Emergent Coherent Lattice Behavior in Kondo Nanosystems

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How many magnetic moments periodically arranged on a metallic surface are needed to generate a coherent Kondo lattice behavior? We investigate this fundamental issue within the particle-hole symmetric Kondo lattice model using quantum Monte Carlo simulations. Extra magnetic atoms forming closed shells around the initial impurity induce a fast splitting of the Kondo resonance at the inner shells which signals the formation of composite heavy-fermion bands. The onset of the hybridization gap matches well the enhancement of antiferromagnetic spin correlations in the plane perpendicular to the applied magnetic field, a genuine feature of the coherent Kondo lattice. In contrast, the outermost shell remains dominated by a local Kondo physics with spectral features resembling the simple-impurity behavior.

In the realm of condensed matter physics, the theory of wave propagation in periodic structures—the Bloch theorem [1]—forms the basis for calculating electronic band structure of solids. It is also a cornerstone for understanding low-temperature Fermi-liquid-like \( \propto T^2 \) transport properties of heavy-fermion (HF) metals which arise from a coherent Kondo screening of periodically arranged \( f \)-shell moments by conduction electrons [2]. The quantum entanglement between localized \( f \)-moments and mobile conduction electrons leads to the enlargement of the Fermi surface which determines, below the coherence scale \( T_{coh} \), transport and thermodynamic properties of HF liquids. A coherent Kondo lattice behavior differs markedly from the single-impurity Kondo physics with a logarithmic increase, below the Kondo temperature \( T_K \), of spin-flip scattering for conduction electrons [3]. Elucidating the crossover between both regimes [4–14] as well as identifying the energy scale \( T_{coh} \) associated with the formation of the coherent HF state is a long-standing issue [15–27].

Starting from an incoherent dilute limit of Kondo impurities one possibility to study how the lattice effects come into play is to increase the concentration of magnetic ions [28–35]. However, the electronic structure of bulk HF materials is essentially three dimensional and one may wonder how the coherence phenomena are affected either by reduced dimensionality or in spatially restricted geometries where both quantum fluctuations and correlations effect are enhanced [36, 37]. In this respect, the experimental realization of artificial \( f \)-electron superlattices [38] has opened a new avenue to investigate the onset of coherence in a two-dimensional (2D) regime followed up by theoretical studies [39].

In recent years, new insight into the Kondo physics at the nanoscale has come from scanning tunneling microscopy (STM) [40, 41]. It allows one to probe Kondo screening at a single magnetic adatom [42, 43], composite nature of the HF quasiparticles in a lattice situation [44–47] and also to image a mutual RKKY interaction between magnetic impurities mediated by conduction electrons as a function of the interatomic distances [48]. The possibility for a systematic and controlled study of the competition between different energy scales in Kondo nanostructures resulted in a resurgence of interest in the interplay between the Kondo effect and magnetic RKKY correlations in adatom dimers [49–52], trimers [53], and multiple impurities [54–56].

Given the distinct difference between a single-impurity Kondo physics and the coherent Kondo lattice behavior, it is natural to ask how many magnetic moments periodically arranged on a metallic surface are needed to resolve a crossover between both regimes? In this paper, we address this question on the basis of the Kondo lattice model (KLM) at half-filling [57],

\[
H_{KLM} = -t \sum_{\langle i,j \rangle, \sigma} c_{i,\sigma}^\dagger c_{j,\sigma} + \sum_i J_i \mathbf{S}_i^F \cdot \mathbf{S}_i^L,
\]

where \( \mathbf{S}_i^F = \frac{1}{2} \sum_{\sigma,\sigma'} c_{i,\sigma}^\dagger \sigma_{\alpha,\sigma'} c_{i,\sigma'} \) are spin operators of conduction electrons and \( \mathbf{S}_i^L = \frac{1}{2} \sum_{\sigma,\sigma'} f_{i,\sigma}^\dagger \sigma_{\alpha,\sigma'} f_{i,\sigma'} \) are localized spins with \( \sigma \) being the Pauli matrices. The Hamiltonian (1) describes localized spin 1/2 magnetic moments coupled via site-dependent antiferromagnetic (AF) exchange interaction \( J_i \) to conduction electrons on a square lattice. By switching on and off the values of \( J_i \) we can investigate a crossover from the zero-dimensional Kondo effect to the 2D particle-hole symmetric Kondo lattice where coherence of individual Kondo screening clouds opens up a hybridization gap at the Fermi level and gives rise to the Kondo insulating phase. We study the most challenging regime \( J/t = 1.6 \) close to the quantum critical point of the 2D KLM [58] where the RKKY interaction and Kondo screening are of the same order of magnitude and thus it is crucial to treat them on equal footing. To this end, we use a finite-temperature version of the auxiliary-field quantum Monte Carlo (QMC) algorithm as implemented in Ref. [59].

We consider the situation depicted in the inset of Fig. 1(a): starting with a single magnetic impurity deposited in the middle part of the metallic surface, in consecutive steps we add closed shells of magnetic moments.
around the initial impurity so as to keep the coordination number fixed. This line of research has explicit experimental relevance: atomically precise engineering with STM was used to study the evolution of local density of states (LDOS) from an isolated iron(II) phthalocyanine molecule to the 2D superlattice on a Au(111) surface [60]. Although the underlying physics is complicated by the SU(4) Kondo effect [61], Ref. [60] provides a novel route to interpolate between the physics of a single Kondo impurity and the 2D Kondo lattice behavior.

First, we focus on local properties at the central impurity \( \mathbf{r} = 0 \): \( f \)-spin susceptibility \( \chi_{f,0} \) and c-electron double occupancy \( D_{c,0} \) (a), c-electron LDOS \( N_{c,0}(\omega) \) (b), and \( f \)-operator LDOS \( N_{f,0}(\omega) \) (c) at the central site \( \mathbf{r} = 0 \) of a Kondo superlattice (see inset) upon increasing the number of impurity shells \( n \) coupled to a \( L \times L \) lattice of c-electrons at temperature \( T \) much below the corresponding single-impurity Kondo scale \( T_K \approx 1/8 \) [23]. Inset: construction of the superlattice; extra impurity shells around the initial impurity (open circle) are indicated using different symbols.

The KLM forbids charge fluctuations on the \( f \)-orbitals. Instead, we examine a local spectral function \( \tilde{N}_{f,0}(\omega) \) extracted from the Green’s function \( G_{f,0}(\tau) = \sum_{\sigma} \langle \tilde{j}_{0,\sigma}^{\dagger}(\tau) \tilde{j}_{0,\sigma}(0) \rangle \) where the \( \tilde{f} \)-operator is defined as 
\[
\tilde{f}_{0,\sigma}^{\dagger} = \sum_{\sigma} (\tilde{c}_{0,\sigma}^{\dagger} \tilde{c}_{0,\sigma} + c_{0,\sigma}^{\dagger} c_{0,\sigma}) \tilde{j}_{0,\sigma}. 
\]

The \( \tilde{f} \)-operator is derived from a single-impurity Anderson model (SIAM) using the Schrieffer-Wolff transformation [64, 65] and describes the so-called cotunneling process [66–68]: the tunnelling of an electron from the STM tip into the conduction sea involves a spin-flip of the local magnetic \( f \)-moments. As apparent, \( \tilde{N}_{f,0}(\omega) \) reproduces the Abrikosov-Suhl resonance of the SIAM in the Kondo model with a single impurity, see Fig. 1(c). Upon increasing \( n \), the resonance splits first, then is replaced by a pseudogap, and finally a full gap opens up at \( n = 3 \) coinciding with the gap that appears in \( N_{c,0}(\omega) \). While the splitting of the Abrikosov-Suhl resonance is already observed in the interimpurity spin-singlet state of the two-impurity Anderson model with a direct Heisenberg interaction between the \( f \)-spins [69–71], here we assume the absence of a direct overlap between the \( f \)-orbitals. Thus, the opening of the gap seems to signify the onset of Kondo coherence. Moreover, given that upon further increasing \( n \) peaks on the flanks of the gap sharpen and become reminiscent of those found in the lattice limit [20, 72, 73], it is tempting to assume that they stem from nearly flat HF bands with predominantly \( f \)-character.

Next, we consider spatial properties of superlattices summarized in Fig. 2. The build up of intersite AF spin correlations \( S_{ij}(\mathbf{r}) = 4\langle \tilde{S}_{i,\sigma}^{\dagger} \tilde{S}_{j,\sigma} \rangle \) measured relative to the central impurity gives rise to a collective-like screening of the \( f \)-impurities seen as the enhancement of \( \chi_{f,0}^{-1}(\mathbf{r}) \) at inner shells. It also assists localization of c-electrons reflected in the reduction of \( D_{c,\mathbf{r}} \) in the center. In contrast, the outermost shell, in particular the corner sites, clearly stand out. It stems from two effects: (i) a reduced number of nearest-neighbor impurities at the edge makes the RKKY interaction less important and (ii) since the edge is immersed in a conduction electron sea, a locally enhanced density of c-electrons states \( \rho_{c,\mathbf{r}} \) available for Kondo quenching of the edge impurities introduces a site-dependent Kondo temperature \( T_{K,c}(\mathbf{r}) \sim \rho_{c,\mathbf{r}}^{-1/2}(\omega_0 = 0) \). Furthermore, while the spatial characteristics of the \( n = 3 \) and larger superlattices are very much alike, cf. Figs. 2(g–i) and 1S in Ref. [65], one can discern a nonmonotonous evolution of both \( \chi_{f}^{-1}(\mathbf{r}) \) and
$D_c(r)$ when moving from the center towards the corner atom of the $n=2$ system. Together with the initial oscillating behavior of both quantities upon increasing $n$ in Fig. 1(a) it is indicative of a strong competition between the local Kondo physics at the edges and lattice effects in the center.

Given the onset of a Kondo insulating phase in the core of systems with $n \geq 3$, one would like to know what happens at the surface of the insulator immersed in a conduction electron sea and, in particular, whether and to what extent the insulator is penetrable to those electrons? To get more insight into this issue, we show in Fig. 3 how the $\tilde{f}$-operator LDOS $N_{\tilde{f}r}(\omega)$ evolves when moving from the central $(0,0)$ impurity to the corner $(n,0)$ site for superlattices with different number of shells $n$. As apparent, independently of whether the gap at the central site is full or partial, the corner always develops the Kondo effect, independently of whether the gap at the central site is full or partial; similarly, $N_{\tilde{f}r}(\omega = 0)$ at the other $(4,1)$ and $(3,2)$ sites of the edge grows steadily with reducing $T$ lending further support for the emergent peaks at our lowest $T = t/30$, see Fig. 3(d). They signal the penetration of conduction electron gas into the correlated superlattice via the Kondo effect and stem from the single-impurity Kondo physics which prevails locally over intersite correlations. As such they are specific to the geometry of a superlattice. Indeed, interfacing the Kondo lattice layer with a noninteracting metal leads merely to softening of the hybridization gap in the Kondo lattice layer [74].

An important issue that one might be concerned about is whether opening of a direct (optical) gap in the local quantity $N_{\tilde{f}r}(\omega)$ can be considered as unambiguous evidence of coherence, which is a global phenomenon in the system? Indeed, in a translation-invariant system, the appearance of a hybridized HF band structure with a small indirect gap is usually inferred either from the momentum-dependent single-particle spectral function $A(k, \omega)$ [75–81] or from the $T$-dependence of transport properties [82, 83]. Given that the translatinal symmetry is broken in our superlattices, one has to use an alternative strategy to assess the formation of HF bands. One possibility stems from the fact that the 2D particle-hole symmetric KLM subject to a magnetic field features a phase transition from the Kondo insulator to a canted
AF state [84–86]. The AF phase is understood as a spin-density-wave instability driven by perfect nesting of the particle and hole hybridized bands with opposite spin indices. Thus it is legitimate to consider the field-induced transverse antiferromagnetism as the criterion of the coherent Kondo lattice behavior.

With this considerations in mind, we perform QMC simulations of the KLM (1) augmented by a Zeeman term,

$$H_B = -g\mu_B B \sum_i (S^z_{i,i} + S^z_{\bar{i},\bar{i}}),$$

and calculate real-space transverse-spin correlations $S^{xy}_f(r)$ relative to the central impurity of the $n = 2$ (a-c), $n = 3$ (d-f), and $n = 5$ (g-i) systems subject to increasing magnetic field $\mu_B B/t = 0.06$ (left), 0.13 (middle), and 0.2 (right). Parameters: $L = 16$ and $T = t/30$.

Finally, since the QMC algorithm maps a quantum system in $d$ spatial dimensions onto a $d + 1$-dimensional classical problem with an additional imaginary-time dimension $\beta = 1/T$, strengthening of in-plane AF spin correlations should be equally seen in the imaginary-time-displaced correlation function $S^{xy}_{f,0}(\tau) = 2\langle (S^y_{f,0}(\tau)S^y_{f,0}(0)) + (S^y_{f,0}(\tau)S^y_{\bar{f},\bar{0}}(0)) \rangle$ at the central site. Figure 5 displays how the corresponding dynamical transverse-spin structure factor $S^{xy}_{f,0}(\omega)$ at the central impurity as a function of out-of-plane magnetic field $B$ for the system with $n = 2$ (a), $n = 3$ (b), and $n = 5$ (c) shells. From bottom to top: $\mu_B B/t = 0, \ldots, 0.2$. Parameters: $L = 16$; $T = t/30$ (solid) and $T = t/15$ (dashed).

FIG. 4. Real-space transverse-spin correlations $S^{xy}_f(r)$ relative to the central impurity of the $n = 2$ (a-c), $n = 3$ (d-f), and $n = 5$ (g-i) systems subject to increasing magnetic field $\mu_B B/t = 0.06$ (left), 0.13 (middle), and 0.2 (right). Parameters: $L = 16$ and $T = t/30$.

FIG. 5. Dynamical transverse-spin structure factor $S^{xy}_{f,0}(\omega)$ at the central impurity as a function of out-of-plane magnetic field $B$ for the system with $n = 2$ (a), $n = 3$ (b), and $n = 5$ (c) shells. From bottom to top: $\mu_B B/t = 0, \ldots, 0.2$. Parameters: $L = 16$; $T = t/30$ (solid) and $T = t/15$ (dashed).
with exactly one conduction electron per impurity spin. It leads to the nesting-driven enhancement of the RKKY interaction which assists the opening the hybridization gap on top of that from coherent scattering off Kondo singlets. It motivates future QMC studies of the composite heavy quasiparticle formation in depleted Kondo nanosystems where some impurity spins are removed in a regular way promoting metallicity [88, 89].

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Supplemental Material for: Emergent Coherent Lattice Behavior in Kondo Nanosystems

Schrieffer-Wolff transformation and electron cotunneling

For simplicity we consider a symmetric single-impurity Anderson model (SIAM) defined as:

\[
H_{\text{SIAM}} = \sum_{\mathbf{k},\sigma} \varepsilon(\mathbf{k}) c_{\mathbf{k},\sigma}^\dagger c_{\mathbf{k},\sigma} + U \left( f_\uparrow f_\downarrow - 1/2 \right) \left( f_\uparrow f_\downarrow - 1/2 \right) + \frac{V}{\sqrt{N}} \sum_{\mathbf{k},\sigma} \left( c_{\mathbf{k},\sigma}^\dagger f_\downarrow + f_\downarrow c_{\mathbf{k},\sigma} \right).
\]

The Abrikosov-Suhl resonance of the SIAM emerges in the single-particle spectral function of the \( f \)-electrons:

\[
G_f(\tau) \equiv \sum_{\sigma} \langle f_\sigma(\tau) f_\sigma^\dagger(0) \rangle = \int d\omega K(\tau, \omega) \text{Im} G^R_{ret}(\omega),
\]

where

\[
\text{Im}G^R_{ret}(\omega) = \frac{\pi}{Z} \sum_{n,m,\sigma} \left( e^{-\beta E_m} + e^{-\beta E_n} \right) \times |\langle n | f_\sigma^\dagger | m \rangle|^2 \delta(\omega + E_n - E_m),
\]

and the Kernel is given by:

\[
K(\tau, \omega) = \frac{1}{\pi} e^{\tau \omega} \frac{1}{1 + e^{\beta \omega}}.
\]

We now carry out the Schrieffer-Wolff canonical transformation required to eliminate the hybridization term in first order and to derive the Kondo model. Let

\[
S^\dagger = -S, \text{ and } [S, H_0] = -H_1,
\]

then

\[
e^S H e^{-S} = H_0 + \frac{1}{2} [S, H_1] + O(\epsilon^3).
\]

Here, we have formally assumed that \( H_1 \) and the generator \( S \) are of order \( \epsilon \). Under this canonical transformation, the imaginary-time-displaced Green’s function:

\[
G_f(\tau) \equiv \sum_{\sigma} \langle e^{S} f_\sigma e^{-S}(\tau) e^{S} f_\sigma^\dagger e^{-S}(\tau = 0) \rangle,
\]

should reproduce the Abrikosov-Suhl resonance of the SIAM in the Kondo model with a single impurity. The calculation gives:

\[
S = \frac{V}{\sqrt{N}} \sum_{\mathbf{k},\sigma} \left( \frac{c_{\mathbf{k},\sigma}^\dagger f_\sigma P_o}{\varepsilon(\mathbf{k}) + U/2 - i0^+} + \frac{c_{\mathbf{k},\sigma}^\dagger f_\sigma P_e}{\varepsilon(\mathbf{k}) - U/2 - i0^+} - H.c. \right),
\]

where \( P_o \) (\( P_e \)) is the projection of the even, \((-1)\sum_o f_\sigma^\dagger f_\sigma = 1 \) (odd, \((-1)\sum_o f_\sigma^\dagger f_\sigma = -1 \)) respectively, parity sector of the \( f \)-electron. We can now transform the creation operator for \( f \)-electrons, and constrain the Hilbert space to \( P_o \) so as to obtain:

\[
f_\sigma^\dagger \approx e^S f_\sigma^\dagger e^{-S} \approx \frac{V}{\sqrt{N}} \sum_{\mathbf{k},\sigma} \left( \frac{c_{\mathbf{k},\sigma}^\dagger f_\sigma^\dagger}{\varepsilon(\mathbf{k}) + U/2 - i0^+} - \frac{f_\sigma c_{\mathbf{k},\sigma}}{\varepsilon(\mathbf{k}) + U/2 - i0^+} \right).
\]

The first (second) term involves a virtual doubly (empty) occupied \( f \)-site. Assuming that \( U \) is the largest scale so that we can set \( \varepsilon(\mathbf{k}) \) to zero gives the final result:

\[
f_\sigma^\dagger \approx -\frac{V}{U} \sum_{\sigma} \left( c_{\mathbf{r} = 0, \sigma}^\dagger f_\sigma f_\sigma^\dagger + f_\sigma^\dagger c_{\mathbf{r} = 0, \sigma} f_\sigma \right).
\]

Using the relation, \( 2 f_\sigma^\dagger f_\sigma = n_f + \sigma' 2 S_f' \), where \( \sigma' \) takes the value 1 (−1) for up (down) spin degrees of freedom, \( S_f' = \frac{1}{2} \sum_{\sigma} \sigma' f_{\sigma}^\dagger f_{\sigma} \), and \( n_f = \sum_{\sigma} f_{\sigma}^\dagger f_{\sigma} \), the above operator can be rewritten as:

\[
\tilde{f}_{\sigma'}^\dagger \approx \frac{2V}{U} \left( c_{\mathbf{r} = 0, -\sigma'}^\dagger S_f' + \sigma' c_{\mathbf{r} = 0, \sigma} S_f' \right).
\]

Here \( S_f' = f_{\sigma'}^\dagger f_{-\sigma'} \) and we have used the fact that in the Kondo regime \( n_f = 1 \). The corresponding impurity spectral function for the Kondo model matches that derived in Ref. [91] using the equation of motion for the \( c \)-electron Green’s function.

### Supplemental data

The relation between the number of shells \( n \) and the corresponding total number of impurities \( N_{\text{imp}} \) in a superlattice is summarized in Table I. Figure 1S shows the spatial characteristics of the \( n \in \{4,5\} \) superlattices. The behavior of transverse-spin correlations \( S_{f'}(\mathbf{r}) \) upon increasing the external magnetic field \( B \) is documented more quantitatively in Fig. 2S.

| TABLE I. Total number of magnetic impurities \( N_{\text{imp}} \) in a nanosystem with \( n \) shells. |
|---|
| \( n \) | 0 | 1 | 2 | 3 | 4 | 5 |
| \( N_{\text{imp}} \) | 1 | 5 | 13 | 25 | 41 | 61 |
FIG. 1S. Real-space spin correlations $S_f^z(r)$ relative to the central impurity (left) and spatial dependence of: inverse $f$-spin susceptibility $\chi_f^{-1}(r)$ (middle) and c-electron double occupancy $D_c(r)$ (right) in the $n = 4$ (a-c) and $n = 5$ (d-f) systems. Parameters: $J/t = 1.6$, $T = t/30$, and $L = 16$.

FIG. 2S. Transverse-spin correlations $S_f^{xy}(r)$ between the $(i,0)$ site with $i = 1, \ldots, n$ and the central $(0,0)$ impurity as a function of out-of-plane magnetic field $B$ for the system with $n = 1$ (a), $n = 2$ (b), $n = 3$ (c), $n = 4$ (d), and $n = 5$ (e) shells.