Orbital-selective Mott transition and heavy fermion behavior in a bilayer Hubbard model for $^3$He

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Inspired by recent experiments on $^3$He films between one and two atoms thick, we consider a bilayer Hubbard model on a triangular lattice. Our results are obtained in the framework of a cluster dynamical mean-field calculation with a quantum Monte Carlo impurity solver. For appropriate model parameters, we observe an enhancement of the effective mass as the first layer approaches integer filling and the second remains only partially filled. At finite temperatures, this increase of the effective mass—or, equivalently, the decrease of the coherence temperature—leads to a crossover to a state where the first layer fermions localize, drop out of the Luttinger volume, and generate essentially free local moments. This finite temperature behavior is shown to be robust against the cluster size above some critical temperature. The zero temperature phase diagram, however, depends on the cluster topology. In particular, for clusters with an even number of unit cells, the growth of the effective mass is cut off by a first-order, orbital-selective Mott transition.

I. INTRODUCTION

The solidification of $^3$He monolayers has been interpreted as a density-driven Mott transition in which the effective mass diverges. Below the critical density, the system is metallic, nearly localized Fermi liquid; beyond the critical density, it is a solid, the magnetic properties of which are dominated by antiferromagnetic two-body exchange processes. It is now possible to realize bilayers of $^3$He (atop a frozen $^4$He substrate, itself adsorbed onto graphite) with the special property that the second layer begins to form before the first has solidified. Since the first layer is close to a Mott transition, the $^3$He fermions in this layer are slow (i.e. heavy), whereas those in the second layer are fast. This combination of fast and slow dynamics—corresponding to wide and narrow fermion conduction bands—is completely analogous to the situation in electronic heavy fermion materials, albeit without the complication of crystal field and spin orbit effects.

According to this picture, one expects, prior to solidification of the first layer, an enhanced effective mass and a Luttinger volume that counts both the first- and second-layer populations. Moreover, one naively anticipates that the first-layer effective mass to diverge, in coincidence with the solidification of the first layer, an enhanced effective mass and a Luttinger volume. In experiment, the effective mass is indeed observed to increase as a function of the total $^3$He concentration, but its growth is interrupted by an intervening phase. The fact that this phase is ferromagnetic indicates that three-body exchange processes come to dominate in the solid phase of the first layer.

The motivation of this article is to consider a simple lattice model that goes a good way towards reproducing the essential features of the above experimental situation. As shown in Figs. 1(a) and 1(b), we adopt a stacking of billiard balls modeling of bilayer $^3$He on a triangular lattice defined by $\mathbf{a}_1 = (1/2, \sqrt{3}/2, 0)$ and $\mathbf{a}_2 = (1, 0, 0)$. Each unit cell accounts for two $^3$He positions, $r_f = 0$ and $r_e = \frac{1}{2}\mathbf{a}_1 - \frac{1}{2}\mathbf{a}_2 + (0, 0, a_3)$, measured relative to the lattice. This geometry presupposes a particular stacking arrangement for the second $^3$He layer.

Our model can be viewed as a honeycomb lattice whose
equivariant sites (corresponding to $^3$He positions in the upper and lower layers) are populated by two species of fermion, which we label $c$ and $f$. The tight-binding parameters include a nearest-neighbor (interlayer) hopping $V$ and next-nearest-neighbor (intralayer) hoppings $t_c$ and $t_f$. See Fig. 1(c). With the inclusion of onsite Coulomb repulsion terms, the Hamiltonian reads

$$H = \sum_{k,\sigma} \left( \varepsilon_c(k) - \mu \right) c_{k,\sigma}^\dagger c_{k,\sigma} + \sum_{i} \left( \hat{n}_{c,i} - 1 \right)^2 + U_c \sum_{i} \left( \hat{n}_{f,i} - 1 \right)^2 + U_f \sum_{i} \left( \hat{n}_{f,i} - 1 \right)^2. \quad (1)$$

Here, the mixing element $V(k) = V(3 + 2\gamma_k)^{1/2}$ and the dispersion $\varepsilon_c(k) = -2t_c\gamma_k + \varepsilon_c^0$ are expressed in terms of the connection $\gamma_k = \cos(k \cdot a_1) + \cos(k \cdot a_2) + \cos(k \cdot (a_2 - a_1))$ of the underlying Bravais lattice. The operator $\hat{n}_{c,i} = \sum_{\sigma} c_{i,\sigma}^\dagger c_{i,\sigma}$ is the local $^3$He density in the upper layer. Similar definitions hold for $\varepsilon_f(k)$ and $\hat{n}_{f,i}$.

Except for the complication of the layer stacking (and the resulting $k$-dependent hybridization), this bilayer Hubbard model reduces to the Periodic Anderson Model as $t_f \to 0$, a limit in which the bare mass of the $f$ fermions diverges. Similar models have been considered for the description of bilayer $^3$He in Refs. 8 and 9 within a slave boson mean-field calculation. Here we go a significant step further and perform calculations within the cellular dynamical mean field theory (CDMFT) approximation (Sec. II). Our strategy is to systematically investigate the model of Eq. (1) as a function of the cluster size. In Sec. III, we will see that clusters with an odd number of unit cells have a radically different low-energy behavior than those with an even number. Given this situation, the extrapolation to the large cluster size limit is delicate and is relegated to the conclusions in Sec. IV. Part of this work has already appeared in a preprint.12

II. CELLULAR DYNAMICAL MEAN FIELD THEORY

By construction, the CDMFT approach exactly accounts for the temporal fluctuations at each site and thereby captures the physics of the local moments—both their formation and their screening via the Kondo effect. But the spatial fluctuations extend only over the simulation cluster; insofar as the true correlation length scale exceeds the linear size of the cluster, the results will suffer from finite size effects. To mitigate this, we have considered various cluster topologies ranging from one unit cell (a single $c$ and $f$ site) to four unit cells as defined in Fig. 1(c). For a given supercell, the resulting single-particle Green function, $G(K, i\omega_n)$, is a $2N_u \times 2N_u$ matrix with crystal momentum $K$ in the Brillouin zone of the supercell lattice. The CDMFT calculation involves neglecting momentum conservation and thereby obtaining a $K$-independent self-energy $\Sigma(i\omega_n)$. This quantity is extracted from a cluster of $N_u$ unit cells embedded in a dynamical mean field that is determined self-consistently. We have solved this cluster problem using a standard Hirsch-Fye approach and have symmetrized the cluster Green function to obtain the corresponding quantity on the lattice:

$$G(K, i\omega_n)_{\mu,\nu} = \frac{1}{N_u} \sum_{\alpha,\beta} \epsilon^{K}(x_{\alpha} - x_{\beta})G(K, i\omega_n)_{(\mu,\alpha), (\nu,\beta)} \quad (2)$$

Here $x_{\alpha}$ denotes the unit cell positions within the supercell, $\mu$ and $\nu$ run over the $c$ and $f$ orbitals within each unit cell, and $K$ and $K$ differ by a reciprocal lattice vector of the supercell Bravais lattice. The rotation to real frequencies was carried out with a stochastic analytical continuation technique.12

III. RESULTS

We consider the following model parameters: $t_c = t_f = t$, $U_c/t = U_f/t = 12$, $V/t = 1/2$, $\varepsilon_c^0/t = 3$, and $\varepsilon_f^0/t = 0$. We have chosen large values of $U_c$ and $U_f$ to reflect the contact repulsion of the $^3$He atoms and to guarantee that each single layer is well within the Mott insulating phase at half-band filling.10 These values of the Hubbard interaction lead to low double occupancy, thus generating local moments. The difference $\varepsilon_c^0 - \varepsilon_f^0 > 0$ is a crude accounting for the van der Waals forces (both $^3$He–$^3$He and $^3$He–$^4$He) that preferentially fill the first layer.

A. Layer densities

The generic Mott insulating state is characterized by a density $\langle \hat{n} \rangle = 1$ and a vanishing charge susceptibility: i.e., $\chi_{ch} = \partial \langle \hat{n} \rangle / \partial \mu = 0$, where $\mu$ denotes the chemical potential. Figure 2 plots the layer-resolved densities $\langle \hat{n}_c \rangle$ and $\langle \hat{n}_f \rangle$ as a function of the chemical potential, which controls the overall $^3$He concentration. For both the odd and even cluster sizes, $\langle \hat{n}_f \rangle$ shows a plateau feature centered around $\langle \hat{n}_f \rangle = 1$, whereas $\langle \hat{n}_c \rangle$ grows smoothly. In contrast to the Mott insulating state, $\chi_{ch}^f = \partial \langle \hat{n}_f \rangle / \partial \mu$ never vanishes. Hence, charge fluctuations between the layers are allowed and the simple picture of a complete decoupling of the layers never holds. Although the plateau feature is common to all cluster sizes, the data show distinct odd-even effects. For $N_u = 1$ and $N_u = 3$, $\langle \hat{n}_f \rangle$ is a continuous function of the chemical potential for all temperatures considered. In contrast, for the even clusters, $N_u = 2$ and $N_u = 4$, a discontinuity in $\langle \hat{n}_f \rangle$ emerges below a critical temperature $T_c$ and at a critical chemical potential. For $N_u = 4$, a robust discontinuity is present at $T_c \simeq t/20$, whereas for $N_u = 2$ this feature already appears at $T_c \simeq t/15$. Since $\langle \hat{n}_c + \hat{n}_f \rangle = 0$, $\partial F / \partial \mu$, where $F$ is the free energy, the jump in the total fermionic density signals a density-driven first-order transition. In a canonical ensemble,
FIG. 2. The average occupation number is plotted for the upper- (top row) and lower-layer (bottom row) fermions as a function of chemical potential. Results are reported for both odd- (leftmost two columns) and even-numbered (rightmost two columns) clusters. Solid lines connect data sets with a common temperature. Vertical dashed lines mark the location of the emerging low-temperature discontinuity in $\langle \hat{n}_f \rangle$.

states with total density lying within the jump are phase separated.

B. First-layer effective mass and low temperature spectral functions

We can estimate the $f$ fermion’s effective mass as a function of the chemical potential by considering its cluster-averaged self-energy, $\Sigma_f(i\omega_m) = \frac{1}{N_u} \sum_{\alpha=1}^{N_u} \Sigma_{(f,\alpha),(f,\alpha)}(i\omega_m)$, and extracting the quantity

$$\frac{t}{T_0} = \frac{m^*}{m} \propto Z^{-1} = 1 - \frac{\text{Im} \Sigma_f(i\omega_m = i\pi T)}{\pi T}. \quad (3)$$

This estimate of the effective mass (or, equally, of the inverse of the coherence temperature $T_0$) is valid provided that the real space dependence of the self-energy is small and that the temperature $T$ is extrapolated to zero. Data on the $N_u = 4$ cluster presented in Ref. 11 shows that for $\mu < \mu_c$, the assumption of a local self-energy is valid.

At $N_u = 1$ (see Fig. 3), the effective mass increases as a function of chemical potential. This effect is also evident in the evolution of single particle spectral functions,

$$A(k, \omega) = -\text{Im Tr} \, G(k, \omega + i0^+). \quad (4)$$

FIG. 3. The inverse of quantity defined in Eq. (3), which is proportional to the coherence temperature $T_0$, inverse effective mass, and quasiparticle residue, is plotted as a function of chemical potential.

plotted in Fig. 4. As exemplified by the data set at $\mu/t = -2.5$ [Fig. 4(a)], the low-energy coherent features
For this cluster smallest size, only the delocalized heavy band with dominant Hubbard band at ω of Fig. 4(b) exhibits typical heavy fermion character: the Fermi energy that hybridizes with a light conduction band. At N_u = 3, the coherence temperature (see Fig. 3) is reduced with respect to the N_u = 1 case but nevertheless shows a similar overall behavior: a rapid decrease as a function of chemical potential followed by saturation at a lower value than for the N_u = 1 case. Within the accessible temperature range of the N_u = 3 cluster, the single particle spectral function shows the same features as for the N_u = 1 case.

For the even site clusters, the initial decrease of the coherence temperature is cut off by the first-order transition. Far below μ_c, as exemplified by μ/t = −2.5, the single particle spectral function is very similar to that observed on the N_u = 1 cluster [cf. Figs. 4(a) and 4(b)]. With increasing chemical potential, the effective mass of the f band grows, and beyond μ_c the f band drops out of the low-energy physics altogether. This can be understood at the static mean field level by a conventional slave-boson theory in competition with local singlet formation in the first layer. The transition is signaled by the appearance of an anomalous expectation value Δ_u ^ {2} ∼ (t_f ^2/U_f) ∑σ ⟨f_i^σ f_j^σ⟩. The inset of Fig. 5(c) shows the band structure that results when this singlet order parameter breaks down the original lattice symmetry to that of the N_u = 2 supercell.

C. Spin susceptibilities and correlations

We can extract from the cluster the local spin susceptibility as defined by

\[ \chi_f(iΩ_m) = \frac{1}{N_u} \sum_i \int_0^β dτ e^{iΩ_mτ} \langle S^f_i(τ) \cdot S^f_i(0) \rangle. \] (5)

A Fermi liquid below its Fermi temperature is Pauli paramagnetic, and hence \( \chi_f(iΩ_m = 0) \) is constant. On the other hand, a local moment is characterized by a Curie-Weiss law, \( \chi_f(iΩ_m = 0) \propto 1/(T + Θ) \) at temperatures \( T \gg Θ \). As is apparent in Fig. 6, \( \chi_f(iΩ_m = 0) \) always exhibits a smooth crossover from the high-temperature Curie-Weiss to the low-temperature Pauli behavior, irrespective of the lattice topology. For the odd lattice sizes, the crossover point tracks the coherence temperature. The same holds for the even lattice at \( μ < μ_c \).

It is worth emphasizing that this qualitative change in magnetic response pinned to the coherence temperature (also denoted by \( T_0 \) in Ref 7) has been observed in the \(^3\)He bilayer experiment. Hence, at high temperatures, a local moment generated by the Hubbard interaction is present. The screening of this local moment, or in other words the quenching of its entropy, is at the origin of the different behavior between the odd- and even-numbered lattices.

N_u = 1. For this cluster smallest size, only the delocalized c fermions are available to screening the local moment. This is precisely the Kondo effect, and one can view the heavy fermion paramagnetic state as originating...
from the coherent, Bloch-like superposition of individual Kondo screening clouds. Within a periodic Anderson model, this screening of the local moment is linked to a delocalization of the $f$ fermion. Hence, above $T_0$, when screening is absent, we expect the $f$-quasiparticle band to drop out of the low-energy physics. This is evident from Figs. 4(b) and 4(c) upon comparison of the high- and low-temperature spectral functions at $\mu = 0$.

$N_u = 3$. This cluster size shows behavior very similar to that of the $N_u = 1$ system, albeit with a lower coherence temperature. At values of the chemical potential where the $f$ layer is approximately half filled, a magnetic superexchange interaction $J = 4t^2/U_f$ is dynamically generated and the spin degrees of freedom on the first layer are described by a Heisenberg model on a three-

FIG. 5. The amplitude of the single particle spectral function for the $N_u = 4$ cluster. The inset in panel (c) shows the single $c$-only band completely decoupled from the gapped, nearly flat band of the singlet-bound $f$ fermions. The mean-field calculations were carried out on the $N_u = 2$ cluster and the QMC on the $N_u = 4$ systems.

FIG. 6. Temperature dependence of the inverse local spin susceptibility for the (a) $N_u = 3$ and (b) $N_u = 4$ clusters.

FIG. 7. (a) Normalized nearest-neighbor $f$-fermion spin-spin correlations for on the $N_u = 3$ cluster. On a three-site ring, the value of this quantity for the Heisenberg model is given by $-1/3$. (b) Local dynamical spin structure factor.
site lattice. The ground state is fourfold degenerate corresponding to a spin-1/2 degree of freedom with either positive or negative chirality. As in the \( N_u = 1 \) case, the only way to quench this residual entropy is via Kondo screening by the \( c \) fermions. To confirm this interpretation, we have computed the nearest-neighbor spin-spin correlations on the first layer, \( S_f(r) = \langle S_i^f \cdot S_{i+r}^f \rangle \). Comparison with the Heisenberg result is best achieved by normalizing the QMC data by the magnitude of the local moment, \( S_f(r = 0) \). As shown in Fig. 2(a), the nearest-neighbor antiferromagnetic spin-spin correlations are considerable. At high temperatures the energy scale at which they decay is set by the superexchange coupling \( J/t = 1/3 \); and at \( \mu/t = -0.75 \), where we observe a Curie-Weiss law down to our lowest temperature, they compare favorably to the Heisenberg ground state result: \( S_f(\mathbf{r})/S_f(0) = -1/3 \). Fig. 7(b) plots the dynamical local spin structure factor,

\[
S_f(\omega) = \text{Im} \frac{\chi_f(\omega)}{1 - e^{-\beta\omega}}, \tag{6}
\]

at \( \beta t = 25 \) and as a function of the chemical potential. As mentioned above, at \( \mu/t = -0.75 \) the residual entropy is not quenched. Consequently, a low frequency sharp feature in \( S_f(\omega) \) marks the spin degenerate ground state of the three-site, half-filled Hubbard model. A feature at \( \omega/t \approx 0.5 \) corresponds to the first spin excitation, which for the three-site Hubbard model at \( U_f/t = 12 \) takes the value \( \Delta_{\text{sp}}/t = 0.49 \). As we decrease the chemical potential from \( \mu/t = -0.75 \) to \( \mu/t = -1.5 \), the weight in the high-energy feature remains approximately constant, but the sharp low-energy feature decreases in intensity and is shifted to slightly higher energies. This screening of the residual entropy by the \( c \) fermions competes with the nearly localized antiferromagnetic fluctuations in the first layer and is at the origin of the upturn in \( S_f(\mathbf{a}_1)/S_f(0) \) (see Fig. 2) at low temperatures.

Hence, at \( N_u = 3 \) spin correlations between the \( f \) fermions quench part of the entropy associated with the formation of the local moments. The residual entropy is Kondo screened by the \( c \) fermions, and in comparison to the \( N_u = 1 \) case leads to a suppressed coherence temperature.

\( N_u = 2, N_u = 4 \) The even site clusters show a band-selective Mott transition and a low-energy decoupling of the first and second layers. As in the \( N_u = 3 \) case, we can consider the effective Heisenberg model on the first layer. For even cluster sizes the ground state is unique and is spin singlet. The first-order transition we observed in Fig. 2 arises from competing screening mechanisms of the local moments generated by the nearly localized \( f \) fermions. On the one hand, the local moments can be Kondo screened by the light \( c \) fermions, thereby generating heavy fermion behavior. On the other hand, they can form (among themselves) a spin singlet state entirely in the first layer. The \textit{gapping} of the spin and charge degrees of freedom of the \( f \) quasiparticles at \( \mu > \mu_c \) allows for the decoupling of \( f \) and \( c \) quasiparticles: a \( f \) quasiparticle at the Fermi level cannot scatter off a \( f \) quasiparticle due to the absence of phase space. To support the picture of a sudden change in the screening mechanism, we plot in Fig. 8 intra- and interlayer equal-time spin-spin correlations for the \( N_u = 4 \) cluster size. At the critical chemical potential, we observe a \textit{sudden} growth of the antiferromagnetic correlations between nearest-neighbor \( f \) fermions and a decrease in the intracell \( c-f \) spin-spin correlations. Figure 8 also shows the local dynamical spin structure factor. One observes a depletion of spectral weight at low energies on both sides of the transition and a considerable sharpening of the line shape in the band-selective Mott insulating state. At \( \mu < \mu_c \), we can interpret the data within an itinerant fermion picture where the mass enhancement prior to the band-selective Mott transition is taken into account by a renormalization of the hybridization \( V \) and hopping \( t \) as in a slave boson approach. Following this modeling, the peak position in \( S_f(\omega) \) is expected to track the coherence temperature or, equivalently, the inverse effective mass. An explicit comparison of those quantities is provided in Ref. 11. At \( \mu > \mu_c, S_f(\omega) \) should be interpreted within a localized \( f \) fermion picture, in which case the peak position is a measure of the excitation energy required to break the singlet state of the \( f \) fermions. On a four-site Hubbard cluster, corresponding to the \( f \) layer in the \( N_u = 4 \) case, this quantity is given by 0.214\( t \) and compares favorably to the data in Fig. 8.

IV. CONCLUSIONS

Our calculations are best summarized by the phase diagrams plotted in Fig. 9. At high temperatures \( (T > T_c) \)
spin susceptibility. In the bilayer the high temperature phase and results in a Curie-Weiss local moments. The local moment is the key feature of heavy fermions. As the first layer approaches half band-filling the results are independent of the cluster size and topology. In the framework of Ref. 20 where the f fermions interact solely through the RKKY interaction, and in agreement with our numerical simulations, it is conjectured that for even site lattices, $T_c$ is set by the small FS magnitude of the Kondo screening and $\mu/t$ where the Kondo coupling between the $f$ fermions is generated by the hopping matrix element $t_f$, which again in the local moment regime, the Kondo coupling between the layers has been discussed in Ref. 16. On the other hand, if no symmetries are broken such that a spin liquid state is realized on the first layer, fractionalized Fermi liquids as proposed in Refs. 18 and 19 could be realized.

It is interesting to recast our results in terms of the $Q$-$K$ phase diagram for heavy fermions proposed by Coleman and Nevidomskyy.23 Here, $K$ corresponds to the magnitude of the Kondo screening and $Q$ is a measure of frustration between the $f$ fermions. For our model in the local moment regime, the Kondo coupling between the two layers is dynamically generated starting from second-order perturbation theory in the hybridization. The frustration between the $f$ fermions is generated by the hopping matrix element $t_f$, which again in the local moment regime leads to a superexchange interaction between the $f$ fermions. In the framework of Ref. 20 and in agreement with our numerical simulations, it is the frustration between the $f$ fermions that drives the Kondo breakdown or band-selective Mott transition. For a recent review in this domain, we refer the reader to Ref. 21. This is in contrast to the Kondo lattice model, where the $f$ fermions interact solely through the RKKY interaction, and no Kondo breakdown is observed in cluster simulations.21

Let us finally return to the bilayer $^3$He experiment. Heavy fermion character is clearly seen by the increase of the effective mass (or decrease of the coherence tem-
temperature). Furthermore, and as seen in our calculations, \( T_0 \) marks the crossover between a Curie-Weiss and Pauli behavior of the spin susceptibility. The \( Q \) or competing interaction which localizes the \( f \) fermions to the first layer are the three-body exchange processes. These processes, which in solid \(^3\)He can dominate the two-body antiferromagnetic exchange, lead to the observed ferromagnetic behavior. Given this interpretation of the experiment, an extremely important issue would be to pin down the experimental value of the Weiss constant. Above \( T_0 \) it should be positive and essentially track the Kondo scale. The transition to the ferromagnetic state should be accompanied by a vanishing and subsequently negative value of the Weiss constant.

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