Orbital Order and Hund's Rule Frustration in Kondo Lattices

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We analyze a microscopic origin of the Kondo effect–assisted orbital order in heavy-fermion materials. By studying the periodic two-orbital Anderson model with two local electrons, we show that frustration of Hund's rule coupling due to the Kondo effect leads to an incommensurate spiral orbital and magnetic order, which exists only inside the Kondo screened (heavy-electron) phase. This spiral state can be observed in neutron and resonant x-ray scattering measurements in U- and Pr-based heavy-fermion compounds, and realized in cold atomic gases, e.g., fermionic ^{173}Yb.

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Introduction.—The dichotomy between the localized and itinerant behavior of electrons in solids often leads to a rich variety of quantum states of matter with fascinating physical properties. In some materials $4f$ or $5f$ electrons physically move on and off the ionic site. Even when such valence fluctuations are suppressed, virtual transitions lead to hybridization of these electrons with the conduction band. For one f electron in a single orbital per site, the resulting Hamiltonian describes an interplay between Kondo screening of the local spin by the conduction band (which yields a large Fermi surface with heavy quasiparticles) and local moment magnetism due to the long-range Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. This competition leads to intriguing physics of heavy fermion (HF) metals and results in complex phase diagrams [1] including superconducting and magnetically ordered phases [2].

Materials where localized electrons occupy several (nearly) degenerate atomic orbitals display an even richer physics because the orbital degree of freedom becomes an active participant in establishing the Kondo screened phase [1,3]. If different orbital configurations are not related by the time reversal symmetry, the ground state (GS) of an f-electron ion may have a finite electric multipole moment [4]. The phases associated with long-range ordering of these multipoles were observed in various compounds, e.g., antiferroquadrupole states in CeB_6 [5,6], $PrPb_3$ [7], and $PrIr_2Zn_{20}$ [8], or octupole order in NpO_2 [9] and $Ce_{0.7}La_{0.3}B_6$ [10]. As a result orbital physics in f-electron materials has received much recent theoretical attention [11,12]. It was also suggested that orbital fluctuations provide a glue for unconventional superconductivity [13–15], and are responsible for the ''hidden'' order in URu_2Si_2 [16–18].

Conventional microscopic mechanisms for the orbital order in f-electron systems include an RKKY-like exchange between the multipoles mediated by conduction electrons [7,19] and a direct Heisenberg-like multipole interaction arising in the strong-coupling (t-J–like) limit of a purely f-electron model without the conduction band [20]. In these cases the Kondo screening and multipole order are antagonistic towards each other. Here we show that under certain conditions a long-range orbital order in f-electron materials may exist due to the Kondo effect.

The low-energy electronic configuration of an f-electron ion in the lattice is determined by a hierarchy of energy scales (the j-j coupling scheme) [21]. First, the atomic spin-orbit interaction and crystal electric field (CEF) splitting determine the GS multiplet in accordance with the point symmetry double group [22]. The remaining degeneracy is partially lifted by the Hund's rule interaction. For an isolated multiorbital Kondo impurity, the Hund coupling suppresses the Kondo temperature T_K [23,24]. Conversely, formation of the Kondo resonance aims at restoring the orbital degeneracy thus frustrating the Hund interaction.

In this Letter we use the above intuition to show that the competition between Hund coupling and Kondo screening can give rise to combined orbital and magnetic (generally incommensurate spiral) orders in Kondo lattices (KLs). We consider a single-channel two-orbital periodic Anderson model with two local electrons (the f^2 configuration) in the Kondo regime. In the absence of Hund's splitting J, the mixing of high- and low-spin states of an f-electron ion due to their hybridization with the conduction band yields an emergent $SO(4)$ symmetry of the problem which involves spin and orbital f-electron degrees of freedom on an equal footing, and results in a macroscopic degeneracy of the Kondo screened GS. For a finite J, this symmetry is broken down to $SU(2)$ and the GS degeneracy is partially lifted. Quantum fluctuations inside the HF liquid lead to an effective RKKY-like interaction between magnetic and orbital degrees of freedom, which stabilizes a

long-range orbital order. In contrast to the previous works, this order cannot exist away from the Kondo regime.

Our results are directly applicable to U- and Pr-based HFs in which tetravalent U^{4+} and Pr^{4+} ($5f^2$ and $4f^2$ configurations, respectively) ions have a Γ_8-type CEF GS, and the Hund interaction is small compared to the CEF splitting. They are also relevant for ultracold fermion gases in optical lattices, especially in light of recent proposals to realize KL models with either special optical superlattice structures [25], or alkaline atoms [26] (e.g., ^{173}Yb).

Emergent SO(4) structure in the two-orbital KL model.—We derive the KL model from a two-orbital Anderson model with two localized electrons. First, let us consider a two-orbital Anderson impurity model with a single conduction channel [27]

$$H = \sum_{k\sigma} \varepsilon_k c^\dagger_{k\sigma} c_{k\sigma} + \frac{1}{\sqrt{N}} \sum_{p\sigma a} (v_{pa} c^\dagger_{p\sigma} f_{a\sigma} + \text{H.c.}) + (J - \epsilon_f) N_f - J(S_f^2 + N_f^2/4) + U N_f (N_f - 1)/2,$$

which describes a system of conduction electrons $c_{k\sigma}$ (with momentum k, spin $\sigma = \{\uparrow, \downarrow\}$, and band dispersion ε_k) hybridized with electrons created in two impurity orbitals by $f^\dagger_{a\sigma}$ ($a = 1, 2$) via the orbital-dependent amplitude v_{pa}. The two orbitals correspond to a CEF GS multiplet with the binding energy $-\epsilon_f < 0$. In the above expression, N is the number of lattice sites, $N_f = \sum_{a\sigma} f^\dagger_{a\sigma} f_{a\sigma}$, and $S_f = (1/2) \sum_a f^\dagger_{a\alpha} \sigma_{\alpha\beta} f_{a\beta}$ (σ are the Pauli matrices) define the electron number and spin of the impurity, respectively, and U is the Coulomb repulsion between localized electrons (for simplicity, we assume identical inter- and intraorbital interactions). Finally, $J \geqslant 0$ is the strength of Hund's rule coupling.

Energy levels E_f for an isolated impurity with $J \ll U \sim \epsilon_f$ are presented in Fig. 1(a). The GS sextet belongs to the sector $N_f = 2$ if $1 \leqslant \epsilon_f/U \leqslant 2$, and can be broken into subspaces with total spins $S_f = 0$ and $S_f = 1$ each containing three states, as shown in Table I.

We derive the Kondo Hamiltonian via a generalized Schrieffer-Wolff transformation [28], $\tilde{H} = e^{\mathcal{S}} H e^{-\mathcal{S}}$, with the generator

$$\mathcal{S} = \frac{v}{\sqrt{N}} \sum_{k a \sigma E_f' E_f} \left(\frac{c^\dagger_{k\sigma} P(E_f') f_{a\sigma} P(E_f)}{E_f' - E_f} - \text{H.c.} \right), \quad (1)$$

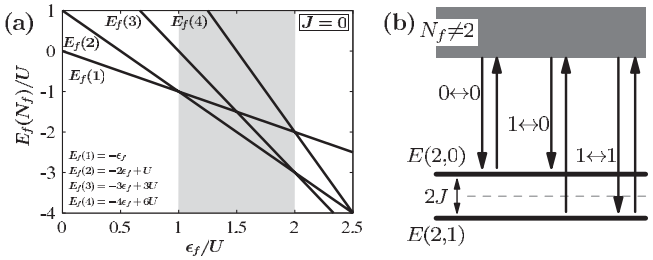

FIG. 1. Impurity states in the two-orbital Anderson impurity model. (a) Energies $E_f(N_f)$ vs localized level depth ϵ_f in the absence of Hund's rule coupling (the S_f argument can be omitted when $J = 0$). In the shaded region the GS has exactly $N_f = 2$ electrons. (b) Fluctuations $S_f \leftrightarrow S_f'$ in the $N_f = 2$ GS due to hybridization with conduction electrons which contribute to the transformation (1).

where E_f in the sum denotes the full set of quantum numbers $\{N_f S_f S_f^z\}$ corresponding to a level with energy $E_f(N_f, S_f)$, and $P(E_f) = |N_f S_f S_f^z\rangle\langle N_f S_f S_f^z|$ is the projector on this multiplet. In writing Eq. (1) we took the hybridization $v_{pa} = v$ to be independent of momentum (as is usually done in deriving the Kondo Hamiltonian [1]) and orbital index [29], and assumed that $t, v^2/U, v \ll U, \epsilon_f$ in order to omit the conduction electron bandwidth (and Hund interaction) in the denominators.

The transformation (1) decomposes the $N_f = 2$ sextet into a doublet $\{|00\rangle, |s\rangle\}$ and a quartet $\{|10\rangle, |1 \pm 1\rangle, |a\rangle\}$. These subspaces have different parities with respect to interchange of the orbitals (see Table I) and are not coupled by hybridization with the conduction band. The resulting six-level "Kondo" Hamiltonian is a direct sum $H_K = H_4 \oplus H_2$. The term $H_2 \sim (1 + \sigma^x) n_0^c$ has an Ising structure (n_0^c is the conduction electron density at the impurity site), and does not involve either spin flips in the conduction channel or transitions between impurity orbital states. Therefore it is irrelevant for the Kondo physics.

The block H_4 contains coupling of the $S_f = 1$ triplet as well as the singlet $|a\rangle$ to the spin of conduction electrons, and can be straightforwardly generalized to the lattice

$$H_4 = \sum_{k\sigma} \xi_k c^\dagger_{k\sigma} c_{k\sigma} + 2J_K \sum_i \Sigma_i s_i^c - 2J \sum_i \Sigma_i A_i. \quad (2)$$

Here $\xi_k = \varepsilon_k - \mu_c$, $s_i^c = (1/2) c^\dagger_{i\alpha} \sigma_{\alpha\beta} c_{i\beta}$, $i = x_i$ denotes lattice sites, the Kondo coupling is $J_K = 2v^2 U/[(\epsilon_f - U)(2U - \epsilon_f)] > 0$, chemical potential μ_c controls the

TABLE I. GS multiplet for an isolated impurity with $N_f = 2$. $|0_f\rangle$ denotes a state with no local fermions.

$S_f = 0$, $E_f = -2\epsilon_f + U + J$	$S_f = 1$, $E_f = -2\epsilon_f + U - J$
$\|00\rangle = (1/\sqrt{2})(f^\dagger_{1\uparrow} f^\dagger_{2\downarrow} - f^\dagger_{1\downarrow} f^\dagger_{2\uparrow})\|0_f\rangle$	$\|1, +1\rangle = f^\dagger_{1\uparrow} f^\dagger_{2\uparrow} \|0_f\rangle$
$\|s\rangle = (1/\sqrt{2})(f^\dagger_{1\uparrow} f^\dagger_{1\downarrow} + f^\dagger_{2\uparrow} f^\dagger_{2\downarrow})\|0_f\rangle$	$\|1, -1\rangle = f^\dagger_{1\downarrow} f^\dagger_{2\downarrow} \|0_f\rangle$
$\|a\rangle = (1/\sqrt{2})(f^\dagger_{1\uparrow} f^\dagger_{1\downarrow} - f^\dagger_{2\uparrow} f^\dagger_{2\downarrow})\|0_f\rangle$	$\|1, 0\rangle = (1/\sqrt{2})(f^\dagger_{1\uparrow} f^\dagger_{2\downarrow} + f^\dagger_{1\downarrow} f^\dagger_{2\uparrow})\|0_f\rangle$

conduction band filling, and the last term describes singlet-triplet level splitting due to Hund's interaction [30].

The vectors $\boldsymbol{\Sigma}_i$ and $\boldsymbol{A}_i$ are spin-1/2–like objects ($\boldsymbol{\Sigma}_i^2 = \boldsymbol{A}_i^2 = 3/4$) that generate two independent (commuting) $su(2)$ algebras. They can be expressed in terms of the on-site Hubbard operators $X_i^{M,a} = |1M\rangle\langle a|$, $X_i^{a,M} = (X_i^{M,a})^\dagger$, and $X_i^{M'M} = |1M'\rangle\langle 1M|$ as

$$\begin{aligned}
&\boldsymbol{\Sigma}_i = (\boldsymbol{S}_i + \boldsymbol{T}_i)/2, \quad \boldsymbol{A}_i = (\boldsymbol{S}_i - \boldsymbol{T}_i)/2, \\
&S_i^+ = \sqrt{2}(X_i^{0,-1} + X_i^{1,0}), \quad T_i^+ = \sqrt{2}(X_i^{1,a} - X_i^{a,-1}), \\
&S_i^z = X_i^{1,1} - X_i^{-1,-1}, \quad T_i^z = -(X_i^{0,a} + X_i^{a,0})
\end{aligned} \tag{3}$$

with $S_i^- = (S_i^+)^\dagger$ and $T_i^- = (T_i^+)^\dagger$. By construction $\boldsymbol{S}_i$ has matrix elements only within the spin-triplet sector and reduces to the $S = 1$ spin operator in the limit of large J, while $\boldsymbol{T}_i$ contains transitions between singlet and triplet local orbital states. Since the Hubbard operators satisfy the constraint $X^{a,a} + \sum_M X^{M,M} = 1$, and $X_i^{a,a} = \boldsymbol{T}_i^2/3 - \boldsymbol{S}_i^2/6$ and $\sum_M X_i^{M,M} = \boldsymbol{S}_i^2/2$, the last term in Eq. (2) can be written up to a constant as $2J\sum_i n_i^a$, where n_i^a is the occupation of each singlet state, and hence is the Hund's energy cost. From Eq. (2) it follows that spin of conduction electrons couples not only to the impurity spin $\boldsymbol{S}$ but also to the orbital component $\boldsymbol{T}$. This interaction can be viewed as a special spin-orbit term originating from many-body correlations.

In the following we will call a state $|\psi_0\rangle$ orbitally ordered if $\langle\psi_0|\boldsymbol{T}_i|\psi_0\rangle \neq 0$, and introduce the local operators for an f-electron ion $\tau_i^\mu = (1/2)\sigma_{\alpha\beta}^\mu f_{i,1\alpha}^\dagger f_{i,2\beta} +$ H.c. which generate transitions between the two orbitals. One can easily verify that within the quartet subspace $\boldsymbol{T}_i = \boldsymbol{\tau}_i$.

Operators $\boldsymbol{S}_i$ and $\boldsymbol{T}_i$ generate an $so(4)$ algebra [31], and commutativity of $\boldsymbol{\Sigma}_i$ and $\boldsymbol{A}_i$ reflects the decomposition $so(4) = su(2) \otimes su(2)$. The orbital component $\boldsymbol{T}$ is analogous to the Runge-Lenz vector in the hydrogen atom [32]. This hidden $so(4)$ structure of Eq. (2) is distinct from the explicit $SU(M)$ symmetry of the multiorbital Coqblin-Schrieffer Hamiltonian [33]. A model similar to Eq. (2) with a single impurity arises in the context of Kondo tunneling through quantum dots [32,34–36], but to our knowledge has never been applied to KLs.

Orbitally ordered heavy-fermion state in the two-orbital KL model.—There are two important observations about the Hamiltonian (2): (i) despite the GS of each f-electron site being an $S_f = 1$ triplet, the conduction electrons are coupled to a spin-1/2 object $\boldsymbol{\Sigma}_i$, and (ii) the Hund's interaction is the only term that involves $\boldsymbol{A}_i$, and therefore cannot be neglected. Equation (3) implies that both of these operators act on physical spin and orbital degrees of freedom of the f-electron ions. In the fully Kondo screened state $|\mathrm{HF}_{c\Sigma}\rangle$, $\boldsymbol{\Sigma}$'s form a singlet with the conduction band, and any magnetically or orbitally ordered state corresponds to an ordering of $\boldsymbol{A}$ pseudospins. While magnetic order may persist outside of the HF regime, the orbital order exists only inside the Kondo phase.

Indeed, if we treat $\boldsymbol{\Sigma}$ and $\boldsymbol{A}$ as classical vectors, Kondo screening does not occur, and Eq. (2), with $J \geqslant 0$, describes a double-exchange model [37]. The pseudospins $\boldsymbol{\Sigma}_i$ form a spiral whose precise shape depends on the conduction band filling, ratio J_K/t, and lattice topology [38]. The vectors $\boldsymbol{A}_i$ will follow exactly the same spiral due to the ferromagnetic Hund's coupling. Since $\boldsymbol{\Sigma}_i$ and $\boldsymbol{A}_i$ are always locally parallel, the situation is the same as if J were infinite, i.e., at each site $S_f = 1$ with no admixture of singlet component, which is equivalent to having a pure spin-1 spiral without orbital order. This is not surprising because even for an infinitesimal J the local triplet state has a lower energy than the singlet. The above analysis shows that a nontrivial orbital order inevitably frustrates the (ferromagnetic) Hund term in Eq. (2). This local frustration arises because quantum fluctuations associated with the Kondo screening dynamically restore orbital degeneracy by allowing $\boldsymbol{\Sigma}_i$ to form a singlet with the conduction band. Below we focus on the regime $J \ll J_K$ to demonstrate how the competition between the Kondo effect and Hund's rule coupling leads to a long-range orbital order.

Returning to the quantum case, for $J = 0$ the fields $\boldsymbol{A}_i$ decouple and the GS of Eq. (2) is macroscopically degenerate $|\psi_0(\{A_i^z\})\rangle = |\mathrm{HF}_{c\Sigma}\rangle \otimes |\{A_i^z\}\rangle$, where $|\{A_i^z\}\rangle$ is one of 2^N states which characterize the free $\boldsymbol{A}_i$ pseudospins. We describe the heavy fermion state $|\mathrm{HF}_{c\Sigma}\rangle$ using the hybridization mean-field approach (HMF) [2] with pseudofermion representation $\boldsymbol{\Sigma}_i = (1/2)h_{i\alpha}^\dagger \boldsymbol{\sigma}_{\alpha\beta} h_{i\beta}$, and self-consistently determine the hybridization order parameter $\chi_0 = (1/\sqrt{2})\langle \mathrm{HF}_{c\Sigma}|c_{i\uparrow}^\dagger h_{i\uparrow} + c_{i\downarrow}^\dagger h_{i\downarrow}|\mathrm{HF}_{c\Sigma}\rangle$. The heavy quasiparticle dispersion becomes $E_{\boldsymbol{k}\tau} = (1/2)(\xi_{\boldsymbol{k}} - \mu_h) + (\tau/2)R_{\boldsymbol{k}}$, with $R_{\boldsymbol{k}} = [(\xi_{\boldsymbol{k}} + \mu_h)^2 + (1/2)(3J_K\chi_0)^2]^{1/2}$ and $\tau = \pm 1$. The h-fermion chemical potential μ_h enforces the constraint $(1/N)\sum_{i\alpha}\langle \mathrm{HF}_{c\Sigma}|h_{i\alpha}^\dagger h_{i\alpha}|\mathrm{HF}_{c\Sigma}\rangle = 1$. The canonical transformation to the quasiparticle states $\gamma_{\boldsymbol{k}\tau\sigma}$ is given by $c_{\boldsymbol{k}\sigma} = \cos(\rho_{\boldsymbol{k}}/2)\gamma_{\boldsymbol{k},+,\sigma} - \sin(\rho_{\boldsymbol{k}}/2)\gamma_{\boldsymbol{k},-,\sigma}$ and $h_{\boldsymbol{k}\sigma} = \sin(\rho_{\boldsymbol{k}}/2)\gamma_{\boldsymbol{k},+,\sigma} + \cos(\rho_{\boldsymbol{k}}/2)\gamma_{\boldsymbol{k},-,\sigma}$, with $\cos\rho_{\boldsymbol{k}} = (\xi_{\boldsymbol{k}} + \mu_h)/R_{\boldsymbol{k}}$ and $\sin\rho_{\boldsymbol{k}} = -3J_K\chi_0/\sqrt{2}R_{\boldsymbol{k}}$.

When $J \ll J_K$ the Hund term in Eq. (2) can be treated within a second-order perturbation theory yielding an RKKY-like effective Hamiltonian acting on the states $|\{A_i^z\}\rangle$

$$\begin{aligned}
H_A &= P_0 V(1 - P_0)(E_0^{\mathrm{HMF}} - H^{\mathrm{HMF}})^{-1}(1 - P_0)VP_0 \\
&= \sum_{ij} J_{ij}^{\mathrm{RKKY}} \boldsymbol{A}_i \boldsymbol{A}_j,
\end{aligned}$$

where P_0 is a projector on the degenerate HF GS manifold and $V = -2J\sum_i \boldsymbol{\Sigma}_i \cdot \boldsymbol{A}_i$. The Fourier transform of the exchange interaction $J_{ij}^{\mathrm{RKKY}} = (1/N)\sum_{\boldsymbol{k}} e^{i\boldsymbol{k}(\boldsymbol{x}_i - \boldsymbol{x}_j)} J_{\boldsymbol{k}}^{\mathrm{RKKY}}$ has the form (for conduction band filling $n^c < 1$)

$$J_k^{\mathrm{RKKY}} = \frac{2J^2}{N}\sum_p \cos^2\frac{\rho_{k+p}}{2}\left[-\frac{\cos^2\frac{\rho_p}{2}\,n^{\gamma}_{p,-}}{E_{k+p,-}-E_{p,-}} + \frac{\sin^2\frac{\rho_p}{2}\,n^{\gamma}_{p,-}}{E_{k+p,-}-E_{p,+}}\right], \tag{4}$$

where $n^{\gamma}_{k\tau}$ is the quasiparticle distribution function.

The semiclassical order of $\boldsymbol{A}_i$ is determined by the minima of J_k^{RKKY}. Figure 2 shows locations of these minima for several electron fillings on a square lattice with nearest-neighbor electron hopping t [$\varepsilon_{\boldsymbol{k}} = -2t(\cos k_x + \cos k_y)$], $N = 10^6$ sites, and $J_K/t = 3$. The incommensurate spiral at low filling gives way to a nearly staggered order and finally, near $n_c = 0.8$, the ordering wave vector becomes small, possibly due to a Nagaoka-like mechanism [39]. Since the HF phase is a singlet ($\langle\psi_0|\boldsymbol{\Sigma}_i|\psi_0\rangle = 0$), $\langle\psi_0|\boldsymbol{A}_i|\psi_0\rangle = -\langle\psi_0|\boldsymbol{T}_i|\psi_0\rangle = \langle\psi_0|\boldsymbol{S}_i|\psi_0\rangle$ [see Eq. (3)].

The state $|\psi_0\rangle$ describes an orbital and real spin spiral of the same pitch. To the zeroth order in J the occupation of the singlet $|a\rangle$ and each of the triplet levels is identical, so the orbital order manifests itself more prominently as a coherent superposition of the triplet and singlet orbital states, rather than the difference in the occupation numbers. For $J \neq 0$ the only globally conserved quantity of the Hamiltonian (2) is the total spin. Hence the symmetry breaking associated with the onset of orbital order is driven by the spin sector. The orbital order appears due to the many-body spin-orbit interaction mentioned earlier.

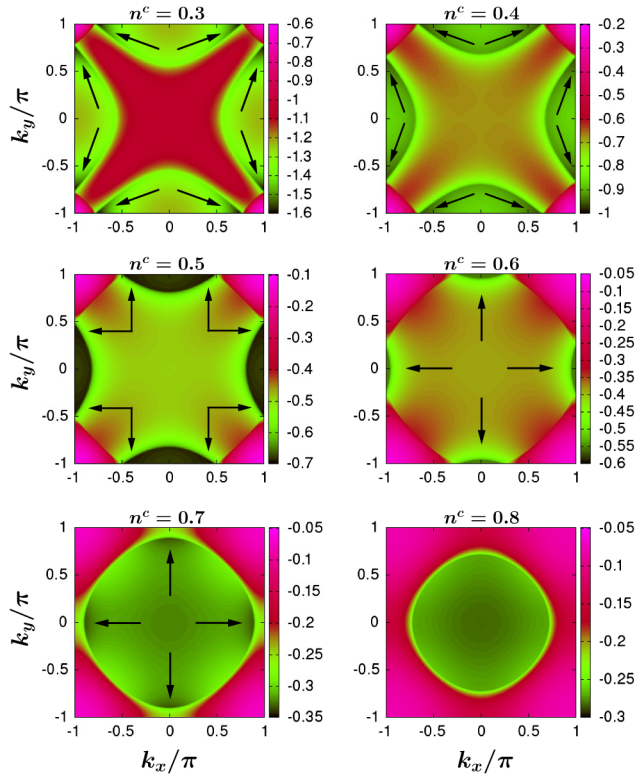

FIG. 2 (color online). Color maps of $tJ_k^{\mathrm{RKKY}}/2J^2$ [Eq. (4)] for several electron fillings n^c. Arrows indicate positions of the minima corresponding to classical spiral states.

Discussion.—In multiorbital heavy-fermion materials the Kondo effect involves both spin and orbital degrees of freedom. Our results highlight the central role of the orbital component, that is the exotic Kondo screening of the pseudo–spin-1/2 object $\boldsymbol{\Sigma}_i$ which stabilizes nontrivial orbital order via a many-body spin-orbit interaction. Formation of the local Kondo singlets competes with Hund's rule coupling by enhancing orbital fluctuations and dynamically restoring orbital degeneracies. This Hund's rule frustration is the fundamental mechanism that leads to a RKKY-like exchange between orbital degrees of freedom and drives their long-range ordering. The RKKY interaction obtained in our work exists due to the Kondo effect, in sharp contrast with theories of the quadrupolar Kondo effect [7,19,20,40], which contend that the RKKY coupling between electric quadrupoles competes with Kondo screening.

Our prediction of coupled orbital and magnetic orders can be tested in neutron and resonant x-ray scattering experiments inside the heavy-electron phase of $5f$ actinide- and Pr-based compounds in which atomic spin-orbit coupling and CEF stabilize an orbitally degenerate local GS. The pitch of the spiral is carrier-density dependent (see Fig. 2) and can be tuned by doping or pressure.

We derived the RKKY interaction (4) for $J \ll J_K$ when the pseudospin $\boldsymbol{\Sigma}$ is Kondo screened while another pseudospin $\boldsymbol{A}$ remains unscreened. For $J \gg J_K$ the singlet $|a\rangle$ is separated by an energy gap from the triplet states, and Eq. (2) reduces to the underscreened $S = 1$ KL model studied in Refs. [41,42]. Using a modified HMF theory [2], it was shown that the system exhibits a coexistence of the Kondo effect and ferromagnetism. In general, the two

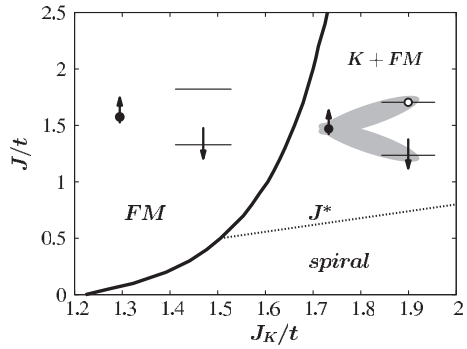

FIG. 3. Schematic phase diagram of the two-orbital KL model. The phases are the local moment ferromagnet (FM), the ferromagnetic Kondo screened phase (K + FM), and the orbital and local-spin spiral state (spiral). The solid line denotes the first-order phase transition. The dotted line $J^*(J_K)$ corresponds to a transition or crossover between K + FM and the spiral state. Shaded ellipses indicate finite Kondo hybridization between conduction electrons (dark circles) and two f orbitals. The system size and electron density are $N = 1600$ and $n^c = 0.6$.

regimes at small and large J are separated by a quantum phase transition because local orbitals realize an irreducible representation of the crystal symmetry group and an orbital order would break at least this discrete symmetry. However, in the presence of the strong spin-orbit interaction the phase transition can become a crossover. Figure 3 presents a schematic phase diagram of the Hamiltonian (2) computed on a square lattice using the mean-field approach of Ref. [34].

The spiral states obtained in our analysis are semiclassical. Since $\boldsymbol{A}_i$ behaves as a spin-1/2 object, quantum fluctuations (especially for frustrated materials [43]) may destabilize static order in favor of quantum disordered phases. In our case quantum effects may lead to even more exotic paraorbital states, e.g., combined orbital and spin liquids, that so far have received little attention.

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