Heavy Antiferromagnetic Phases in Kondo Lattices

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We propose a microscopic physical mechanism that stabilizes coexistence of the Kondo effect and antiferromagnetism in heavy-fermion systems. We consider a two-dimensional quantum Kondo-Heisenberg lattice model and show that long-range electron hopping leads to a robust antiferromagnetic Kondo state. By using a modified slave-boson mean-field approach we analyze the stability of the heavy antiferromagnetic phase across a range or parameters, and discuss transitions between different phases. Our results may be used to guide future experiments on heavy fermion compounds.

Introduction. The study of complex phenomena exhibited by materials with competing ground states is the primary focus of modern condensed matter physics. In systems where local magnetic moments interact with a conduction band there are two opposing quantum many-body effects: Kondo screening (formation of singlets between the local moments and itinerant electrons), and a long-range magnetic [often antiferromagnetic (AF)] order. Since metallic Kondo phase and magnetic ordering involve the same local-moment degrees of freedom, they contest the same entropy. This competition is at the heart of the rich variety of phases observed in heavy-fermion (HF) f-electron materials under tuning of external parameters such as pressure, doping, or magnetic field [1].

In the Kondo screened phase the Fermi surface (FS) volume accounts for the local spins. Quantum (zero-temperature) phase transitions between this HF metal with large FS and small-FS magnetic states remain an actively debated subject [2, 3]. Of particular interest is whether the Kondo screening collapses precisely at the onset of magnetism [4, 5] or the magnetic transition is of a spin-density wave (SDW) type [6, 7] with no concomitant change of the FS volume. Both scenarios are found in experiments [3]. In $Ce_3Pd_{20}Si_6$ [8] Kondo screening disappears inside a magnetically ordered phase, while in $CeCu_2Si_2$ the AF transition is likely of the SDW type [9]. In $YbRh_2Si_2$ data indicate that Kondo screening disappears at the magnetic transition [10, 11], while under Co and Ir doping the two transitions separate [11]. Developing microscopic theories which exhibit such multitude of phases proved difficult [1, 3, 12]. Recently it was conjectured [3, 13] that frustrated magnetic interactions between local moments give rise to a variety of phase that include magnetically ordered as well as paramagnetic states with both large and small Fermi surfaces, but detailed theories are still lacking.

In this Letter we propose a microscopic mechanism which controls the coexistence of the Kondo effect and AF long-range order. We consider the two-dimensional Kondo-Heisenberg lattice model with short-range AF interactions between local moments, and conduction-electron hopping beyond nearest neighbors (NN). By employing the notion of a "spin-selective Kondo insulator" [14, 15] introduced in the context of ferromagnetism (FM) in Kondo lattices we show that in a bipartite Kondo lattice with NN and next-nearest neighbor (NNN) electron hopping the same physics leads to a robust AF Kondo ["K+AF" in Fig. 2(a)] phase. The stability of this state is controlled by the relative magnitude of short- and long-range hopping amplitudes. The AF Kondo phase has a large FS, and is separated by a second-order quantum phase transition from the small-FS AF metal. In contrast to previous studies [16, 17] we focus on the experimentally relevant regime away from half-filling. We find the phases somewhat similar to those suggested in Ref. [12], but the underlying physics is different, see discussion below.

Model and approximations. The essential physics of magnetic HF materials is contained in the Kondo-Heisenberg lattice model (KHLM),

$$H = H_{\text{cond}} + H_{\text{Kondo}} + H_{\text{Heis}} = \tag{1}$$
$$= -\sum_{(ij),\alpha} t_{ij}\left(c_{i\alpha}^\dagger c_{j\alpha} + \text{h.c.}\right) - \mu_c \sum_{i,\alpha} c_{i\alpha}^\dagger c_{i\alpha} +$$
$$+ J_K \sum_i \boldsymbol{S}_i \boldsymbol{s}_i + J_H \sum_{\langle ij \rangle} \boldsymbol{S}_i \boldsymbol{S}_j.$$

This Hamiltonian describes a system of itinerant electrons, $c_{i\alpha}$ with spin indices α, $\beta = \uparrow, \downarrow$, interacting with local spin-1/2 moments $\boldsymbol{S}_i$ via the AF Kondo coupling J_K. We consider this Hamiltonian on a square lattice with sites labeled by i and j. The competition to the Kondo screening is provided by the AF exchange $J_H > 0$ between NN spins. The hopping amplitude $t_{ij} = t > 0$ for NN links $(ij) = \langle ij \rangle$, and $t_{ij} = t'$ for NNN sites, $(ij) = \langle\langle ij \rangle\rangle$; for all other sites $t_{ij} = 0$. The electron spin operator is $\boldsymbol{s}_i = \boldsymbol{\sigma}_{\alpha\beta} c_{i\alpha}^\dagger c_{i\beta}/2$, where $\boldsymbol{\sigma}$ are the Pauli matrices. The chemical potential μ_c controls the conduction band filling.

We employ the hybridization mean-field (HMF) approach [1], where local spins are represented in terms of spin-1/2 pseudo-fermions, $\boldsymbol{S}_i = \boldsymbol{\sigma}_{\alpha\beta} f_{i\alpha}^\dagger f_{i\beta}/2$, subject to the constraint $\sum_\alpha f_{i\alpha}^\dagger f_{i\alpha} = 1$, which removes unphysical

empty and doubly-occupied states from the single-site Hilbert space. The core idea of HMF is to treat this constraint on the average by introducing the pseudo-fermion "chemical potential" μ_f. The interactions in Eq. (1) now contain four fermion operators and are decoupled within the Hartree-Fock approximation. We implement this approach along the lines of Ref. [18], and consider decouplings in all possible channels, namely:

(i) Magnetic channel

$$H_{\text{Kondo}} + H_{\text{Heis}} \rightarrow \qquad (2)$$
$$\rightarrow J_K \sum_i \big(\boldsymbol{M}_i \boldsymbol{s}_i + \boldsymbol{m}_i \boldsymbol{S}_i \big) + J_H \sum_{\langle ij \rangle} \big(\boldsymbol{M}_i \boldsymbol{S}_j + \boldsymbol{M}_j \boldsymbol{S}_i \big),$$

where magnetic order parameters (OPs) are defined as $\boldsymbol{M}_i = \langle \boldsymbol{S}_i \rangle$ and $\boldsymbol{m}_i = \langle \boldsymbol{s}_i \rangle$, and the arrow indicates that we omitted c-number terms.

(ii) Pseudo-fermion dispersion ("spin-liquid") channel

$$H_{\text{Heis}} \rightarrow -\frac{J_H}{4} \sum_{\langle ij \rangle} \boldsymbol{\sigma}_{\alpha'\alpha} \boldsymbol{\sigma}_{\beta'\beta} \big[\langle f_{i\alpha'}^\dagger f_{j\beta} \rangle f_{j\beta'}^\dagger f_{i\alpha} + \text{h.c.} \big]. \quad (3)$$

(iii) Kondo hybridization channel

$$H_{\text{Kondo}} = -\frac{3J_K}{4} \sum_i \chi_{i0}^\dagger \chi_{i0} + \frac{J_K}{4} \sum_i \boldsymbol{\chi}_i^\dagger \boldsymbol{\chi}_i \rightarrow \qquad (4)$$
$$\rightarrow J_K \sum_i \left[-\frac{3}{4} \langle \chi_{i0} \rangle^* \chi_{i0} + \frac{1}{4} \langle \boldsymbol{\chi}_i \rangle^* \boldsymbol{\chi}_i + \text{h.c.} \right]$$

with $\chi_{i\mu} = \sigma_{\alpha\beta}^\mu f_{i\alpha}^\dagger c_{i\beta}/\sqrt{2}$, $\mu = 0 \ldots 3$ and $\sigma_{\alpha\beta}^0 = \delta_{\alpha\beta}$. Physically, the χ-operators are Schwinger bosons [19] which create local singlet and triplet states resulting from the Kondo coupling between localized and itinerant spins. In the usual picture of Kondo singlet formation only χ_0-boson is present. The magnetic order admixes other components, so that the χ-representation of Eq. (4) captures the $SU(2)$ invariance of the Kondo interaction [18].

Since we consider only uniform and commensurate AF states on the square lattice, there are two wavevectors in the problem: $\boldsymbol{q} = 0$ and $\boldsymbol{q} = \boldsymbol{Q}_0 = (\pi, \pi)$. In the AF phase all OPs are site-dependent. Thus the pseudo-fermion density $n_i^f = \sum_\alpha f_{i\alpha}^\dagger f_{i\alpha}$ will acquire an unphysical spatial dependence. To suppress the Q_0 harmonic $n_{Q_0}^f$ we impose an additional constraint, $H \rightarrow H - \mu_Q \sum_i e^{iQ_0 x_i} n_i^f$, where μ_Q is the Lagrange multiplier.

In the rest of the paper we study the phase diagram of the KHLM as a function of t', J_H and temperature, T, at fixed density $n^c = 0.8$. We choose the spin quantization axis along the z-direction and omit the corresponding vector indices whenever possible. At intermediate coupling we need to sum over the entire Brillouin zone and numerically solve the HMF equations in the momentum space on the 32×32 square lattice with periodic boundary conditions, which is close to the thermodynamic limit. Indeed increasing system size by a factor of 4 leads to only $\sim 1\%$ correction to the results below.
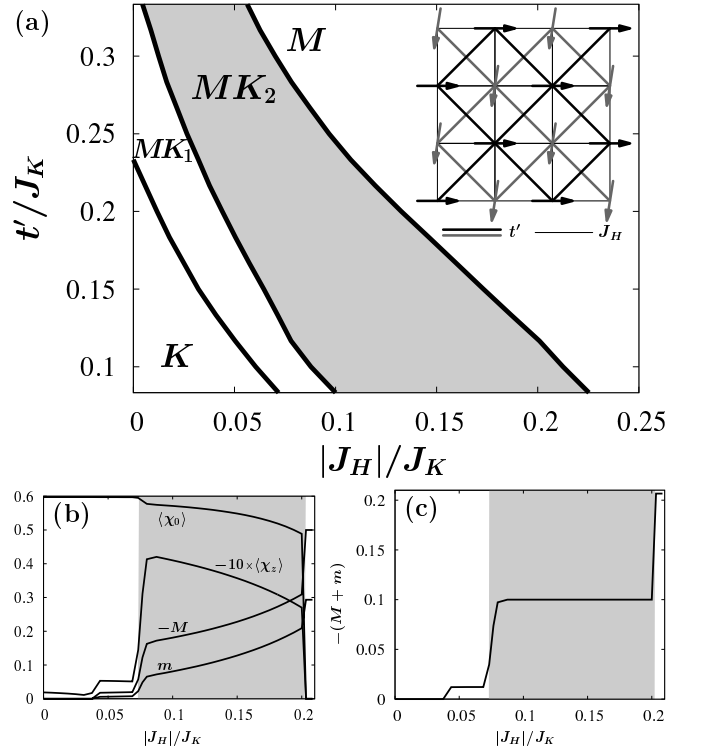

FIG. 1. KHLM with $t = 0$. (a) $T = 0$ phase diagram: "K" denotes singlet Kondo phase, "M" is the local moment magnetic phase, $MK_{1,2}$ are magnetic Kondo phases. M and $MK_{1,2}$ are ordered at $\boldsymbol{q} = 0$ (FM) if $J_H < 0$ and (π, π) (AF) for $J_H > 0$. All phase transitions are 1st order. Inset: Square lattice with $t = 0$. The arrows denote local moments. (b) and (c) Hybridization $\langle \chi_{0,z} \rangle$, magnetic ($M$ and m) OPs, and total magnetization $M + m$ for $t'/J_K = 2/15$. The plateau in (c) signals that Eq. (5) is satisfied inside MK_2 but not MK_1.

Origin of the AF Kondo phase: $t = 0$ limit. This limit allows a simple analysis and guides subsequent discussion of physical regimes. It is known [14, 15] that the Kondo lattice Hamiltonian with $J_H = 0$ and NN hopping exhibits a ferromagnetic HF state characterized by finite Kondo hybridization and magnetic polarization. In that "spin-selective Kondo insulator" phase [14], away from half-filling, all minority-spin and the corresponding fraction of the majority-spin conduction electrons screen part of the f-electron spin, while the magnetic moments of the remaining conduction and local f-electrons are antiparallel to take advantage of the Kondo coupling.

To reveal the physical mechanism responsible for coexistence of the Kondo effect and antiferromagnetism, it is instructive to start with a limiting case with NN hopping $t = 0$, but finite NNN hopping t'. The model Eq. (1) then reduces to two interpenetrating square Kondo (sub)lattices, coupled by the Heisenberg term [see inset in Fig. 1(a)]. When $J_H = 0$ and t'/J_K is large enough, each sublattice enters a ferromagnetic Kondo state with non-zero $\langle \chi_{0,z} \rangle$ and magnetic OPs. The ground state of

this system is continuously degenerate with respect to the angle between the magnetizations of the two sublattices, similar to the J_1-J_2 antiferromagnet with NN (J_1) and NNN (J_2) interactions for large J_2/J_1 [20]. Finite J_H lifts this degeneracy and stabilizes long-range magnetic order coexisting with Kondo singlets across the entire lattice. For $t = 0$ only the Heisenberg interaction couples the sublattices, and, at least at the level of HMF, the ground state energy is an even function of J_H. The FM (AF) states are stabilized for $J_H < 0$ ($J_H > 0$). Below, m and M denote uniform (for $J_H < 0$) and staggered (for $J_H > 0$) z-axis magnetizations in the c- and f-channels respectively. The vector part of the hybridization, $\langle \chi_z \rangle$, has the same Fourier components as m and M, while the singlet hybridization amplitude $\langle \chi_0 \rangle$ is always uniform.

Fig. 1(a) shows the $T = 0$ mean field phase diagram of the KHLM with $t = 0$. For small t' and $|J_H|$ the system resides in the Kondo singlet state. In the opposite limit, the Kondo effect is suppressed in favor of magnetism. In the intermediate range of parameters several magnetic Kondo states (MK$_{1,2}$) are stabilized. All phase transitions in Fig. 1(a) are discontinuous as illustrated by the $|J_H|$-dependence of the hybridization and magnetic OPs in Fig. 1(b). The states MK$_{1,2}$ can be distinguished based on the commensurability condition [14]

$$2(M + m) = |1 - n^c|, \qquad (5)$$

satisfied only inside MK$_2$. Consequently, the state MK$_2$ has a plateau in the total magnetization, shown in Fig. 1(c). We also note that the phase MK$_1$ is quite fragile and may be an artifact of the HMF approximation.

Coexistence of antiferromagnetism and Kondo effect. The above picture survives in the physically relevant limit $0 \leqslant t'/t \leqslant 1$. Similar to the $t = 0$ case, when $J_H = 0$ and J_K are small compared to the electron bandwidth the system undergoes a transition to a ferromagnetic Kondo state. To avoid this, we fix the Kondo coupling at $J_K = 6t$ (bandwidth is $\sim 8t$), so that for $J_H = 0$ and any $t' \in [0, 1]$ the ground state is the non-magnetic uniform HF phase.

Numerical solution of Eqs. (1)–(4) exhibits a complex phase diagram shown in Fig. 2(a). Its most important feature is the existence of a critical point D at $t'_c/t \approx 0.46$. For $t' < t'_c$ there is a direct 1st order transition between the large-FS HF and small-FS AF states. On the other hand, for $t' > t'_c$ there is a large parameter range (shaded region in the figure) where Kondo screening coexists with antiferromagnetism. The hybridization OP is uniform in the Kondo phase, $\langle \chi_{i0} \rangle = \langle \chi_0 \rangle$, and vanishes in the AF state. The latter is characterized by $M_i^z = M e^{iQ_0 x_i}$ and $m_i^z = m e^{iQ_0 x_i}$. In the coexistence phase (K + AF) the singlet component of the hybridization acquires a small staggered component, $\langle \chi_{i0} \rangle = \langle \chi_0 \rangle + \langle \tilde{\chi}_0 \rangle e^{iQ_0 x_i}$ ($|\langle \tilde{\chi}_0 \rangle / \langle \chi_0 \rangle| \sim 0.01$), while the vector part of the Kondo hybridization is purely staggered, $\langle \chi_{iz} \rangle = \langle \chi_z \rangle e^{iQ_0 x_i}$, tracking the spatial variations of the magnetization.
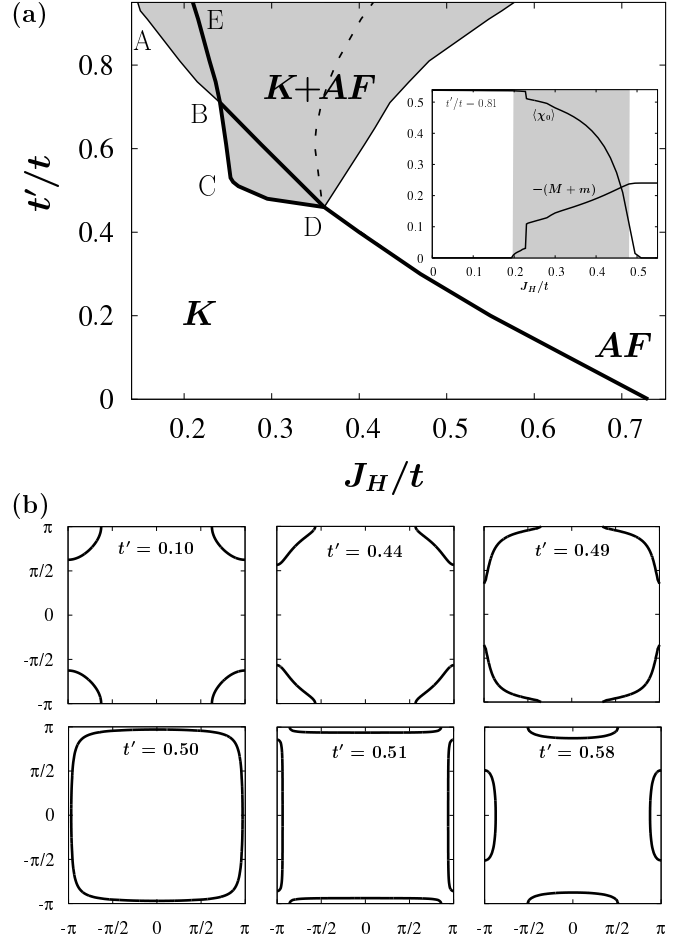

FIG. 2. KHLM at $T = 0$. (a) Phase diagram. "K" is the singlet Kondo phase, and "K + AF" is the large-FS AF Kondo state. The dashed line is a continuation of the boundary between K and AF phases, obtained if their coexistence is prohibited in the calculation. Thick (thin) lines denote 1st (2nd) order transitions. Inset: Uniform part of the Kondo hybridization OP and total staggered magnetization for $t'/t = 0.81$. (b) Evolution of the heavy quasiparticle FS with t' for $J_H = 0$. The Lifshitz transition at $t'/t \sim 0.5$ corresponds to point C in panel (a).

The coexistence phase is separated from the AF state by a 2nd order transition. However, its boundary with the pure Kondo state is more complex. In Fig. 2(a) the line AB is the 2nd order transition, line DCB is weakly 1st order, and BD and BE are strongly 1st order transitions. The AF Kondo state inside regions ABE and BCD differs from the rest of the intermediate phase only in its value of the staggered magnetization $M + m$, see inset of Fig. 2(a). Absence of the magnetization plateau in this phase [cf. Fig. 1(c)] indicates that the commensurability condition (5) no longer holds in the presence of the NN hopping.

The kink in the phase boundary at point C is due to the change in the topology of the heavy quasiparti-
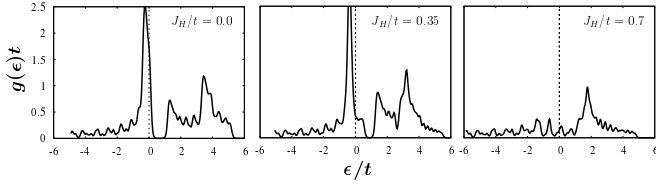
FIG. 3. Quasiparticle DOS for a fixed $t'/t = 0.81$ and J_H inside the singlet Kondo phase (left), magnetic Kondo state (middle) and AF metal (right). The energy ϵ is measured relative to the Fermi level. In the left and middle panels the Kondo peak is apparent.

cle FS (Lifshitz transition) at $t'/t \sim 0.5$ in Fig. 2(b) [21]. Hence, the intermediate phases ABE and BCD may signify tendency towards incommensurate ordering, rather than the (π, π) AF state that we consider. More studies are needed to determine the exact spatial structure of the coexistence state. Although the AF Kondo state exists only because of NNN hopping, increasing t' eventually destroys it when J_K becomes small compared to the bandwidth.

Finally, in Fig. 3 we present the quasiparticle density of states (DOS) in the three phases along the line $t'/t = 0.81$ in Fig. 2(a). As expected, the DOS in the AF Kondo phase shows a peak near the Fermi surface, similar to the singlet Kondo state, and is very different from the DOS in the small-FS AF metal.

Finite temperature behavior. To assess thermal stability of the AF Kondo state, in Fig. 4(a) we present the finite-T phase diagram of the KHLM computed along the $\prod$-shaped path centered at point D of Fig. 2(a). In the HMF analysis the Kondo screened phase (K) always vanishes via a 2nd order phase transition to a small-FS metal at a critical temperature $T_K(J_H, t')$. Depending on J_H and t' the HF metal may become unstable towards either a pure AF state or a "$K + AF$" phase [shaded regions in Fig. 4(a)] at a Néel temperature $T_N(J_H, t') < T_c$. The latter phase extends over a sizeable range $T_N/T_c \lesssim 1/4$ and is separated from the singlet Kondo state by a 2nd order transition. This is illustrated in Fig. 4(b) which shows the temperature dependence of all OPs. In contrast, pure AF phase is separated from the HF state by a 1st order transition.

It is interesting to observe that Fig. 4 resembles the phase diagram of a spin-1 underscreened KLM [22] if T_N is replaced by the Curie temperature. Finally, we note that, although spontaneous continuous symmetry-breaking in a pure 2D system is prohibited by the Mermin-Wagner theorem [23], an infinitesimal coupling to the third dimension will be sufficient to stabilize the ordered phases in Fig. 4.

Discussion. Our study identifies the long-range electron hopping as a physical mechanism responsible for the robust coexistence of antiferromagnetism and Kondo ef-
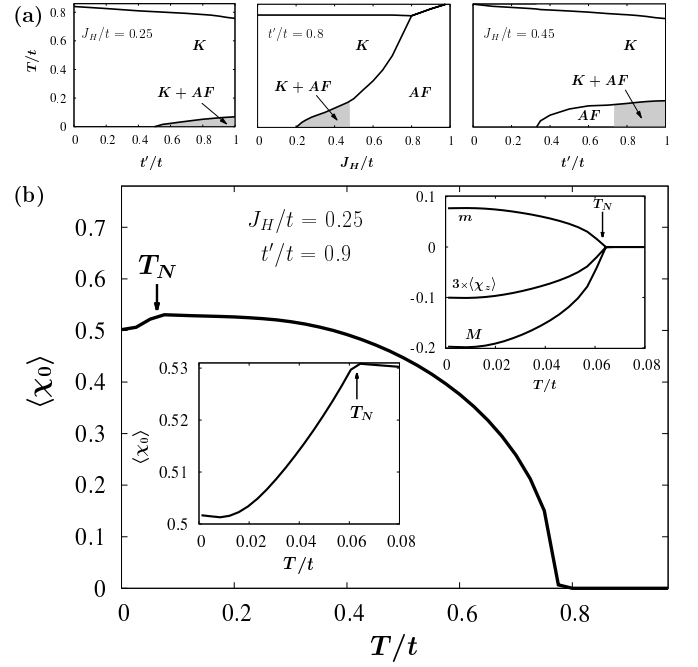

FIG. 4. KHLM at finite T. (a) Phase diagrams exhibiting competing orders, see text. Notations are the same as in Fig. 2. Shaded regions denote AF Kondo phases. The transition between Kondo and AF (AF Kondo) states is 1st (2nd) order. Unmarked phases are usual Fermi liquids. (b) OPs in the AF Kondo phase for a fixed $J_H/t = 0.25$ and $t'/t = 0.9$: $\langle \chi_0 \rangle$ (main panel and lower inset), and $\langle \chi_z \rangle$ and staggered magnetizations (upper inset).

fect in the description of HF materials via the Kondo-Heisenberg lattice model. The parameter t'/t controls whether the Kondo screening can vanish precisely at the onset of the magnetic order, or via and intermediate coexistence regime, covering the range of experimental data. Although we considered a square lattice, our results can be straightforwardly applied to any bipartite graph.

The phase diagram in Fig. 2(a) can be tested in HF systems by changing t'/t with pressure. However, as J_H is also usually pressure-sensitive, the exact path cut in Fig. 2(a) by such an experiment is not clear and either an AF or K + AF phase may be realized. Chemical pressure, such as substitution of Ni for Pd, or Co for Rh in Ce- or Yb-based HF compounds, can also be used to explore the phases and the transitions between them. Study of the critical behavior near point D would be particularly intriguing.

We emphasize that our study is distinct from previous works, notably Ref. 12 which considered the model (1) with $t' \equiv 0$ and found a phase diagram that included the heavy SDW state similar to our AF Kondo phase. However, their analysis involved enforcing the particular form of the spin-liquid order and imposing unequal coupling constants in Eqs. (2) and (3) to stabilize it (in our case

they are both equal to J_H). We provided a framework where the fully self-consistent treatment of the KHLM allows controlled analysis to the different OPs. Crucially, while we find the non-zero "spin-liquid" order parameter in all the Kondo screened phases, it vanishes in the local moment antiferromagnet, and therefore the spinon FS is absent in that phase.

It is known that the finite-T HMF phase transition between HF and small-FS metal phases becomes a crossover when fluctuations beyond HMF are taken into account. However, the condensation of bosons χ_0 and $\boldsymbol{\chi}$ does remain a phase transition at $T = 0$ [12], and hence salient features of the quantum phase diagram of Fig. 2(a) remain unchanged, although phase transition lines may shift. At finite T we expect a wide quantum critical regime above those transition lines.

We focused on commensurate magnetic phases. However, at least in the classical KHLM such states are often suppressed in favor of incommensurate spiral [24] or skyrmion [25] phases. Therefore it would be interesting to establish whether such states are realized in the *quantum* KHLM. We leave this for future investigations.

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