) the GW approximation may still be a reasonable tool to use in solving the effective models (6) and (8). This allows us to make a proper comparison between the "exact" self-energy and the Hubbard model self-energy. If the assumption of static U is valid, the Hubbard self-energy and the true self-energy (both within the GWA) should be close to each other, at least for small energies. Or equivalently, the spectral function for small energies should resemble that of the full one.

III. RESULTS AND DISCUSSIONS

A. Comparing self-energies

The screened interaction with and without the 3d-3d transitions is shown in Fig. 1 in the case of paramagnetic nickel. Here and in all the following, a spin-unpolarized (paramagnetic) solution is considered. At low energies, the real part of the (partially) screened interaction W_r without the 3d-3d transitions is larger than the full one W, and at high energies they approach each other, as anticipated. Related calculations have also been performed by Kotani.[7]

We first compare the self-energy obtained from a GW treatment of the full system, given by

$$\Sigma(r,r';\omega) = \frac{i}{2\pi} \int d\omega' e^{i\eta\omega'} G(r,r';\omega+\omega') W(r,r';\omega') \quad (9)$$

to the self-energy obtained from the effective model (6) with an energy-dependent interaction $U(\omega)=W_r(\omega)$. Because of Eq. (3), the screened interaction corresponding to this $U(\omega)$ is simply W, and the corresponding self-energy reads

$$\Sigma_d(\omega) = \frac{i}{2\pi} \int d\omega' e^{i\eta\omega'} G_d(\omega+\omega') W(\omega'). \quad (10)$$

The difference between this expression and the GWA for the full system [Eq. (9)] is that in Eq. (10) only the d block of the Green's function has been included (since the effective action was written for the d-band only). Hence, the two self-energies differ by a term $G_r W$, with $G_r = G - G_d$. We expect that the wave-function overlap between two 3d states (one from G_d and the other from the 3d state appearing in the matrix element of Σ_d) and other non-3d states is small so that Σ_d should be close to the true Σ. In Fig. 2, the two self-energies are displayed (more precisely, in this figure and in all the following, we display the matrix element of the self-energy in the lowest 3d state (band number 2), at the Γ-point, corresponding to an LDA eigenenergy −1.79 eV). We observe that the two self-energies are rather close to each other. Hence, we conclude that the effective Hubbard model (6) for the d subspace, with an energy-dependent interaction, provides a reliable description of the real system.

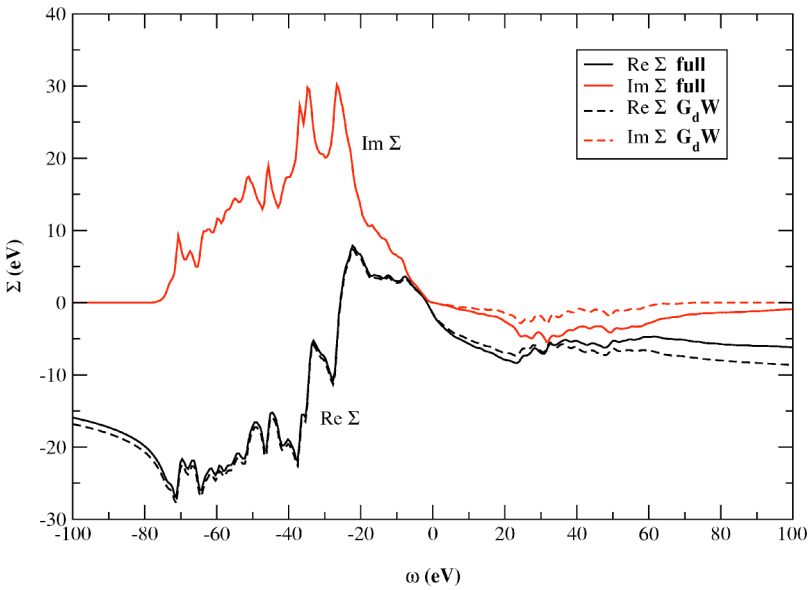

FIG. 2. The matrix elements of the self-energy of paramagnetic nickel in the lowest *d* state corresponding to the LDA eigenvalue of −1.79 eV at the Γ point. The solid curves correspond to the real system (i.e., full *GW* calculation) and the dashed curves to the Hubbard model with a frequency-dependent *U*.

We now turn to the self-energy associated with the static Hubbard model (8) with $U=W_r(0)\simeq 3.5$ eV. The local screened interaction for this model, within the GW approximation, is given by

$$W_d(\omega) = [1 - UP_d(\omega)]^{-1}U, \tag{11}$$

and the self-energy of the Hubbard model in the GWA thus reads

$$\Sigma_d^H(r,r';\omega) = \frac{i}{2\pi}\int d\omega' e^{i\eta\omega'} G_d(r,r';\omega+\omega')W_d(r,r';\omega'). \tag{12}$$

Note that the difference between this static Hubbard model self-energy and that of the effective model with a frequency-dependent interaction (6) relies in the use of a different form of the screened interaction W_d instead of the full *W*.

In Fig. 3 the imaginary part of the self-energy is shown. Here we see that $\mathrm{Im}\,\Sigma_d^H$ for the Hubbard model is peaked around 5 eV since there are no states above or below the 3*d* band. On the other hand, Im Σ of the real system is peaked at around the average plasmon energy around 30 eV (Fig. 2). But within an energy region spanning about twice the 3*d* bandwidth, the imaginary part of the Hubbard model self-energy is in reasonable agreement with the full one.

In Fig. 4, the real part of this self-energy for nickel is shown and compared with that of the full GWA self-energy. In contrast to Im Σ, the real part of the self-energies obtained with a static and frequency-dependent *U* are quite different. This is understandable from Fig. 3 since Re Σ is simply the Hilbert transform of Im Σ. Since the energy scale of the self-energy of the real system is determined by the bare Coulomb interaction *v* whereas the Hubbard self-energy is set by *U*, the latter has been shifted so that it is equal to the former at the LDA eigenvalue (−1.79 eV) of the band we have consid-

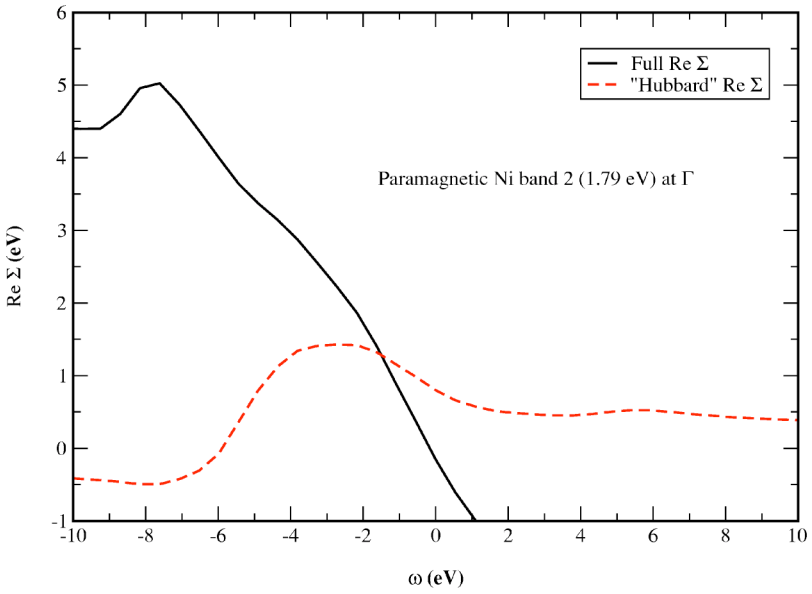

FIG. 3. The imaginary parts of the self-energy of the real systems (solid) and the Hubbard model (dash) in the GWA.

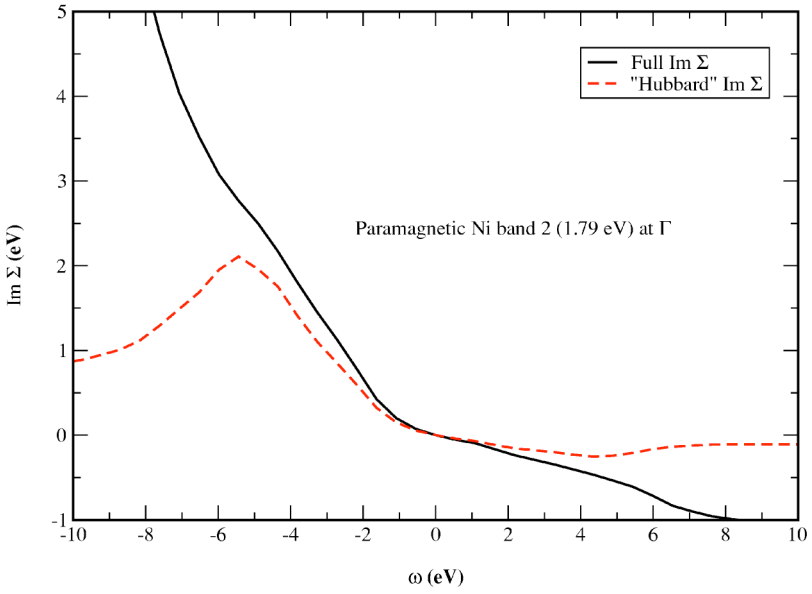

FIG. 4. The real parts of the self-energy of the real system (solid) and the Hubbard model (dash) in the GWA.

ered at the Γ point. The difference in magnitude of the self-energies is not important since it simply shifts the spectrum (or, said differently, we have compared differences $\Sigma-\mu$ from the values of the chemical potential obtained in the various schemes). However, the difference in the variation of the self-energy with respect to energy matters, since it will give a different quasiparticle weight $Z=[1-\partial \mathrm{Re}\Sigma/\partial\omega]^{-1}$ and affect the spectral function. As can be deduced from the figure, the Z factor of the Hubbard model taken at the energy of the quasiparticle band at the Γ point ($\simeq -1.79$ eV) is much closer to unity as compared to the true (full GW) one because the former already contains the renormalization from the plasmon. Hence, neglecting frequency dependence directly affects the physical results, *even in the low-energy range*. We shall see below, however, that it is possible to modify the static model in such a way that an accurate approximation is obtained at low energy.

These findings can be understood in more detail by considering explicit expressions of the self-energy obtained within the GW approximation, for an effective model of the d subspace defined on the periodic lattice. The imaginary part reads

$$\mathrm{Im}\,\Sigma_n(k,\omega)=\sum_{q,m}\mathrm{Im}\,W_{nm}(q,\omega-\epsilon^m_{k-q})[n_F(\epsilon^m_{k-q}) + n_B(\omega-\epsilon^m_{k-q})]. \tag{13}$$

In this expression, n,m are band indices, ϵ^n_k corresponds to the nth noninteracting band, and n_F (resp. n_B) is the Fermi (resp. Bose) function. Im W is the spectral function associated with the effective interaction (to be taken as W if the self-energy of the frequency-dependent effective model is considered, and as W_d if that of the static Hubbard model is considered). From this expression, one sees that if one considers an energy ω, the bands contained in the energy interval $[0,|\omega|]$ are the only ones contributing significantly to Im Σ. This is the reason why the *imaginary part* of the self-energy at low energy will be correctly reproduced by the effective low-energy model (provided, of course, that the spectral function Im W is correctly approximated at low energy). In contrast, the real part of the correlation self-energy is obtained from the Hilbert transform or the Kramers-Krönig relation in the form

$$\mathrm{Re}\,\Sigma_n(k,\omega)=-\frac{1}{\pi}P\int_{-\infty}^{+\infty}d\nu\sum_{q,m}\mathrm{Im}\,W_{nm}(q,\nu)\frac{n_F(\epsilon^m_{k-q})+n_B(\nu)}{\omega-\epsilon^m_{k-q}-\nu}. \tag{14}$$

Because the principal-part integral extends over the whole frequency range, high-frequency contributions influence the self-energy even at low frequency. As a result, for the case we are considering, an accurate description of the real part of the self-energy cannot be obtained within the static Hubbard model because the effective interaction is not correctly approximated over the whole frequency range. This formula also suggests a way to appropriately modify the effective static model in order to obtain an accurate description at low energy, as discussed below.

B. The puzzle of the satellite

Figure 5 compares the spectral function obtained for the full system within the GWA, and that of the static Hubbard model (8). A striking difference between these two results is the absence of the 6 eV satellite in the GWA for the full system, while the static Hubbard model displays a satellite feature. This is to be expected from the structure of the self-energy. The GWA self-energy of the full system continues to grow and reaches a maximum around the plasmon excitation at around 25–30 eV, while the self-energy of the static Hubbard model has a maximum around the width of the 3d band (4 eV). In comparison with the true plasmon, the "plasmon" of the Hubbard model has a much lower energy, of the order of U, which results in the "6 eV satellite."

In contrast to the electron gas, where the plasmon excitation contained in Im W is quite distinct in energy and well

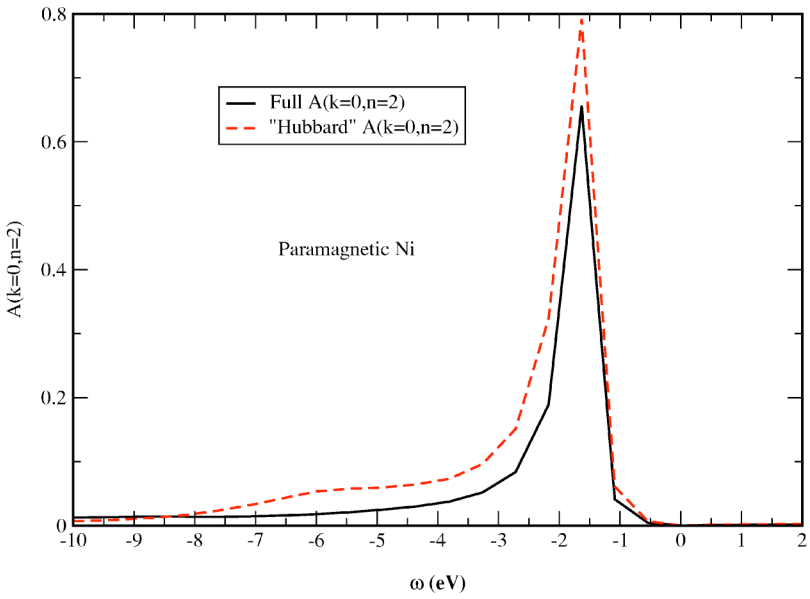

FIG. 5. The spectral functions of the real system (solid) and the Hubbard model (dash) in the GWA.

separated from the valence band, there is no well-defined plasmon excitation in transition metals. As can be seen in Fig. 1, in the case of nickel or transition metals Im W has a rather broad distribution of weight extending down to the low-energy region. It is this weight at low energy that gives a significant contribution to Im Σ that in turns washes out the satellite structure presence in the static Hubbard model. If Im W is dominated by a sharp plasmon well separated from the valence band, then it follows from Eq. (13) that the corresponding Im Σ has little weight in the region of the valence band. Consequently, we expect the static Hubbard model to give a reliable spectrum at low energy. However, when Im W has a significant weight at low energy, we expect this weight will contribute to Im Σ, and consequently influence the spectral function. The preceding argument has been based on the GWA but it seems quite feasible that the conclusion drawn from this argument is also true in the exact case. We know, for instance, that the experimental loss spectrum of nickel, which is proportional to Im W, is well reproduced by the RPA.

We may divide the frequency region into three parts: a low-energy region $\omega \lesssim \Lambda$, where Λ is of the order of the d bandwidth, an intermediate region $\Lambda < \omega < \omega_p$, where ω_p is the average plasmon energy, and a high-energy region $\omega \lesssim \omega_p$. By a judicious choice of a noninteracting G it should still be possible to construct a Hubbard model with a static U that gives a reliable description of the spectrum or quasiparticle energies in the low-energy region $\omega \lesssim \Lambda$, as proposed in the following section. For the intermediate and high-energy regions, however, the static Hubbard model may have a problem since Im Σ plays an important role in determining the structure of the spectrum like the shake-up satellite or the lower Hubbard band.

One could blame the appearance of a satellite in our static Hubbard model calculation on the fact that we have used a (non-self-consistent) GW approximation. However, more accurate treatments of the static Hubbard model [such as dynamical mean-field theory (DMFT)] do preserve this feature, which has in this context a natural interpretation as a lower Hubbard band. On the other hand, it could be that the experimentally observed nickel satellite has a somewhat different physical origin and that the satellite obtained in static Hubbard model calculations is spurious in the context of nickel (e.g., because it is not legitimate to use a low-energy effective model in this energy range). In our view, this issue is an open problem which deserves further work. Although the calculations have been carried out within the GWA, our finding indicates a possible problem with a static Hubbard model even when treated exactly.

C. Improving the effective static model

The preceding discussion shows that an effective model for the d band can be constructed, which accurately reproduces the full results over an extended energy range, provided the energy dependence of $U(\omega)$ is retained. However, performing calculations with an energy-dependent Hubbard interaction is exceedingly difficult. A more modest goal is to obtain an effective model which would apply to some low-energy range only, $|\omega| < \Lambda$, with Λ a cutoff of the order of the d-bandwidth. In order to achieve this goal, we propose to adopt a renormalization-group point of view, in which high energies are integrated out in a systematic way. Following this procedure, an appropriate low-energy model with a static U can be appropriately constructed. As we shall see, the bare Green's function defining this low-energy model does not coincide with the noninteracting Green's function in the d-subspace (we have seen that this does not lead to a satisfactory description, even at low energy).

Let us illustrate this idea within the GW approximation. The full Hilbert space is divided into the Hubbard space, comprising the $3d$ orbitals, and the downfolded space, comprising the rest of the Hilbert space. This approach is complementary to the one in Refs. 2 and 13, where the division is done in real space (on site and off sites). Within the GWA we may write the full self-energy as follows:

$$GW = G_d W_d + G_d(W - W_d) + G_r W, \tag{15}$$

where $G = G_d + G_r$. The term $G_d(W - W_d)$ represents the high-energy contribution of the screened interaction. This is the

main source of error in the static limit, as discussed in the previous sections. The term G_rW is *not* obtainable within the Hubbard model, even when a frequency-dependent U is employed since G_r resides in the downfolded space. This term was shown to be small. Its effect at low-energy can also be taken into account by appropriate modifications of the one-particle propagator, but we shall neglect it for simplicity.

We consider the following Hubbard model with a static $U=W_r(0)$, but a modified one-particle propagator $\tilde{G}$, defined by the action

$$S_H = -\int d\tau d\tau' \sum d^\dagger_{Rn}(\tau)\tilde{G}^{-1}_{Rn,R'n'}(\tau-\tau')d_{Rn'}(\tau') + \frac{1}{2}\int d\tau \sum :d^\dagger_{R_1n}(\tau)d_{R_2n'}(\tau):U_{R_1nR_2n',R_3mR_4m'} :d^\dagger_{R_3m}(\tau)d_{R_4m'}(\tau):. \quad (16)$$

The self-energy of this static Hubbard model in the GWA is

$$\tilde{\Sigma}^H_d = \tilde{G}_d\tilde{W}_d. \quad (17)$$

where the new effective interaction is $\tilde{W}_d=U[1-U\tilde{P}_d]^{-1}$, with $\tilde{P}_d$ constructed from the new Green's functions $\tilde{G}_d$ (schematically $\tilde{P}_d=\tilde{G}_d\cdot\tilde{G}_d$). We request that the interacting Green's function of this modified static model coincides with that of the Green's function calculated with the frequency-dependent interaction *in the low-energy range* $|\omega|<\Lambda$:), that is

$$\tilde{G}^{-1}_d - \tilde{\Sigma}^H_d \simeq G^{-1}_d - G_dW_d - G_d(W-W_d) \text{for} |\omega| < \Lambda. \quad (18)$$

Using the identity $G^{-1}-GW=[1-UP]^{-1}G^{-1}$, this can be rewritten as

$$[1-U\tilde{P}_d]^{-1}\tilde{G}^{-1}_d \simeq [1-UP_d]^{-1}G^{-1}_d - G_d(W-W_d) \text{ for} |\omega| < \Lambda. \quad (19)$$

This is an integral equation which determines, in principle, the modified bare Green's function $\tilde{G}_d$ to be used in the "downfolded" static action (16). To first approximation, one can neglect the polarization terms in this equation, and obtain the first-order modification of $\tilde{G}_d$ as

$$\tilde{G}^{-1}_d = G^{-1}_d - G_d(W-W_d) + \cdots. \quad (20)$$

The first correction appearing in this equation is precisely the contribution coming from the high-energy part of the screened interaction. We have explained above that this correction is not small, which is the reason for the failure of the static Hubbard model using the noninteracting G_d. Dividing the screened interaction into a low-energy part for $|\omega|<\Lambda$ and a high-energy part for $|\omega|>\Lambda$, we can use the explicit forms (13) and (14) given in the preceding section to obtain this first correction in the form

$$\tilde{G}^{-1}_d(k,\omega)_n = G^{-1}_d(k,\omega)_n + \frac{1}{\pi}\int_{|\nu|>\Lambda} d\nu \sum_{q,m} \text{Im } W_{nm}(q,\nu)\frac{n_F(\epsilon^m_{k-q})+n_B(\nu)}{\omega-\epsilon^m_{k-q}-\nu} + \cdots. \quad (21)$$

In particular, we see by expanding this expression to first order in ω, that the low-frequency expansion of the modified one-particle propagator reads $\tilde{G}^{-1}_d(k,\omega+i0^+)_n=(1+\alpha_{k,n})\omega-\varepsilon^n_k+\cdots$. The coefficient $\alpha_{k,n}$ is a partial contribution to the quasiparticle residue $\alpha_{k,n}=-(1/\pi)\Sigma_m\int_{|\nu|>\Lambda}d\nu \text{Im } W_{nm}(q,\nu)\times[n_F(\varepsilon^m_{k-q})+n_B(\nu)]/(\nu-\varepsilon^m_{k-q})^2$. In practice, this integral can be easily evaluated using the well-known plasmon pole approximation, which should contain most of the high-energy contribution. This correction ensures that the quasiparticle residue is obtained correctly from this "downfolded" static model. We do not, however, expect that this improvement solves the problem with the satellite discussed in the preceding section.

D. Beyond the *GW* approximation

In strongly correlated systems, it is known that the GWA is not sufficient and improvement beyond the GWA is needed. Much of the shortcoming of the GWA probably originates from the improper treatment of *short-range* correlations within the RPA. Thus, one would like in the first instance to attempt to improve these short-range correlations, which are essentially captured by the Hubbard model. The formula (15) suggests a natural way to do this, as follows. The contribution from the frequency-dependent U as well as the contribution from the downfolded Hilbert space are treated within the GWA. The self-energy G_dW_d corresponding to the *GW* self-energy of the Hubbard model with a static U can then be replaced by that obtained from more accurate theories such as DMFT (Ref. 14) or from exact methods such as Lanczos diagonalization and path-integral renormalization-group (PIRG) method.[17] Thus, if we use the LDA to construct G_d, the correction to the LDA exchange-correlation potential reads:

$$\Delta\Sigma = \Sigma_H + G_d(W-W_d) + G_rW - v_{xc}, \quad (22)$$

where Σ_H is the Hubbard model self-energy obtained from more accurate methods, replacing G_dW_d. We note also that the scheme in (15) avoids the problem of double counting, inherent in LDA+U[15] or LDA+DMFT methods.[16] In this way, it is possible to calculate the Hubbard U using the response function constructed from the LDA bandstructure.

The application of DMFT requires the Hubbard U for the impurity model. To calculate the Hubbard U for an impurity model, it is necessary to downfold contributions to the polarization P from the neighboring sites. Here, the identity in Eq. (3) shows its usefulness. Since the formulation in Eq. (3) is quite general, we merely need to redefine P_d to be the on-site polarizations, or equivalently, we redefine P_r to also include polarizations from the neighboring sites. The computational result for Ni shows that the difference between the lattice and the impurity Hubbard U is rather small, essen-

tially negligible. Somewhat different but related calculations of the Hubbard U for the impurity model has also been performed in Ref. 7 for Fe and Ni, which confirm the result obtained in our formulation.

Another feasible approach for calculating physical quantities of the derived Hubbard model is by using the path integral renormalization group (PIRG) method.[17] One advantage of this method is the possibility of using the lattice Hubbard model as opposed to the impurity model, thus including spatial fluctuations and possible symmetry breaking. The method is also suited for studying single-particle Green's functions as well as thermodynamic quantities, in particular for accurate determination of the phase diagram, which often requires a determination of the possible symmetry breaking of the Hubbard Hamiltonian after taking account of spatial and temporal fluctuations on an equal footing. In calculating the self-energy, one may substitute $G_d W_d$ by the self-energy obtained within the correlator projection method[18] together with the PIRG. These form a future challenge in the field of strongly correlated materials.

IV. SUMMARY AND CONCLUSION

In conclusion, we have investigated the construction of effective models for the correlated orbitals in materials with a natural separation of bands. In particular, we propose a simple procedure for calculating the Hubbard $U(\omega)$, which can include off-diagonal matrix elements within the d multiplets as well as off-site matrix elements. We have shown for the case of nickel that if one retains the full frequency dependence of the local components of the screened interaction, an accurate effective model can be obtained over an extended energy range. Simply neglecting this energy dependence and using the noninteracting Hamiltonian into a static Hubbard model does not provide an accurate description even at low energy. However, a proper modification of the bare propagator, obtained by integrating out high energies, allows for the construction of an effective Hubbard model which describes the low-energy physics in a satisfactory manner.

Although the present study has been based on the GWA, our finding indicates that a static Hubbard model may have a problem in describing the spectrum at intermediate and high-energy regions when the loss spectrum or Im W has a broad distribution of weight, extending down to low-energy region. This is in contrast to the case where Im W is dominated by a distinct plasmon excitation, well separated in energy from the valence band, in which case the static Hubbard model is expected to work well. It is desirable to carry out more sophisticated calculations with a frequency-dependent Hubbard model in order to have a clearer insight into the limitations of the static Hubbard model. Work in this direction is now in progress.

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