Spin-selective electronic reconstruction in quantum ferromagnets: A view from the spin-asymmetric Hubbard model

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Using the tight-binding treatment for the spin-asymmetric Hubbard model we explore the effect of electronic interactions in the ferromagnetic, partially filled Lieb lattice. As a key result we demonstrate the formation of correlation satellites in the minority spin channel. In addition, we consider the role played by transverse-field spin fluctuations in metallic ferromagnets. We quantify the degree of electronic demagnetization, showing that the half-metallic state is rather robust to local spin flips. Not being restricted to the case of a partially filled Lieb lattice, our findings are expected to advance the general understanding of spin-selective electronic reconstruction in strongly correlated quantum ferromagnets.

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I. INTRODUCTION

Metallic ferromagnetism results from the existence of permanent magnetic moments in correlated electron bands [1]. Historical examples in this class of metallic quantum systems are three-dimensional, monoatomic chains of elemental transition-metal ferromagnets like iron, cobalt, and nickel [2]. The microscopic mechanism underlying band ferromagnetism is known to be one of the most fundamental many-particle problems in condensed matter physics [3]. It is believed to be due to the interplay between Coulomb, Hund, or double-exchange interactions and kinetic energy as determined by the Pauli principle. In other words, itinerant quantum ferromagnetism results from a nontrivial interplay between the kinetic energy of itinerant electrons and many-body interactions (on-site and intersite components) as well as disorder in the solid [4]. Microscopic many-particle models for understanding the origin of itinerant ferromagnetism have been proposed independently by Hubbard [5], Kanamori [6], and Gutzwiller [7] but this remains a major unsolved and open debate problem [1]. On general grounds, ferromagnetic ground states might occur if one of the several bands of the Coulomb correlated model is dispersionless, determining the so-called Lieb (or flat-band) ferromagnetism [8,9].

The Hubbard model is known to be the canonical description of strongly correlated electron systems. It was introduced to explain ferromagnetism in transition metals [5], and since then, it has been considered a model for antiferromagnetism, unconventional superconductivity, and fractionalized phases of quantum matter. In spite of its apparent simple form, the Hubbard model encodes nontrivial many-body physics that can only be treated exactly at particular one-dimensional [10] and high-lattice-dimensional [11] limits. Importantly, this model is now considered to be one of the most fundamental among various microscopic theories for understanding hidden properties of correlated electron systems, including as shown here the formation of Hubbard satellites in fully polarized one-band ferromagnets. However, even for this one-band model Hamiltonian, a comprehensive picture of the mechanism that drives ferromagnetic order is still missing [12]. The difficulty in understanding the origin of itinerant or half-metallic ferromagnetism [13] is mainly due to the fact that the ordering and the formation of local moments are a strong-coupling, multiorbital [14,15] phenomenon and thus in general the ferromagnetic magnetic ground state is not easily described by purely perturbative techniques. Nonetheless, some exactly known results for the Hubbard model have been used as a test frame for approximate theories. The Nagaoka theorem [16], for example, states that a saturated ferromagnetic order is the ground state for the $U = \infty$ Hubbard model when one hole/electron is introduced into the half-filled band for the simple cubic lattice in three dimensions. The Mermin-Wagner theorem [17], on the other hand, rules out at finite temperatures both the ferromagnetic and the antiferromagnetic ordered state in the one-band Hubbard model below two dimensions [18]. Moreover, in addition to many-particle interaction effects, important details of the noninteracting electronic structure induced by lattice topologies, e.g., the asymmetry of the local density of states (DOS) [19] as well the existence of van Hove singularities near the Fermi energy, E_F, must be considered in some cases [20]. Thus, based on extant studies, it seems that two main ingredients favor ferromagnetism in the Hubbard model: an asymmetric DOS with singularities near E_F (e.g., the fcc DOS) [21] and nonbipartite lattices with frustration in the antiferromagnetic correlations, which can be generated by introducing next-nearest-neighbor hopping terms [20]. In this work we focus our attention on the effect of electron-electron interactions on the partially filled Lieb lattice, which, in addition to the two cases above, possesses intrinsic flat-band ferromagnetism [8]. It is noteworthy, however, that our theory and results are not restricted to the Lieb lattice and

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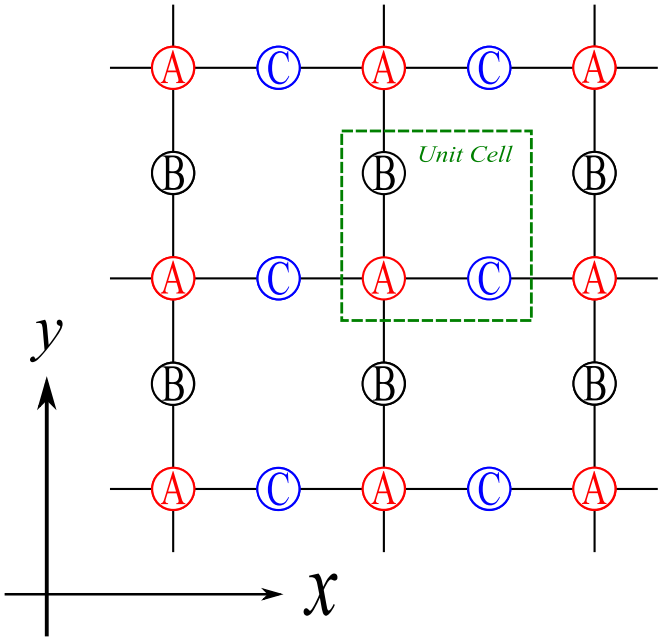

FIG. 1. Two-dimensional Lieb lattice. Its unit cell, which contains three atoms (A, B, C), is shown as the dashed line. The corresponding density of states for the noninteracting case can be found, for example, in Refs. [25] and [26].

are expected to be generally applicable to understanding the correlated electronic structure of itinerant and half-metallic one-band ferromagnets [22] within the dynamical mean-field theory (DMFT) approximation.

The two-dimensional Lieb lattice, or the line-centered square lattice, has been studied with considerable interest due to specific properties induced by its topology [23–26] as well as to understanding the interplay between on-site electronic interactions [27] and orbital degrees of freedom [27,28]. In recent years, Lieb lattices have been realized in photonic crystals [29,30] as well as in cold-atom systems [31]. Additionally, artificial Lieb lattices have been designed on the metallic copper surface [32]. The lattice is characterized by a unit cell containing three atoms (see Fig. 1) [9,23,25] and a one-particle energy spectrum showing two fully polarized (with particle-hole symmetry) bands and a dispersionless band [25,26,30]. Thus, for a lattice system, the three energy bands touch one another at the center of the spectrum (usually taken as the zero energy), and the low-energy spectrum exhibits a V-shaped Dirac cone near E_F at half-filling. In this decorated square lattice [25], when the flat band is at the Fermi energy, magnetic phase transitions can occur even with infinitesimally small electron interactions because the DOS at E_F is extremely high. Although theoretical studies on correlated quantum magnetism in the Lieb lattice have been performed in recent years [33], the role played by dynamical electron-electron interactions in the ferromagnetic state for the one-band Hubbard model remains unclear. Indeed, the general consensus dictates that dynamical correlations induced by the local Coulomb interaction U is irrelevant since the majority spin sector (spin $\uparrow$) is not scattered by the fully polarized minority spin channel. In this work we show that this understanding holds true for itinerant (spin-$\uparrow$) electrons, however, the spin-$\downarrow$ channel is shown to be strongly affected by U within the tight-binding treatment of the Hubbard model in infinite dimensions [34].

II. THEORY AND DISCUSSION

The spin-asymmetric (also known as mass-imbalanced) [36] Hubbard model on a d-dimensional lattice is described by the Hamiltonian

$$H = \sum_{\mathbf{k}\sigma} \varepsilon_{\mathbf{k}\sigma} c^{\dagger}_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow} + \sum_{i\sigma} \left(\varepsilon^{0}_{\sigma} - \mu\right) n_{i\sigma}, \quad (1)$$

where $c^{\dagger}_{i,\sigma}$ are the creation operators for electrons with spin $\sigma(=\uparrow,\downarrow)$ at site i and $n_{i\sigma} = c^{\dagger}_{i\sigma} c_{i\sigma}$. In Eq. (1) $\varepsilon_{\mathbf{k}\sigma}$ describes a ferromagnetic, spin-dependent [14,35] electron band dispersion relation for the two spin channels, ε^{0}_{σ} (here chosen to be spin independent [36] and equal to $-U/2$) is the on-site energy level of the localized one-band states, μ is the chemical potential of the system, and U is the on-site Coulomb interaction.

For the sake of simplicity let us focus our attention on the *scattering correction* approximation of the tight-binding treatment in the large-d limit [34], which allows us to treat Eq. (1) self-consistently. This approximation is known to provide the exact solution [11] for the conduction electrons of the simplified Hubbard model (usually dubbed the spinless Falicov-Kimball model) [37] and it is formally identical to that obtained in Ref. [38]. For problems of local disorder the set of equations described below corresponds to the coherent potential approximation, which is the exact solution of the Anderson localization problem [39] within the large-d limit [40]. While an extended description of technical aspects related to the tight-binding (or linked cluster) treatment for the Hubbard Hamiltonian can be found in Refs. [41] and [42], below we briefly describe the self-consistent formalism [34] used here to study Eq. (1) and to understand the role played by electron-electron interactions and one-particle spin-flip fluctuations induced by a local transverse field [43] in itinerant quantum ferromagnets within DMFT.

The linked cluster expansion (or the tight-binding treatment) for the Hubbard model [34,41,42] starts by assuming that the local part of the unperturbed Hamiltonian is given by the last two terms in Eq. (1). The solution of the *local* unperturbed Hamiltonian provides a basis of two coupled spin subspaces, and the corresponding zero-order retarded Green's functions are given by $\mathcal{G}^{0}_{\sigma}(\omega) = \frac{1-\langle n_{\bar{\sigma}}\rangle}{\omega-\varepsilon^{0}_{\sigma}+\mu} + \frac{\langle n_{\bar{\sigma}}\rangle}{\omega-(\varepsilon^{0}_{\sigma}+U)+\mu}$, where $\omega \equiv \omega + 0^{+}$ and $\langle n_{\bar{\sigma}}\rangle$ are the fully renormalized on-site occupancies (or the average electronic density) for the $\bar{\sigma}$ electrons, which encodes the probability of finding a $\bar{\sigma}$ electron at a given site on the lattice. Next we consider the full limit of Eq. (1), where the hopping terms are explicitly taken into account. At any lattice dimension d, an approximate solution to this many-particle problem can be obtained by means of a tight-binding treatment around the atomic limit [42]. Using $\mathcal{G}^{0}_{\sigma}(\omega)$ above the single-particle Green's function within the Hubbard I approximation [5] reads $[G_{\mathbf{k}\sigma}(\omega)]^{-1} = [\mathcal{G}^{0}_{\sigma}(\omega)]^{-1} - \varepsilon_{\mathbf{k}\sigma}$. The next step towards a self-consistent treatment of the Hubbard Hamiltonian is achieved by adding high-order, on-site and intersite corrections to the atomic one-particle Green's function

$\mathcal{G}^0_\sigma(\omega)$ which are encoded in the irreducible nonlocal propagator $\mathcal{G}_{\mathbf{k}\sigma}(\omega)$ [41,42]. It is noteworthy that this propagator, which in the general case gives the momentum and frequency dependence for the many-particle self-energy $\Sigma_{\mathbf{k}\sigma}(\omega) \equiv \omega - \mathcal{G}^{-1}_{\mathbf{k}\sigma}(\omega)$, is irreducible in the sense that it cannot be divided into two pieces by cutting a single hopping line [42]. However, as shown in Ref. [34], in the limit of infinite spatial dimensions the self-energy loses its $\mathbf{k}$ dependence and only the site-diagonal part of the irreducible propagator survives in the large-d limit. Thus, since the $\mathbf{k}$ dependence in this limit is restricted to the tight-binding energies $\varepsilon_{\mathbf{k}\sigma}$, the sum over $\mathbf{k}$ can be directly performed, and the single-particle Green's function at site i for the σ channel is given by

$$G_{ii\sigma}(\omega) = \frac{1}{N}\sum_{\mathbf{k}} \frac{1}{[\mathcal{G}_\sigma(\omega)]^{-1} - \varepsilon_{\mathbf{k}\sigma}}. \tag{2}$$

In the high-dimensional limit [40], one can write the irreducible propagators in terms of the single-site one-particle Green's functions and the dynamical (Weiss) mean field using the relation [34]

$$\mathcal{A}_\sigma(\omega) = [\mathcal{G}_\sigma(\omega)]^{-1} - [G_{ii\sigma}(\omega)]^{-1}. \tag{3}$$

It is worth mentioning here that $\mathcal{A}_\sigma(\omega)$ describes the motion of an electron through the surrounding medium of a site i, i.e., the rest of the lattice [34]. Its effect can be viewed as equivalent to that of a time-dependent (effective) field coupling the lattice site i to two reservoirs, one for each spin direction. We recall here that the basic principle behind the DMFT treatment is to replace the lattice problem with a self-consistently embedded (Anderson) impurity problem [40] and the self-consistency condition requiring the local impurity Green's function to be equal to the local Green's function for the lattice, which, in the tight-binding formalism, reads [34,44]

$$G_{ii\sigma}(\omega) = \frac{1}{N}\sum_{\mathbf{k}} \frac{1}{[G_{ii\sigma}(\omega)]^{-1} + \mathcal{A}_\sigma(\omega) - \varepsilon_{\mathbf{k}\sigma}}. \tag{4}$$

Finally, in order to have the complete set of equations for the relevant one-particle Green's functions as well as the dynamical Weiss fields, an explicit solution of the single-site problem for each spin channel must be obtained. The tight-binding perturbation treatment around the atomic limit provides a direct way of solving the single-site problem by means of a perturbative expansion in the dynamical field $\mathcal{A}_\sigma$ [34]. Starting from the unperturbed local Green's functions, $\mathcal{G}^0_\sigma(\omega)$, two perturbation treatments in the local fields can be summed exactly if local contributions due to spin fluctuations induced by many-particle Coulomb scattering processes are not taken into account [34,38] and in this regime the local one-particle Green's function for both spin channels is given by

$$G_{ii\sigma}(\omega) = \frac{1-\langle n_{\bar\sigma}\rangle}{\omega - \varepsilon^0_\sigma + \mu - \mathcal{A}_\sigma(\omega)} + \frac{\langle n_{\bar\sigma}\rangle}{\omega - \varepsilon^0_\sigma - U + \mu - \mathcal{A}_\sigma(\omega)}, \tag{5}$$

which has exactly the same form as that obtained from the *scattering correction* term of the Hubbard III approximation for the Hubbard model [38].

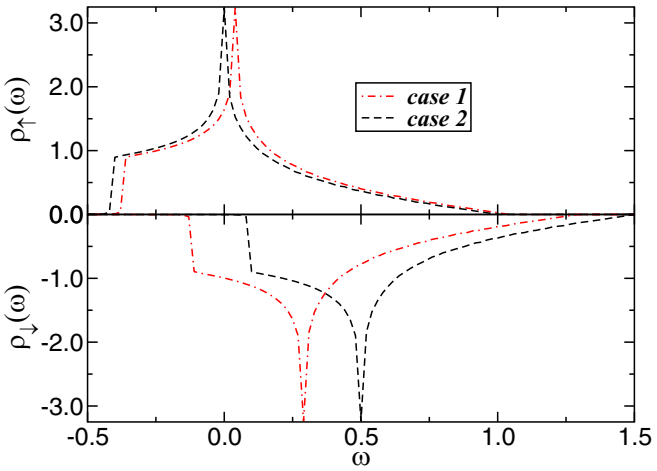

FIG. 2. Uncorrelated density of states for two spin polarization channels at quarter-filling. While the spectral density of the majority (spin-↑) carriers are almost half-filled and span the Fermi energy $E_F(=\omega=0)$, the spin-↓ electron band is completely empty in case 2 as discussed in the text.

Equations (4) and (5) constitute a closed system of equations for the Hubbard Hamiltonian. The self-consistent solution of Eqs. (4) and (5) can be performed numerically for different lattices both in the paramagnetic and for correlated ferromagnetic systems in the full frequency (energy) range. In what follows, we consider the case of a ferromagnetic, quarter-filled Hubbard model in the spin-split Lieb lattice. We focus our attention on the lowest-energy dispersive band and ignore the multiband contributions [9,23] coming from the flat and the dispersive conduction bands. In the partial band filling considered here (quarter-filling in the case of the one-band Hubbard model) the higher-energy conduction band states will form fully empty band dispersions with vanishing Coulomb correlation effects, while the flat bands not explicitly included in our theory will give rise to localized magnetic moments responsible for the ferromagnetic ground state as guaranteed by the Lieb theorem [8]. The dispersion relations we use to describe a (nearly) half-metallic ferromagnetic state for the $2d$ Lieb lattice [9] are written as

$$\varepsilon_{\mathbf{k}\sigma} = \Delta_\sigma - 2\sqrt{t_x^2\cos^2(k_x/2) + t_y^2\cos^2(k_y/2)}, \tag{6}$$

where k_x (k_y) is the momentum component along the x (y) direction, $t_x = t_y = 0.5$, and Δ_σ are the spin-dependent band shifts needed to describe a ferromagnetic ground state [22] with free electron band dispersions as in Fig. 2. Thus, generally speaking and similarly to Ref. [22], where an external magnetic spin splitting Δ was added to the one-band Hubbard Hamilation to mimic the local Hund's polarization present in real multiorbital systems, the constrained (quarter-filled) regime considered here for the spin-asymmetric Hubbard model is characterized by an almost-half-filled majority (spin-↑) electron band and two distinct regimes for the minority (↓) spin channel, with a partially filled (case 1) or fully spin-polarized (case 2) electron band. The electronic states displayed in Fig. 2 are obtained assuming $\Delta_\uparrow = 1.04$, $\Delta_\downarrow = 1.29$ (case 1) and $\Delta_\uparrow = 1.0$, $\Delta_\downarrow = 1.5$ (case 2). In the following we present our analysis of the frequency dependence of the spin-resolved

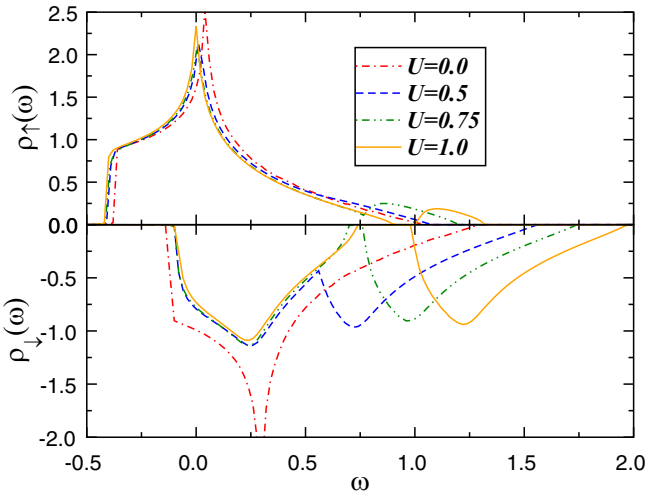

FIG. 3. Effect of electron-electron interactions in the spin-asymmetric Hubbard model. Note the large transfer of spectral weight and the emergence of Hubbard satellites in the minority spin channel with increasing U.

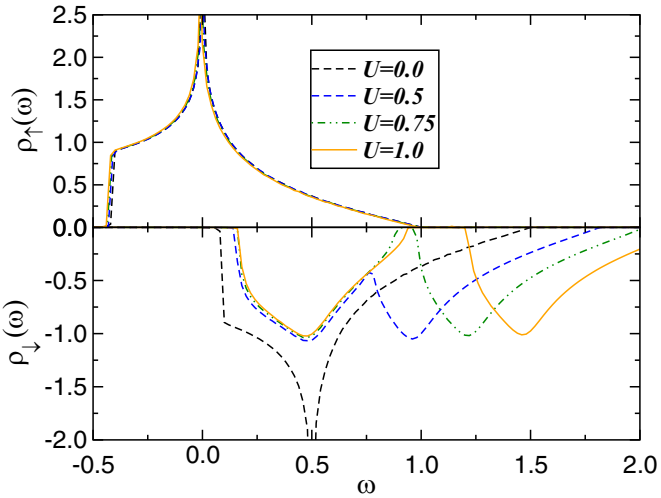

FIG. 4. Local Coulomb correlation effects in the fully polarized, spin-asymmetric Hubbard model. While the majority band is protected against strong electron interactions, a large dynamical transfer of spectral weight is visible in the minority spin channel with increasing U.

one-particle spectral functions, showing the high-energy Mott counterpart of spin-selective Kondo [45] localization in correlated ferromagnets.

A. Role of on-site Coulomb interaction

In Fig. 2 we display the uncorrelated DOS for the two cases described above. As shown the majority (↑-spin) band at quarter-filling spans the Fermi energy ($E_F = \omega = 0$), however, for the minority spin channel, only in case 1 do electronic states set in at E_F. While in this regime the van Hove singularity is found above E_F, for $\Delta_\uparrow = 1.0$ (case 2) it appears exactly at E_F for the majority spin channel. Interestingly, case 2 represents a fully polarized ferromagnet, also referred to as a saturated ferromagnet [14], where the minority spin channel is empty and the electronic states reside only in the conduction band. This saturated scenario is applicable to half-metallic systems like CrO_2, a multiorbital system without minority electronic states crossing E_F [46].

Let us now turn our attention to the self-consistent (DMFT) results in Figs. 3 and 4, where the effect of on-site electron-electron interaction in the two cases introduced above is shown for both spin channels. As expected for one-band itinerant ferromagnets, our results in Figs. 3 and 4 show that the majority spin (↑) channel is weakly renormalized by the on-site Coulomb interaction U. This is linked to the strong polarization of the minority carriers or the low on-site occupancy, $\langle n_\downarrow \rangle$, self-consistently computed using Eqs. (4) and (5). The main effect shown in the upper panel in Fig. 3 is the appearance of an incipient upper Hubbard band at energies $\omega = 0.75$ above E_F in case 1. On the other hand, electrons in the minority spin channel are strongly scattered by the itinerant carriers, with the concomitant appearance of two prominent Hubbard satellites with increasing U. In strongly correlated electron systems the presence of correlation satellites are known to be the precursor of the Mott-Hubbard band splitting, which here is a manifestation of spin-selective high-energy electronic reconstruction in quantum ferromagnets. Interestingly, our results imply that the spin-up electronic configuration (with half-filled conduction band states) causes satellites in the minority spin channel upon the addition or removal of an itinerant electron at each site on the lattice. It is worth mentioning as well that the line shape of the Hubbard satellites and the emergence of a high-energy incoherent electronic structure in the conduction band are many-particle fingerprints of strong electron-electron interactions within DMFT [34,40,42]. However, as expected within the linked cluster framework, we do not observe a first-order electronic reconstruction in the minority spin channel; instead, correlation satellites are accompanied by smooth crossover with increasing U. Taken together, these results imply a nontrivial rearrangement of the conduction band electronic structure which could be tested in future experiments on correlated quantum ferromangnets.

B. Role of local transverse spin-flip fluctuations

Our results above are important for understanding the dynamical nature of high-energy electronic reconstruction and the emergence of Hubbard satellites in the minority electronic states as well as intrinsic low-energy and higher-energy spectral features probed in inverse-photoemission spectroscopy in itinerant ferromagnets like CrO_2 [47]. However, it remains to be understood how robust these results are against local spin fluctuations induced, for example, by spin-orbit interactions in Coulomb interacting ferromagnets [48]. To provide an answer to this fundamental problem we now investigate the role played by local transverse-field spin flips in our strongly correlated DMFT ($U = 1$) results.

For one-band systems the local transverse-field Hamiltonian can be written as $H_\lambda = \lambda \sum_i (c^\dagger_{i\uparrow} c_{i\downarrow} + \text{H.c.})$ [43]. Here, λ is a $\mathbf{k}$-independent (or local) spin-flip term, which is taken as an additional model parameter in our description. Physically, H_λ acts as a transverse magnetic field and locally mixes the

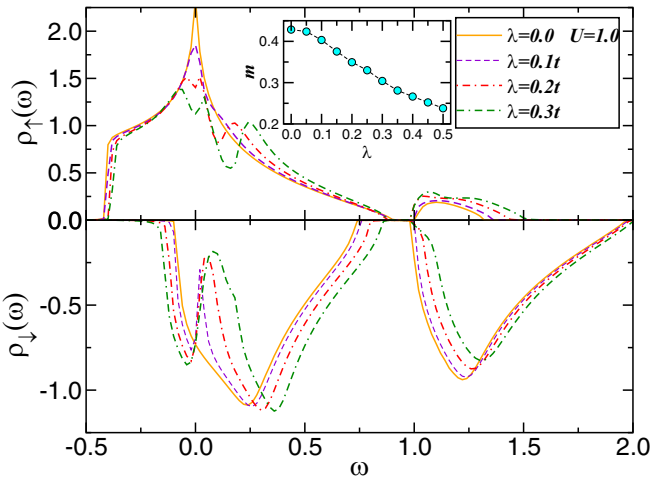

FIG. 5. Effect of transverse-field-induced local spin fluctuations in the spin-asymmetric Hubbard model. Note the substantial changes in the correlated spectral functions of the spin-polarized charge carriers. Interesting is the enhancement of the upper Hubbard band in the majority channel and the changes in spectral weight of the spin-↓ valence band upon increasing the local transverse field λ. Inset: Monotonic reduction of the total magnetization, m, due to electronic depopulation of the majority spin channel.

two spin states described in Eq. (1). As shown in Ref. [43], the formal exact solution of the complete problem, i.e., $\bar{H} = H + H_\lambda$, can be obtained by performing a perturbative treatment on the transverse field (or the spin-dependent hybridization) term. The single-particle Green's function in a transverse field reads

$$G_{ii\sigma}(\omega) = \frac{1}{N}\sum_{\mathbf{k}} \frac{1}{[\mathcal{G}_\sigma(\omega)]^{-1} - \varepsilon_{\mathbf{k}\sigma} - \lambda^2 \mathcal{G}_{\bar{\sigma}}(\omega)}. \quad (7)$$

The DMFT self-consistent solution of these spin-coupled, single-site Green's functions together with Eqs. (3) and (5) provides a direct scheme to describe transverse spin fluctuations of correlated electron systems, including, as shown here, the monotonic switching of ferromagnetic aligned spins in metallic ferromagnets.

With this in mind, in Fig. 5 we display the effect of the local transverse field on the correlated ($U = 1$) spectral function. In the top panel the transverse-field-induced transfer of the spectral weights is visible from low energies to the upper Hubbard band above $\omega = 1$. Though not too pronounced the spectral weight transfer is not rigid in nature. Interesting as well is the fact that the high-energy electronic structure found in the majority spin channel remains nearly the same with increasing λ. On the other hand, the correlated one-particle band gap of the minority carriers shrinks as λ is increased. This high-energy incoherent response is shown in the bottom panel in Fig. 5, where we demonstrate that a strong enough transverse field can induce spectral weight transfer over large energy scales as well as the formation of a pseudogap in the minority spin electron band near E_F. Noteworthily, while the spin-↓ electrons remain metallic up to higher λ values, their electron occupation increases with λ. This result implies a reduction in the electronic polarization of the ferromagnetic state. The overall change in the magnetic behavior with fixed U is illustrated in the inset in Fig. 5 (top panel), where the transverse field leads to decreasing magnetization, $m(\equiv \langle n_\uparrow\rangle - \langle n_\downarrow\rangle)$, as $\langle n_\downarrow\rangle$ grows monotonically at the expense of $\langle n_\uparrow\rangle$. This weak suppression of ferromagnetism is reflected by a reduction in the polarized [14], $P = (\langle n_\uparrow\rangle - \langle n_\downarrow\rangle)/(\langle n_\uparrow\rangle + \langle n_\downarrow\rangle)$, magnetically ordered state due to transverse-field-induced electronic depopulation of the majority spin channel. Our results thus suggest that small changes in the spin-selective band fillings will influence the line shape of correlated electronic states of strongly spin-polarized spin-orbit systems. Nevertheless, the net magnetization m remains finite for realistic (or not too large) λ values and this behavior serves as a proof that the ferromagnetic ground state is rather robust to local spin-flip fluctuations. Future work is needed to identify these specific changes in real spin-orbit ferromagnets.

III. CONCLUSION

In conclusion, we have used the dynamical mean-field theory method to study spin-selective electronic reconstruction in the spin-asymmetric Hubbard model. While our results are expected to be generally applicable to correlated ferromagnetic lattice systems [49], we have discussed them in the context of a partially filled Lieb lattice with spin-dependent electron band dispersions. Electron-electron interactions are shown to induce the emergence of Hubbard satellites in the minority spin channel and high-energy band gaps in the correlated spectral functions at large U. Our model is appropriate for understanding correlation effects in quantum ferromagnets, since it explains why the majority (spin-↑) channel has a noninteracting spectrum while the spin-↓ electrons are shown to be strongly scattered via a local Coulomb repulsion mechanism. Moreover, by investigating the influence of a transverse (magnetic) field, we find that the ferromagnetic properties are sensitive to local spin-flip fluctuations. Finite transverse field (or spin-orbit) effects depolarize the spin-split electron states, with a degree of spectral weight transfer which depends on intrinsic dynamical correlation effects induced by the on-site Coulomb interaction U. Our results are expected to be important for understanding correlation and spin-orbit effects in electronic states at the $LaAlO_3/SrTiO_3$ interface, where the splitting of t_{2g} orbital degeneracy at the polar interface between these two insulating transition-metal oxides leads to a quarter-filled xy band (0.5 electron per Ti) in the top TiO_2 ferromagnetic layer [50].

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