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Kondo screening and Magnetism at Interfaces

A. Euverte[1], F. Hébert[1], G.G. Batrouni[1,2], S. Chiesa[3] and R.T. Scalettar[4]
[1]*INLN, Université de Nice-Sophia Antipolis, CNRS; 1361 route des Lucioles, 06560 Valbonne, France*
[2]*Institut Universitaire de France*
[3] *Department of Physics, College of William & Mary, Williamsburg, VA 23185, USA and*
[4]*Physics Department, University of California, Davis, California 95616, USA*

The nature of magnetic order and transport properties near surfaces is a topic of great current interest. Here we model metal-insulator interfaces with a multi-layer system governed by a tight-binding Hamiltonian in which the interaction is non-zero on one set of adjacent planes and zero on another. As the interface hybridization is tuned, magnetic and metallic properties undergo an evolution that reflects the competition between anti-ferromagnetism and (Kondo) singlet formation in a scenario similar to that occurring in heavy-fermion materials. For a few-layer system at intermediate hybridization, a Kondo insulating phase results where magnetic order and conductivity are suppressed in all layers. As more insulating layers are added, magnetic order is restored in all correlated layers except that at the interface. Residual signs of Kondo physics are however evident in the bulk as a substantial reduction of the order parameter in the 2-3 layers immediately adjacent to the interfacial one. We find no signature of long range magnetic order in the metallic layers.

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Sufficiently strong electronic correlations can cause the formation of an insulating phase at commensurate fillings. In general, there is a non-zero critical interaction strength required for this "Mott transition", so that two uncoupled bands with different degrees of correlation can coexist in metallic and insulating states. The behavior of spectral functions and magnetic and superconducting correlations, when interband hopping or interactions are turned on, is a challenging theoretical problem. The coupling could immediately force both bands to be in the same (metallic or insulating) phase, or coexistence might persist up to some critical degree of coupling [1, 2].

Closely related questions arise as clean interfaces between correlated materials become accessible [3]. Here the role of different orbitals is played by the multiple layers. It has been suggested that it might be possible to "engineer" specific forms of spectral functions at the interface by varying the materials partnered, as well as design other properties arising from electronic interactions [4–6]. Experimental realizations include tunable 2D electron gases in oxide ($SrTiO_3/LaAlO_3$) heterostructures, control of magnetoresistance at manganite interfaces [7], novel magnetic properties at boundaries between cuprate superconductors [8], and observation of magnetic proximity effect in Cu/CuO interfaces [9].

While the detailed chemistry of both multi-orbital and layered materials is complex, an interesting starting point for studying the qualitative properties of metal-insulator interfaces is provided by the multi-layer Hubbard Hamiltonian. In this model, electrons have both intralayer and interlayer hopping, as well as layer-dependent contact interactions. The parameter space is large and in this paper we focus on the simplest realization of the physics of a metal-insulator interface in which all hybridizations are chosen to be equal except the one at the interface; the corresponding Hamiltonian is

$$\begin{aligned}\hat{\mathcal{H}} \;=\; & -t\sum_{\langle ij\rangle,l,\sigma}(c^{\dagger}_{il\sigma}c_{jl\sigma}+\text{h.c.})-\mu\sum_{i,l,\sigma}n_{il\sigma} \\ & +\sum_{i,l}U_l(n_{il\uparrow}-1/2)(n_{il\downarrow}-1/2) \\ & -\sum_{i,\langle ll'\rangle,\sigma}t_{ll'}(c^{\dagger}_{il\sigma}c_{il'\sigma}+\text{h.c.}).\end{aligned} \qquad (1)$$

Here $c^{\dagger}_{i\,l\,\sigma}(c_{i\,l\,\sigma})$ are creation (destruction) operators for fermions of spin σ at site i in layer l. Each layer is an N-site square lattice with a contact interaction U_l chosen to be non zero, $U_l = U$, on "correlated" layers $l = 1, 2, 3, \cdots$, and zero for an additional set of "metallic" layers ($l = -1, -2, -3, \cdots$). Layers are arranged in order of increasing l so that $l = \pm 1$ label the layers at the interface. t and $t_{ll'}$ are the intra and interlayer nearest-neighbor hybridizations. $t_{ll'} = t$ except at the interface where it takes the value $t_{-1,1} = V$. We consider the case where $\mu = 0$ which, as a consequence of particle-hole symmetry, makes all layers half-filled, $\langle n_{il\sigma}\rangle = 0.5$. Recent studies on similar models have found induced magnetic order in the metal [10] and quasi-particle penetration in paramagnetic Mott insulators [11].

Questions that arise in connection with Hamiltonian (1) can be seen as extensions to those typically asked in the context of heavy-Fermion materials [12] and concern, at least at half-filling, the competition of magnetic order and screening of local moments by conduction electrons. Heavy-Fermion materials are modeled by Hamiltonian (1), a bilayer with $l = \pm 1$, or by its strong coupling limit, the Kondo-Heisenberg lattice, where charge fluctuations on the correlated layers are neglected. The fundamental issue we address here is how this competition is affected as the two-layer case crosses over to the 3-dimensional bulk-to-bulk interface.

At small and large interface hybridization, the system is adiabatically connected to, respectively, the $V = 0$ and $V = \infty$ limits. At small V the system is made up of magnetically ordered layers weakly coupled to a metal. At large V the central bilayer decouples and leaves the external layers either metallic ($l \leq -2$) or insulating, with anti-ferromagnetic long range order ($l \geq +2$).

Our results indicate that, for a four-layer system, two interacting and two metallic sheets, and at intermediate interfacial hybridization, there exists an intervening phase where loss of anti-ferromagnetic order is seen in both correlated layers, +1 and +2, despite the fact that the latter is not in direct contact with the metal. We found that the electronic structure of the metal is also profoundly affected and that the overall phase of the quad-layer can be characterized as a Kondo insulator. This is in contrast to our other finding when the interaction region becomes thicker than the metallic one: no loss of magnetic order is found in layers beyond the one immediately adjacent to the metal, regardless of the hybridization strength V. In this case, upon increasing V, a direct transition between the small and large V regimes results.

We addressed the physics of Hamiltonian (1) using determinant Quantum Monte Carlo (DQMC) [13], an exact, finite-T method for solving tight binding Hamiltonians on finite lattices. As we limit our calculations to the perfectly half-filled case, there is no sign problem at any temperature. Our results are averaged over several independent simulations, and the error bars correspond to the standard deviation of the mean. The imaginary-time step is set to $\Delta\tau = t/8$. We present results for the in-plane anti-ferromagnetic structure factor,

$$S_l^{\rm af} \equiv \frac{1}{3N}\sum_{i,j}(-1)^{i+j}\left[2\langle\sigma_{il}^x\sigma_{jl}^x\rangle + \langle\sigma_{il}^z\sigma_{jl}^z\rangle\right], \qquad (2)$$

where $\sigma_{il}^x = c_{il\uparrow}^\dagger c_{il\downarrow} + c_{il\downarrow}^\dagger c_{il\uparrow}$ and $\sigma_{il}^z = c_{il\uparrow}^\dagger c_{il\uparrow} - c_{il\downarrow}^\dagger c_{il\downarrow}$, and the local layer dependent spectral function $A_l(\omega)$, obtained by inverting the integral equation

$$G_l(\tau) = \int_{-\infty}^{+\infty} d\,\omega\,\frac{e^{-\omega\tau}}{1+e^{-\beta\omega}}\,A_l(\omega) \qquad (3)$$

via the maximum entropy method [14]. $G_l(\tau) = \sum_{i\sigma}\langle T\, c_{il\sigma}(\tau)\; c_{il\sigma}^\dagger(0)\,\rangle$ is the quantity directly obtainable by DQMC, and is averaged over 4 boundary conditions corresponding to setting the hopping at the boundary of each layer to $\pm t$ [15]. We also study the in-layer electrical conductivity σ_l which is extracted from the current-current correlation function

$$\Lambda_{xx,l}(k,\tau) = \sum_{i\in l} e^{ik\cdot i}\langle j_x(i,\tau)j_x(0,0)\rangle, \qquad (4)$$

with $j_x(r,0) = it\sum_\sigma(c_{r+x,\sigma}^\dagger c_{r,\sigma} - c_{r+x,\sigma}^\dagger c_{r,\sigma})$. We focused on the intralayer contribution to $\Lambda_{xx,l}$ only, assuming that this correctly characterizes the conductive property of each layer. σ_l is extracted using the approximate form of the fluctuation-dissipation relation, valid at large β and first discussed in [16],

$$\Lambda_{xx,l}(k=0,\tau=\beta/2) = \pi\sigma_l/\beta^2. \qquad (5)$$

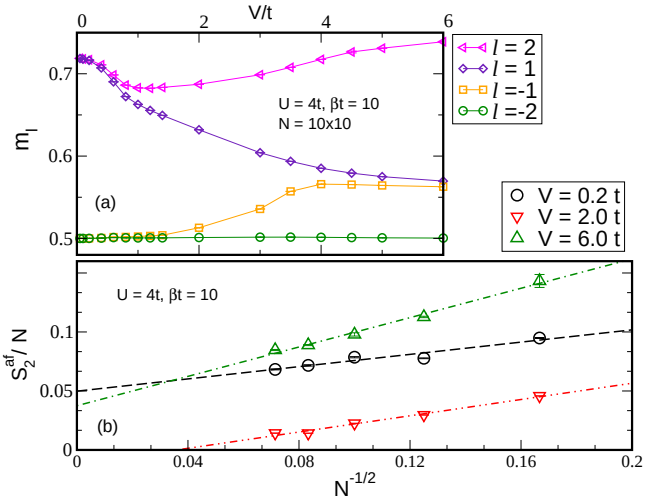

FIG. 1: (color online) (a) V-dependence of local moments, $m_l = \sum_i\langle(\sigma_{il}^z)^2\rangle/N$, on each layer, when two metallic layers are coupled to two correlated ones. (b) Finite size scaling of in-plane structure factor $S_2^{\rm af}$ of the correlated layer ($l = 2$) farthest from the interface. For small V, there is long range order in the thermodynamic limit which vanishes for intermediate V and is recovered for large V. $S_2^{\rm af}$ reaches its ground state value at $\beta t = 10$.

To gain initial quantitative understanding of the evolution of magnetic properties as V increases, we show, in the top panel of Fig. 1, the evolution of local moments in a system of two metallic and two interacting sheets. There are three regimes, most clearly evidenced by the behavior of the metallic layer at the interface. At $V \lesssim t$ local moments on layer -1 are essentially identical to those of a non-interacting system. In $t \lesssim V \lesssim 4t$ the moments monotonically increase and they saturate at $V \simeq 4t$. The evolution of the other layers follows naturally, with layer +1 merging with -1 at large V in a phase that can be best characterized as a band insulator made of weakly interacting dimers. Layer +2 has the only non-monotonic evolution: magnetism is first suppressed and then revived as the central dimer phase gets increasingly stabilized.

We then use finite size scaling on $S_2^{\rm af}$ to investigate whether order in layer +2 is lost in the regime where the moments are most suppressed. As shown in the lower panel of Fig. 1, $S_2^{\rm af}$ scales to a nonzero value for both small and large interface hopping, V, when plotted against the inverse linear system size $1/\sqrt{N}$. For both these regimes, there is long range order in the ground state in the thermodynamic limit [17] as expected from

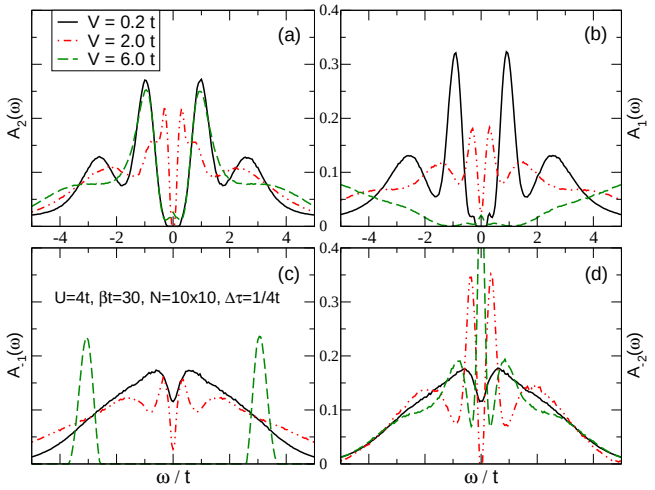

FIG. 2: (color online) (a) Spectral function $A_2(\omega)$ in correlated layer $l = 2$. The Slater gap present at small V due to AF order vanishes at intermediate V and reappears at larger V. (b) In correlated layer $l = 1$, $A_1(\omega)$ resembles $A_2(\omega)$ but unlike $l = 2$ the large V behavior is a broader gap associated with the singlet energy scale. In both non-interacting layers $A_{-1}(\omega)$ (c) and $A_{-2}(\omega)$ (d) a gap opens as V increases. At large V, layer -2 recovers metallic properties while the singlet gap is visible in layer -1.

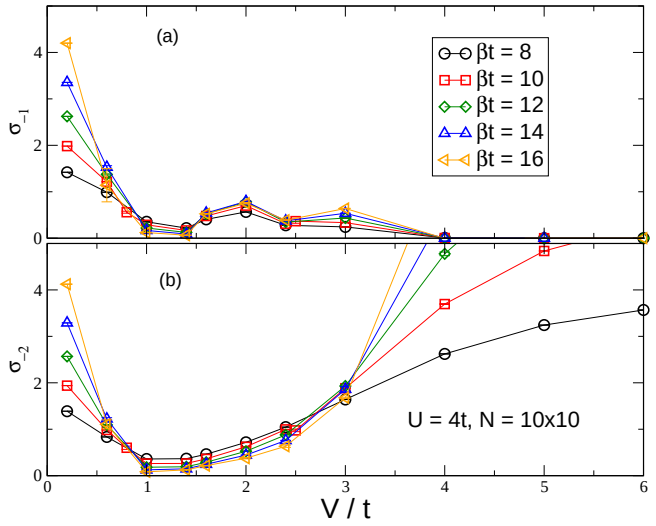

FIG. 3: (color online) (a) In-plane conductivity σ_{-1} in the metallic layer $l = -1$ as a function of V, at several inverse temperature β values. At intermediate V is small but nonzero, but vanishes at $V \geq 4t$ due to dimer formation. (b) In-plane conductivity σ_{-2}. This noninteracting layer becomes insulating for intermediate hybridization, $t \leq V \lesssim 3t$ and then recovers when the pairs are fully formed and pinned at the interface.

the behavior of the local moments. However, in the intermediate regime (starting at $V \geq t$, in rough correspondence to where loss of anti-ferromagnetic order happens in the periodic Anderson model [18] and the bilayer Hubbard model [19]) loss of magnetic order is clearly observed on layer $+2$ as well.

The most likely candidate mechanism for such loss of order involves Kondo screening in both interacting layers. As the formation of a resonance in the single particle spectral density is one of the hallmark of such process, we plot, in Fig.2, the layer dependent spectral density at $T = t/30$. We found that, at $V = 2t$, both interacting layers are characterized by the presence of a Kondo resonance. Due to the fact that we are focusing on a half-filled system, the resonance is split as typically happens for Kondo insulators.

We can gain further insight into the nature of this intermediate phase by looking at the behavior of the non-interacting layers. Figure 3 shows the conductivity in layers $l = -1$ and $l = -2$. At small hybridization, before the loss of magnetic order, the conductivity increases as T is lowered, showing these two sheets to be metallic. In the intermediate regime $t \lesssim V \lesssim 3t$, the conductivity is strongly suppressed in *both* layers, and our inverse temperature results suggest that these layers become insulating around $V = t$ in correspondence with the loss of magnetic order.

From these results we can draw a few significant conclusions. First, that there is a Kondo proximity effect, as already observed using dynamical mean-field theory on a similar model [11]. The novelty of our finding resides in our treatment of non-local correlation. It allows for a proper description of the competition between magnetic order and Kondo screening and shows the latter to be effective in destroying order even on layers not directly coupled to the metal. Furthermore, states from both metallic layers participate in the screening of local moments as best evidenced by the drop in conductivity. This situation is reminiscent of a long-range resonating valence-bond state although it is unclear whether such a description remains meaningful in the present context of itinerant electrons.

The other kind of proximity effect that is expected in such systems is due to the presence of the magnetically ordered layers at smaller values of V. This would seem a likely scenario, especially since the metallic phase lives on layers with nested Fermi surfaces with infinite, $T = 0$, anti-ferromagnetic susceptibility. Our calculations, however, do not find any significant penetration of magnetic order in the metallic layers. Although we cannot exclude that an extremely small order parameter might develop at low T for some range of V, our finding suggests that such an order would not survive the generic scenario of a system with finite susceptibility. This result appears at odds with recent experiments [9] finding evidence for an anti-ferromagnetic proximity effect.

We now consider the question of how the competition of Kondo screening and magnetic order is affected when additional interacting layers $l = 3, 4, \cdots$ are present. The

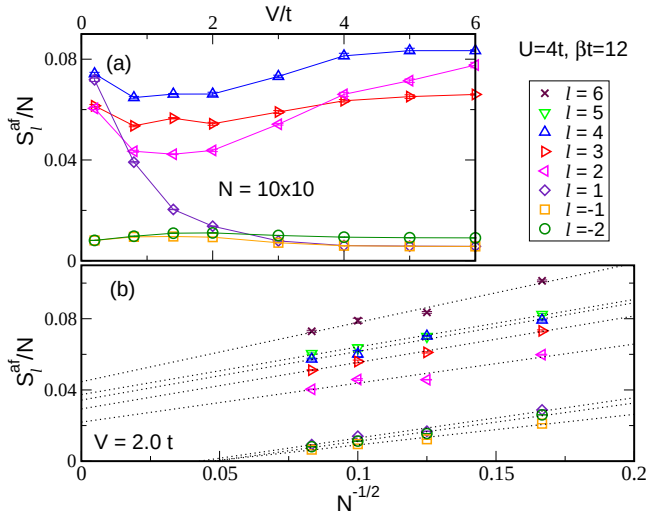

FIG. 4: (color online) (a) In-plane structure factors $S_l^{\rm af}$ as functions of V for a system of four correlated layers coupled to two metallic ones. At intermediate V antiferromagnetic order is suppressed in every correlated layer. At large V the order recovers in every correlated layer but the one at the interface. (b) Finite-size scaling of the in-plane structure factors $S_l^{\rm af}$ for a system of six correlated layers coupled to two metallic ones, at $V = 2t$. A systematic reduction of spin correlations is evident as the metallic interface is approached from the correlated side. Long range magnetic order is completely destroyed in the correlated layer at the junction.

resulting system can be thought of describing the interface between a thin metallic film and a bulk antiferromagnetic Mott insulator or as a heterostructure where the insulating domains are substantially thicker than the metallic ones. A scan of $S_l^{\rm af}$ for different values of V on a 6-layer cluster with 600 sites (top panel in Fig. 4) indicates that the most likely parameter regime to observe loss of magnetic order in layer 2 is for $V/t \in [1, 2]$.

However, for $V = 2t$ (lower panel of Fig. 4), we found that, while $S_1^{\rm af}$ does not extrapolate to a non-zero value, the same does not happen for layers located deeper into the interacting material *i.e.* $l = 2, 3, 4, 5, 6$ are all antiferromagnetically ordered. This revival of magnetic long-range order on layer +2 can be interpreted as a magnetic proximity effect exerted by a bulk anti-ferromagnet on those correlated layers subject to Kondo screening. Note that a clear suppression of the order parameter is still observed for $l = 2$ and several layers deeper in indicating coexistence of Kondo screening and magnetism (the anomalously large value at $l = 6$ is a known phenomenon where surface magnetic correlations are larger than the bulk and has been widely explored experimentally [20]).

In conclusion, we have presented results on a model of metal-insulator interface, the multilayer Hubbard Hamiltonian. Even within this simplified tight-binding model, there are many possible choices of the intralayer and interlayer hoppings. The dominant feature of the coupling of the metal and strongly interacting material is a suppression of magnetic order on the correlated side. We did not observe the converse phenomenon, namely a significant penetration of magnetism into the metal, as has been noted in [10]. It is possible this difference arises from the lower value of the on-site interaction, $U/t = 4$, used here, compared to $U/t \approx 17$ in [10]. Such large couplings are difficult to treat in DQMC. The on-site interactions studied here are relevant to the range of fitted values for a number of interesting materials, e.g. CuO [21].

We showed that for thin insulating layers the Kondo effect embraces correlated and metallic layers that are not in direct contact with each other. Although such an extended Kondo insulating phase does not require any fine tuning, it is not a phase that permeates a large fraction of the "phase space" for such systems. Here we have shown, for instance, that forming Kondo singlets across multiple layers is a process that can be defeated by magnetic proximity effect of layers farther from the interface.

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