Induced magnetism versus Kondo screening in alternating Mott-metal layers

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We investigate the magnetic properties of a heterostructure comprised of alternating metallic and Mott insulating layers of fermions with varying interlayer hybridization. Results from large-scale quantum Monte Carlo simulations at half-filling show clear evidence of induced magnetism in the metallic layers due to coupling to the Mott insulating layers at small to intermediate values of interlayer hopping. The in-plane magnetism is completely suppressed via a Kondo proximity effect when the coupling between adjacent layers is increased beyond a critical strength. The nature of the phase in the Kondo-like insulating regime is investigated and is shown to be dominated by simultaneous in-plane and interplane short-range correlations.

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I. INTRODUCTION

The appearance of novel quantum phases at the interface between correlated materials with mismatched correlations has garnered great interest in recent years. The properties of these emergent phases are often distinct from a simple interpolation between the bulk properties of the constituents. Advances in synthesis methods[1] that can produce atomically smooth surfaces has facilitated the discovery of novel interfacial phases in many composite layered materials. These include tunable high-mobility quasi-2D electron gas in insulating oxide heterostructures,[2,3] giant magnetoresistance,[4] and novel magnetic properties at ferromagnet-superconductor interfaces,[5,6] and suppression of magnetic susceptibility at metal-Mott insulator interfaces.[7] Interest in these systems is driven by both fundamental and technological motivations. The fundamental motivation lies in gaining deeper insight into the mechanism of the emergence of novel quantum phases from the subtle interplay between competing interactions. The technical motivations are driven by the prospect of engineering interfaces with controllable properties. Such tailor-made materials with desired functionalities are widely believed to drive the next generation of electronic devices. Advances in synthesis methods have been accompanied by similar progress in characterization techniques. In particular, following the recent development of standing-wave angle-resolved photoemission spectroscopy (SW-ARPES),[8] it is now possible to probe electronic properties of individual layers in a multilayer system.

On the theoretical front, multiple numerical methods have been used to study these heterostructures. The appearance of metallic interface between oxide band insulators has been investigated with Hartree-Fock[9] and Lanczos methods,[10] whereas dynamical mean-field theory (DMFT)[11–13] and quantum Monte Carlo[14–16] were used to study metal-Mott insulator junctions. Interestingly, initial DMFT results on the inpenetrability of metallicity into the Mott insulator were later contradicted by inhomogeneous DMFT studies that predicted a "fragile" Fermi liquid behavior at sufficiently low temperatures in the insulating layer sandwiched between two metallic layers. Previous QMC studies are consistent with the former picture, although the effects of finite temperature in these studies cannot be ruled out.

In this paper, we use the determinant quantum Monte Carlo (DQMC) method[17–19] to investigate the properties of a composite system comprised of alternating metallic layers (M) comprised of free fermions and Mott insulators (I) with long-range antiferromagnetic (AFM) order. The primary focus of the study is to investigate (i) the potential onset of induced magnetism in the metallic layers due to coupling to the antiferromagnetically ordered insulating layers, (ii) the suppression of in-plane long-range magnetic order via the Kondo screening mechanism, and (iii) the nature of the emergent phase in the Kondo insulating regime. In contrast to previous QMC studies of multilayered structures *with a single interface*,[14,15] we find that the metallic layers acquire induced magnetic order over a finite range of interlayer hopping. This is in agreement with experimental observation in the organic conductor (Me- 3,5-DIP)×[Ni(dmit)$_2$]$_2$.[7] Our results elucidate the mechanism behind the Kondo screening and the nature of the emergent phase at large interlayer hybridization.

II. MODEL AND METHODOLOGY

The low-energy electronic properties at metal-insulator interfaces are adequately described by the multilayer Hubbard model with intra- and interlayer hopping of electrons and layer-dependent on-site interactions. The Hamiltonian is

$$\begin{aligned}\mathcal{H} = &- \sum_{\langle i,j\rangle,l,\sigma} t_l (c^\dagger_{il\sigma} c_{jl\sigma} + c^\dagger_{jl\sigma} c_{il\sigma}) \\ &+ \sum_{i,l} U_l \left(n_{il\uparrow} - \frac{1}{2}\right)\left(n_{il\downarrow} - \frac{1}{2}\right) \\ &- \sum_{i\langle l,l'\rangle,l,\sigma} t_{ll'} (c^\dagger_{il\sigma} c_{il'\sigma} + c^\dagger_{il'\sigma} c_{il\sigma}). \qquad (1)\end{aligned}$$

$c^\dagger_{il\sigma}(c_{jl\sigma})$ creates (annihilates) an electron with spin σ at site i in layer l; $n_{il\sigma}$ is the corresponding number operator. U_l denotes the layer-dependent strength of on-site interaction, while t_l and $t_{ll'}$ denote, respectively, the intralayer nearest-neighbor hopping in layer l and interlayer hopping between nearest-neighbor sites on (adjacent) layers l and l'. Each layer consists of a square lattice of dimension $L \times L$. The odd-l layers are comprised of correlated ($U_l \neq 0$) electrons, whereas uncorrelated (free) electrons populate the even-l layers. We have chosen open boundary conditions in the z direction to capture the behavior of real heterostructures where the outermost layers behave differently from those in the bulk.

The model (1) has a large parameter space—the Hamiltonian parameters can, in principle, be different for each layer. In the present study, we consider the simplest situation where (i) the intralayer hopping amplitudes are the same for every layer, $t_l = t$; (ii) the interlayer hopping is identical between all pairs of adjacent layers, $t_{ll'} = V$; and (iii) the strength of the on-site interaction U is the same for the three correlated layers. The number of particles in each layer is fixed at $\langle n_{il\sigma}\rangle = \frac{1}{2}$. This establishes particle-hole symmetry in the problem. We set the intralayer hopping t as the unit of energy—all the other Hamiltonian parameters as well as the inverse temperature β will be expressed in units of t.

The physics of the Hamiltonian (1) has more than a passing similarity to the Kondo effect in heavy fermion systems. Qualitatively similar interlayer pair formation via the Kondo mechanism is responsible for the suppression of long-range order in both systems, even though the localized moments in the heavy fermion systems do not possess charge degrees of freedom and the interaction between the conduction electrons and localized moments are described by the Heisenberg interaction rather than the interlayer hopping considered here.

We have used the DQMC method to simulate the Hamiltonian (1) on finite-sized lattices. The DQMC is a finite-temperature quantum Monte Carlo (QMC), which is based on the decoupling of the quartic on-site interaction term into quadratic terms (in fermionic operators) coupled to a space- and imaginary-time-dependent auxiliary field via a discrete Hubbard-Stratonovich transformation. Imaginary time discretization is applied in the form of Suzuki-Trotter decomposition. The quadratic fermion terms are integrated analytically and the resultant fermion functional determinants are used as statistical weights for Monte Carlo sampling of the auxiliary fields. Ground-state estimates for various observables are obtained from simulations at sufficiently low temperatures. The DQMC method is free of the negative sign problem for fermions at half-filling on any bipartite lattice in any dimension, making it an ideal choice for studying the current problem. We have used the QUEST simulation package[20] to implement the DQMC algorithm. We have studied six-layer structures of dimension $6 \times L \times L$ ($6 \leqslant L \leqslant 12$) with periodic boundary condition in the layers. An inverse temperature of $\beta = L$ with an imaginary time step of $\Delta\tau = 0.25$ was found to be sufficient for the observables to have converged to their ground-state values.

For a finite lattice, spin rotation invariance is never broken for the Hamiltonian (1). A quantitative measure of the long-range magnetic in different ground-state phases is obtained from the longitudinal and transverse components of the static structure factor defined as

$$S^{zz}(\mathbf{k}) = \frac{1}{N}\sum_{i,j}\left\langle S_i^z S_j^z\right\rangle e^{i\mathbf{k}\cdot(\mathbf{r}_i-\mathbf{r}_j)},$$
$$S^{+-}(\mathbf{k}) = \frac{1}{N}\sum_{i,j}\langle S_i^+ S_j^- + S_j^+ S_i^-\rangle e^{i\mathbf{k}\cdot(\mathbf{r}_i-\mathbf{r}_j)}, \quad (2)$$

where $S_{il}^x = c_{il\uparrow}^\dagger c_{il\downarrow} + c_{il\downarrow}^\dagger c_{il\uparrow}$ and $S_{il}^z = c_{il\uparrow}^\dagger c_{il\downarrow} - c_{il\downarrow}^\dagger c_{il\uparrow}$ are the components of the local spin operator in terms of the fermionic creation and annihilation operators. Long-range Ising (XY) AFM order in the thermodynamic limit is indicated by a finite value of the measured $S^{zz}(\mathbf{Q})[S^{+-}(\mathbf{Q})]$ at the AFM ordering wave vector $\mathbf{Q} = (\pi,\pi)$, when extrapolated to infinite system size. For isotropic interactions (in spin space), as is true in the present Hamiltonian, the two quantities are simply related by $S^{+-}(\mathbf{k}) = 2S^{zz}(\mathbf{k})$ and we have used the average of the two in our simulations,

$$S_l^{\mathrm{AF}} = \tfrac{1}{3}\left[S_l^{+-}(\pi,\pi) + S_l^{zz}(\pi,\pi)\right]. \quad (3)$$

Further insight into the nature of the ground-state phase is obtained from the zero-momentum spectral function, or single-particle density of states $A_l(\omega)$, related to the imaginary-time-dependent single-particle Green's function, $G_l(\tau) = \sum_{i,\sigma}\langle T c_{il\sigma}(\tau) c_{il\sigma}^\dagger(0)\rangle$, by the integral equation

$$G_l(\tau) = \int_{-\infty}^{\infty} d\omega \frac{e^{-\omega\tau}}{1+e^{-\beta\omega}} A_l(\omega). \quad (4)$$

$G_l(\tau)$ is obtained directly from the DQMC simulations. We employ the maximum entropy method[21] to invert the integral equation (4).

III. RESULTS

At small values of the interlayer hybridization V, the system is adiabatically connected to a set of uncoupled correlated and uncorrelated layers. The ground state of the correlated layers are characterized by long-range AFM order; the uncorrelated layers are in a nonmagnetic, metallic state. As the interlayer hopping is turned on, the uncorrelated layers acquire finite long-range induced magnetic order due to coupling to the correlated layers, while at the same time, the magnetic ordering in the correlated layers is reduced. As the hybridization between adjacent layers is increased, interlayer pairing of the fermions destroys the long-range magnetic order at a finite critical V_c that depends on the value of on-site interaction U in the correlated layers. As we shall see later, this phase is characterized by short-range order in the planes. Finally, at a characteristic value of interlayer hybridization that is independent of U, there is a crossover to a phase dominated by interlayer hopping and pairing.

The simulation results for the intraplanar magnetic orders in the individual layers are shown in Fig. 1 (main panel) as a function of varying interplanar hybridization for a representative value of the on-site interaction ($U = 8t$). The results are obtained from extrapolation of data for finite-sized lattices to the thermodynamic limit. For $0 < V \lesssim 2t$, the uncorrelated layers acquire finite magnetic order induced by hybridization with the correlated layers. The in-plane staggered magnetization initially increases and reaches a maximum at an intermediate value of V before decreasing to zero at a critical $V_c \approx 2.4t$. The peak height and position depend on the value of on-site repulsion U. On the correlated layers, the long-range AFM order is reduced by hybridization with the metallic layers—the structure factor decreases monotonically with increasing V and vanishes at approximately the same V_c as the uncorrelated layers. Due to errors associated with extrapolation of the finite-sized data, it is not possible to conclude with certainty from our numerical results if they indeed vanish at exactly the same V_c, although results from finite-size systems appear to support such a conclusion. The

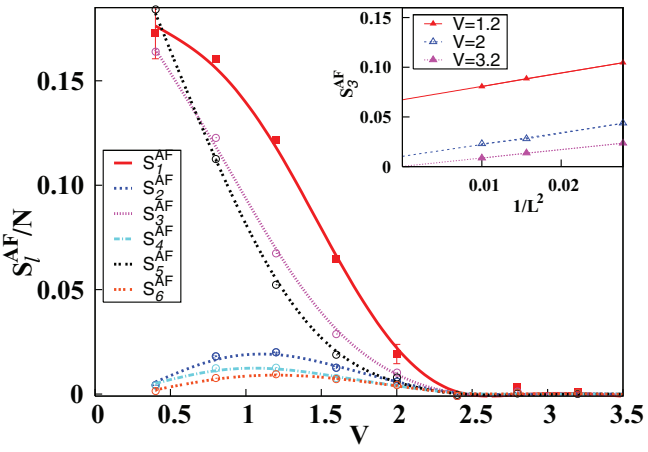

FIG. 1. (Color online) The intraplanar AFM structure factor for the individual layers at $U = 8t$. The main panel shows the extrapolated values of S_l^{AF} as a function of the interlayer coupling, V. The inset shows the extrapolation of the data from finite-sized systems to the thermodynamic limit. For $V \lesssim 2t$ the extrapolated structure factor is nonzero for the metallic layers, indicating the existence of induced long-range order (LRO). With further increase of V, LRO vanishes simultaneously in both correlated and uncorrelated layers.

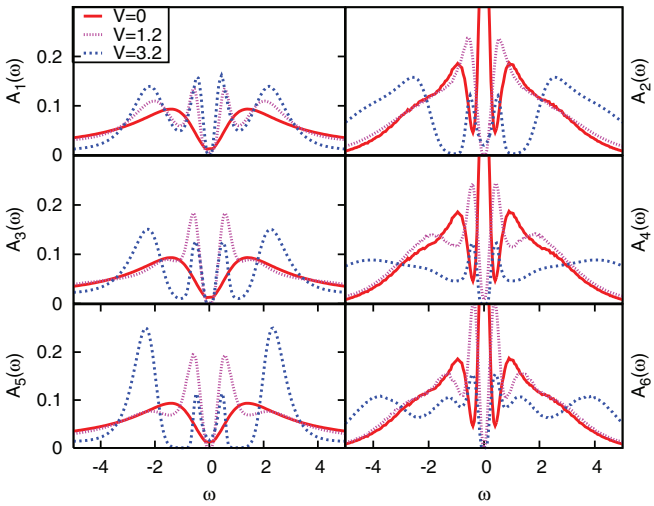

FIG. 2. (Color online) Spectral density for the individual layers at some representative values of V for $U = 8t$. The data for $V = 0$ and 1.2 are for a lattice of size $6 \times 10 \times 10$ at $\beta = 20t$. Results for $V = 3.2$ are for a lattice of dimensions $6 \times 6 \times 6$ at $\beta = 20$. Note: The density of states is broadened by the maximum entropy program.

loss of long-range magnetic order is expected to vanish via a order-disorder transition belonging to the O(3) universality class, but our data is too noisy to test such a hypothesis.

The magnitude of in-plane ordering varies with the number of interfaces. Since layer 1 has one interface with an uncorrelated layer as opposed to layers 3 and 5 that have two interfaces each with uncorrelated layers, the suppression of long-range AFM order in layer 1 is smaller than those in layers 3 and 5. Consequently, the induced magnetism in the uncorrelated layer 2, which is coupled to layer 1, is higher than that measured in layers 4 and 6. Interestingly, despite the difference in magnitudes at intermediate V, the vanishing of long-range magnetic order occurs at the same V_c (within error bars) for all the layers. As discussed in the Introduction, our results agree with experimental observation in the organic conductor (Me- 3,5-DIP)$\times$[Ni(dmit)$_2$]$_2$. On the other hand, previous QMC studies have shown that such induced magnetism is is absent in multilayer M-I systems with a single interface. The necessity of multiple M-I interfaces to induce magnetism in the uncorrelated layers underscores the weakness of the induced magnetism and its fragility towards suppression by interaction with other metallic layers. This is crucial for any future device design using such systems.

The single-particle spectral density $A(\omega)$ provides valuable insight into the nature of ground-state phases in any interacting many-body system. The density of states at the Fermi surface, $N_0 = A(\omega = 0)$, distinguishes a metallic phase ($N_0 > 0$) from an insulating one ($N_0 = 0$). Furthermore, the nature of suppression of the zero-frequency spectral weight provides information regarding the origin of the insulating behavior. Importantly, the spectral density can be measured experimentally with ARPES. As mentioned earlier, the recent breakthrough in the development of SW-ARPES makes it possible to probe strong correlations layer by layer in a multilayer system. In fact, ARPES is more versatile and measures the full momentum-dependent single-particle spectral function $A(k,\omega)$ in a translationally invariant system such as the one described by (1), but in the present study we restrict ourselves to the investigation of only $\sum_k A(k,\omega)$.

The simulation results for the spectral density are shown in Fig. 2 for the individual layers at some representative values of V. For $V = 0$, the metallic layers feature a prominent peak at $\omega = 0$ (finite density of states). For the correlated layers, the strong on-site interaction (data for $U = 8$ are shown in the figure) induces a Mott gap which is reflected in a vanishing $A(\omega = 0)$. At intermediate values of the interlayer hopping, $0 < V < V_c$, the induced magnetization in the metallic layers is accompanied by the opening of a (much smaller) Slater gap reflected in the emergence of resonance peaks in the spectral density for the metallic layers. At the same time, the penetration of metallicity in the correlated layers due to coupling to the uncorrelated layers results in a reduction of the magnitude of the gap, but does not induce a finite density of states. Strictly speaking, the gap in this regime is not a true Mott gap. Instead, its nature can at best be qualitatively described as being intermediate between a Mott and a Slater gap. For $V > V_c$, a vanishing density of states and strong Kondo resonance peaks in the spectral density for both the correlated and uncorrelated layers mark the complete suppression of intraplanar long-range order by the Kondo proximity effect. The spectral density data reinforce the evidence of penetration of Mott-ness in the metallic layers in an alternating layer geometry, in contrast to previous studies of multilayer systems with a single interface.

What is the nature of the ground state at $V > V_c$? It is generally acknowledged that the destruction of magnetism at a Mott-metal interface occurs via a "Kondo proximity effect" at large values of interlayer hopping V. A strong on-site interaction U in the correlated layers results in a large charge gap that prevents the free hopping of fermions between the metallic and correlated layers at small values of interlayer

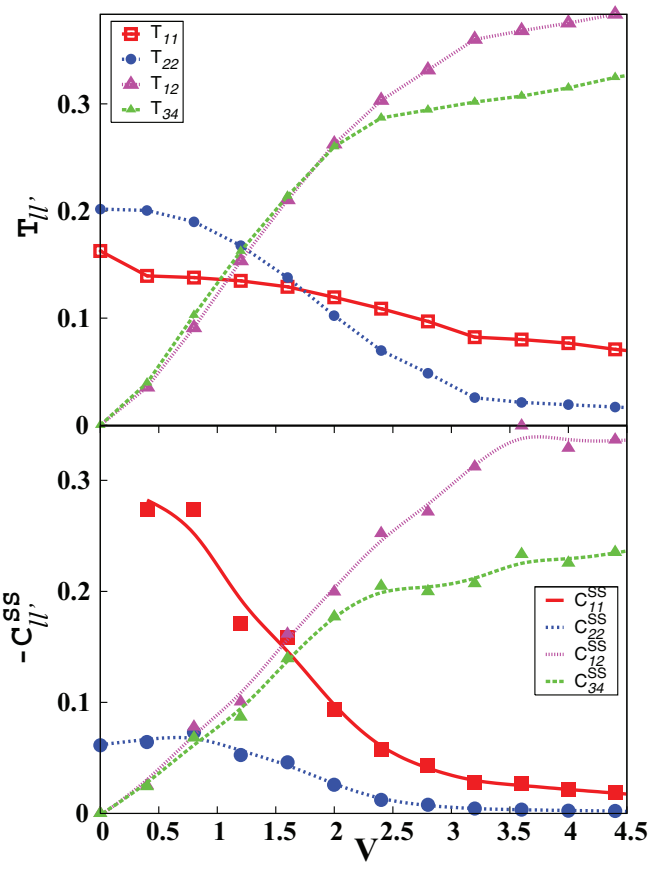

FIG. 3. (Color online) Short-range correlations. Top: Intralayer hoppings T_{11}, T_{22} and interlayer hopping T_{12} at the surface and T_{34} in the "bulk." Bottom: Nearest-neighbor intralayer spin-spin correlation for correlated (C_{11}^{SS}) and uncorrelated (C_{22}^{SS}) layers and interlayer spin-spin correlation C_{12}^{SS} and C_{34}^{SS} at and away from the surface. For $V \gtrsim 2$, both the interlayer hopping and spin-spin correlations are reduced for layers away from the surface.

hybridization. Large values of V, on the other hand, favor spin exchange between neighboring sites on adjacent layers, thereby forming local interlayer pairs and destroying long-range magnetic order within the layers. The exact nature of the ground sate resulting from the Kondo proximity effect when the localized moments carry simultaneous charge degrees of freedom, i.e., finite U, is not well established. To gain an insight into the ground state in this parameter regime, we study the intra- and interlayer hopping

$$T_{ll'} = \langle c_{il\sigma}^{\dagger} c_{il'\sigma} \rangle \tag{5}$$

as well as short-range (near-neighbor) spin-spin correlation

$$C_{ll'}^{SS} = \langle S_{il}^{z} S_{il'}^{z} \rangle. \tag{6}$$

The results are shown in Fig. 3. With increasing V, intraplane nearest-neighbor hopping and spin-spin correlations decrease monotonically and eventually reach a constant value at $V_s \approx 4t$. The corresponding interplane observables, on the other hand, increase monotonically with V and saturate at V_s. Interestingly, the value of the saturation interlayer coupling V_s is found to be independent of the value of on-site coupling over a wide range $-0 \leqslant U \leqslant 10$ (data not shown here). Previous studies of a insulator-metal bilayer with *attractive* U has shown that V_s is a function of the density of fermions (assumed to be identical for both the metallic and the insulating layers). Unfortunately, the negative sign problem prevents us from extending our QMC study in this multilayer system with $U > 0$ to densities away from half-filling. As with the in-plane magnetic ordering, it is observed that the pairing amplitude as well as interlayer hopping are enhanced at the interfaces nearest to the boundaries where one of the layers has only one interface with a layer of the opposite type. In the bulk, where each layer has two interfaces with layers of the opposite type, the interlayer pairing/hopping and spin-spin correlations compete at the two interfaces and reduce the overall response. It would be interesting to investigate whether the surface-bulk response is reversed if each layer consists of more than one plane of each type, e.g., MM-II-MM-II-. . . geometry.

IV. SUMMARY

We have used DQMC simulations to study the magnetic properties of a system of alternating metallic and Mott insulating layers with variable interlayer hybridization V. Our results show clear evidence of induced magnetism in the metallic layers at small to intermediate values of V, in contrast to previous studies of multilayer systems with a single interface. The in-plane magnetism vanishes at a critical V_c due to the formation of interplane pairs via the Kondo proximity effect. The evolution of the density of states is studied across the entire range of V. Finally, the nature of the ground state at large values of interlayer hopping is investigated and its nature is explained in terms of short-range correlations.

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