

Destabilization of local magnetic anisotropy in heavy-fermion compounds

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The local magnetic anisotropy of a typical crystalline compound is usually attributed to the combined effect of crystal electric fields and spin-orbit coupling. We show that this simple local picture is transformed in heavy-fermion compounds by the development of coherent electron scattering from local spin degrees of freedom. Provided the dominance of the coherence energy scale over the magnetic energy scale is strong enough, the fractionalization and delocalization of the spins destabilizes their single-ion anisotropy by generating an opposing anisotropy in the exchange. Experimentally, this can manifest as competing splittings in the Curie-Weiss constants and effective moments. We show that in the presence of orthorhombic or tetragonal symmetry, the destabilization of the anisotropy can result in either ferromagnetic or antiferromagnetic order that is perpendicular to the high-temperature easy axis. In the absence of destabilization, we show that the order is more likely to be antiferromagnetic. In agreement with our theory, we also observe that the temperature at which the anisotropy of the uniform magnetic response changes tracks the coherence energy scale in a wide range of actinide and lanthanide compounds.

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I. INTRODUCTION

Heavy-fermion compounds entangle electronic and local magnetic degrees of freedom leading to a rich variety of unusual magnetic phenomena such as high-field superconductivity mediated by spin fluctuations [1–4], with multiple superconducting and highly anisotropic phases, or metamagnetic jumps in the magnetization [5]. The interweaving begins with the formation of the flat heavy-fermion bands as the scattering of electrons from local magnetic moments becomes coherent. The heavy Fermi liquid excitations that emerge coexist with the residual local moments that form at the same time. Although many general questions remain, this “two-fluid” picture is a useful perspective in which to view a number of experimental observations [6–9], as well as theoretical results [10–12].

The rich phenomenology of magnetism in heavy-fermion metals is generally sensitive to the underlying crystal structure, crystal electric fields (CEFs), spin-orbit coupling, or valency of the local-moment ions. Nonetheless, across a wide range of lanthanide and actinide compounds, some robust universal features emerge [4,13,14]. One example is the reorientation of the magnetic easy axis with temperature, which is strongly correlated with the presence of the coherence maximum in the material’s resistivity. The resistivity rises logarithmically under cooling and reaches a maximum at a characteristic temperature T_{coh}^* , before dropping dramatically

as the Kondo scattering of electrons from local spin degrees of freedom becomes coherent (see the bottom panel of Fig. 1). Many heavy-fermion compounds with orthorhombic or tetragonal symmetry that order magnetically below this coherence temperature do so at 90° to the preferred direction of the local moments at high temperatures. The easy axis in the ordered state is thus perpendicular to the easy axis at temperatures above the coherence and exchange energy scales, where it is set by the single-ion anisotropy. Previous works [15,16] have highlighted the competition between local anisotropy and exchange anisotropy as the key driver of the easy-axis reorientation. Why so many heavy-fermion systems happen to have opposing CEF and exchange anisotropies has been a puzzle for some time now. Here, starting from a Kondo lattice model, we show that it is the development of flat heavy-fermion bands that destabilizes the local anisotropy by generating an opposing anisotropy in the exchange.

Before the onset of magnetic order, the destabilization of the single-ion anisotropy can also be signaled by a change in the anisotropy of the uniform magnetic response, which is seen as a crossing of uniform magnetic susceptibilities at a temperature T_{cross} for fields along two perpendicular directions (see the bottom panel of Fig. 1). The connection between T_{coh}^* and T_{cross} was first noticed across a wide range of Yb- and Ce-based Kondo ferromagnets [13], where the susceptibilities cross just before the onset of magnetic order and the crossing is preceded by a coherence maximum at a higher temperature. Here, we widen the universality of this phenomenon by including Eu- and U-based heavy-fermion ferromagnets, antiferromagnets, and paramagnets. Unlike in Ce- or Yb-based materials, in Eu- or U-based heavy-fermion compounds, the crossing of the susceptibilities can take place far away from the onset of magnetic order, or even in its absence, showing that it is unlikely to be caused by the collective effects

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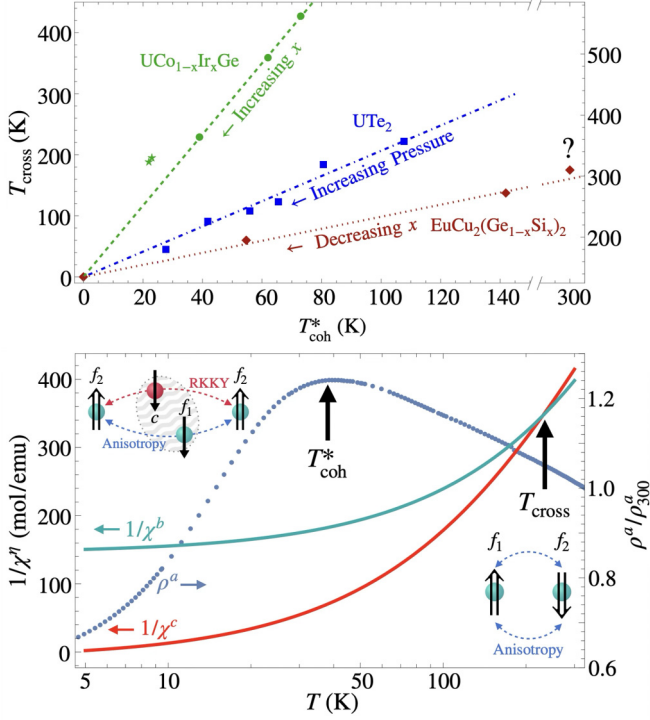


FIG. 1. Top: The susceptibility crossing temperature T_{cross} against the coherence temperature T_{coh}^* for three heavy-fermion compounds. The *direct proportionality* fits suggest that the same energy scale is responsible for the development of coherence and easy-axis reorientation. (For $\text{UCo}_{1-x}\text{Ir}_x\text{Ge}$, the fit does not include the stars, and for $\text{EuCu}_2(\text{Ge}_{1-x}\text{Si}_x)_2$ it does not include the highest T_{cross} , which is a tentative prediction). Bottom: A plot of the a -axis resistivity and modified Curie-Weiss fits of b - and c -axis susceptibilities for $\text{UCo}_{0.98}\text{Ir}_{0.02}\text{Ge}$ identifying T_{coh}^* and T_{cross} . A single-ion anisotropy, which favors antialignment of the two components of the spin-1 moment along the c axis, leads to a lower paramagnetic response along this direction at high temperature. Around T_{coh}^* , when one of the components delocalizes and becomes part of the heavy-fermion sea, the anisotropy interaction becomes nonlocal and a stronger Ruderman–Kittel–Kasuya–Yosida (RKKY) exchange along the c axis is generated. (A detailed description of all the plotted data and any extrapolations used can be found in Table I).

associated with the onset of magnetic ordering. Remarkably, we find that, in these compounds, the crossing and coherence temperatures appear to be *directly proportional*, showing that the same energy scale is responsible for both [see Fig. 1 (top)], with a detailed description of all the plotted data given in Table I].

We find that single-ion anisotropy is most likely to be destabilized in heavy-fermion compounds that display more itinerant rather than localized magnetism. They are characterized by clear resistivity maxima at a temperature T_{coh}^* that is significantly higher than the temperature at which potential magnetic order sets. As shown in Table I, they also often possess higher Sommerfeld coefficients and smaller ordered moments, again pointing to a greater degree of itinerancy. For all the compounds in Table I, if T_{coh}^* is significantly higher (at least three times) than the temperature at which magnetic order sets in, the uniform susceptibilities cross and

the magnetic order (if present) is along the high-temperature hard axis. This therefore appears to be the most predictive experimental marker of local anisotropy destabilization. As the coherence and RKKY energy scales get closer, we show that T_{cross} approaches the magnetic ordering temperature before the susceptibility crossing vanishes entirely. When magnetic order takes place in the presence of anisotropy destabilization, it can be either ferromagnetic or antiferromagnetic. In the absence of destabilization, antiferromagnetic order appears to be more common.

The universal features of magnetism in heavy-fermion metals are not limited to the reorientation of the easy axis. In U- and Eu-based magnets, we notice that the susceptibility crossing is often linked to the presence of a maximum in the temperature dependence of the susceptibility along the axis that had the stronger magnetic response at high temperatures [17–22]. This so-called hard-axis maximum and the following decay of the hard axis susceptibility with cooling are in turn connected to a metamagnetic transition when a high magnetic field is applied along the axis at lower temperatures [17–19,22]. In fact, looking at all the heavy-fermion metals that we have surveyed (see Table I), it appears that a metamagnetic transition in a high field applied along the hard axis implies a maximum in the susceptibility along this axis at a higher temperature, which in turn implies that it is the easy axis at an even higher temperature.

In some compounds, e.g., URhGe [23], coherence develops at a temperature T_{coh}^* that is much higher than the temperature at which fairly large moments order. This points to the underscreened nature of the underlying Kondo lattice, where in spite of the Kondo energy scale T_K being much higher than the RKKY energy scale T_{RKKY} , there are not enough conduction electrons to fractionalize the local spin degrees of freedom entirely and a so-called residual moment remains.

Motivated by the above observation, we present a generalization of the standard large- N Read-Newns theory to the underscreened Kondo lattice. We will focus on the $S = 1$ case.

II. MODEL

The underscreened $S = 1$ Kondo lattice with a generic single-ion anisotropy, which is endowed to the spins through the combined effect of CEF and spin-orbit coupling, is described by the following Hamiltonian:

$$H = J_K \sum_i \mathbf{S}(\mathbf{r}_i) \cdot \mathbf{s}(\mathbf{r}_i) + \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}} c_{\sigma}^{\dagger}(\mathbf{k}) c_{\sigma}(\mathbf{k}) + \sum_{\eta i} D^{\eta} (S^{\eta}(\mathbf{r}_i))^2 - \sum_i \mathbf{h}_i \cdot (\mathbf{S}(\mathbf{r}_i) + \mathbf{s}(\mathbf{r}_i)), \quad (1)$$

where $J_K > 0$ is the Kondo coupling between local spin $\mathbf{S}_i$ and the conduction electron spin $\mathbf{s}_i$, σ denote the up/down spin indices, $c_{\sigma}(\mathbf{k})$ are the usual conduction electron annihilation operators, and $\mathbf{h}$ is an external magnetic field. For simplicity, we have assumed that electrons and spins occupy the same Bravais lattice sites, and $\epsilon_{\mathbf{k}}$ is the dispersion of the single conduction band with bandwidth W . Unless stated otherwise, we will be using a constant density of states $\rho = 1/W$. $D^{x,y,z}$ is a general orthorhombic single-ion anisotropy for spin-1, and we have set its average value to zero. We will work away from

TABLE I. A table of the coherence temperature T_{coh}^* defined as the maximum in resistivity; T_{cross} , the temperature of the susceptibility crossing; $T_{\text{max}}^{\text{hard}}$, the temperature of the maximum in the low-temperature hard-axis susceptibility; T_C (K) or T_N (K), the Curie or Néel temperature; x , the doping fraction; P , pressure (GPa); orien., the orientation of the magnetic moment; HTEA, the high-temperature easy axis; μ (μ_B), the size of the ordered moment; and γ ($\text{mJ mol}^{-1} \text{K}^{-2}$), the Sommerfeld coefficient. NM, nonmagnetic; NR, not resolved; and prin., principally. The superscript on T_{coh}^* values indicates the axis for which the resistivity measurement was taken. Otherwise, a polycrystalline sample was used.

Compound	P (GPa)	T_{coh}^* (K)	T_{cross} (K)	$T_{\text{max}}^{\text{hard}}$ (K)	T_N (K)	T_C (K)	Orien.	HTEA	μ (μ_B)	γ ($\text{mJ mol}^{-1} \text{K}^{-2}$)
UTe ₂ (ortho.) [17,24,25]	0	108 ^a _e	222	37			NM	<i>b</i>		120
Crossing $b \rightarrow a$	0.6	81 ^a _e	184	31			NM	<i>b</i>		–
Metamagnetism observed	0.9	65 ^a _e	123	24			NM	<i>b</i>		–
along b at ambient pressure	1.1	56 ^a _e	108	22			NM	<i>b</i>		–
	1.4	42 ^a _e	91	16			NM	<i>b</i>		–
	1.7	28 ^a _e	45		3		<i>a</i> ?	<i>b</i>	>0.3	–
Compound	x	T_{coh}^* (K)	T_{cross} (K)	$T_{\text{max}}^{\text{hard}}$ (K)	T_N (K)	T_C (K)	Orien.	HTEA	μ (μ_B)	γ ($\text{mJ mol}^{-1} \text{K}^{-2}$)
UCo _{1-x} Ir _x Ge (ortho.) [18,26,27]	0.00	74 ^a	427 ^b			2.5	<i>c</i>	<i>b</i>	0.07	60
Crossing $b \rightarrow c$	0.02	63 ^a	359 ^b				NM	<i>b</i>		55
Metamagnetism observed	0.07	40 ^a	229				NM	<i>b</i>		40
for $x > 0.84$ along b	0.24	24 ^a	187	35			NM	<i>b</i>		56
	0.40	24 ^a	194	30			NM	<i>b</i>		65
	0.67	15 ^a	45	25			NM	<i>b</i>		69
	0.84	25 ^a	32	13	10.2		prin. <i>c</i>	<i>b</i>	–	40
	0.89	31 ^c	27	18	12.2		prin. <i>c</i>	<i>b</i>	–	30
	0.92	29 ^c	36	19	14.4		prin. <i>c</i>	<i>b</i>	–	20
	1.00	32 ^c	21	29	16.5		prin. <i>c</i>	<i>b</i>	0.36	19
URh _{1-x} Ir _x Ge (ortho.) [3,4,19,23,28]	0.00	156	97 ^f			9.5	<i>c</i>	<i>b</i> ^f	0.43	163
Crossing $b \rightarrow c$	0.14	–	69 ^b			9.1	<i>c</i>	<i>b</i>	0.39	160
Metamagnetism observed	0.43	–	34 ^b			6.2	<i>c</i>	<i>b</i>	0.24	175
along b	0.45	–	45 ^b	~11		3.9	<i>c</i>	<i>b</i>	–	120
	0.58	–	34 ^b	12	7.0		<i>c</i>	<i>b</i>	–	70
	1.00	32 ^c	21	29	16.5		prin. <i>c</i>	<i>b</i>	0.36	19
EuCu ₂ (Ge _{1-x} Si _x) ₂ (tetra.) [20,29,30]	≤0.6	0 ^a _c	0 ^d		18.8			NR	≥5.3	–
Crossing $a \rightarrow c$	0.7	55 ^a	~60 ^d		15.5		<i>c</i> ?	NR	<5.3	191
	0.8	142 ^a	138				NM	<i>a</i>		137
	1.0	300 ^a	~320?	~230			NM	<i>a</i>		25
UIr ₂ Si ₂ (tetra.) [21]		97 ^c	60	6	5.5		<i>c</i>	<i>a</i>	0.1	292
Crossing $a \rightarrow c$,										
UPt ₂ Si ₂ (tetra.) [31,32]		176 ^a	76.5		32		<i>c</i>	<i>a</i>	2.5	–
Crossing $a \rightarrow c$										
Metamagnetism observed along a										
UIrSi ₃ (tetra.) [33,34]		224 ^c	93		41.7		<i>c</i>	<i>a</i>	–	31
Crossing $a \rightarrow c$										
EuNi ₂ P ₂ (tetra.) [22]		135 ^a	76	46			NM	<i>ab</i>		93
Crossing $ab \rightarrow c$										
Metamagnetism observed along ab										
YbNi ₄ P ₂ (tetra.) [13,35,36]		19 ^c	0.16			0.15	<i>ab</i>	<i>c</i>	<0.05	2000
Crossing $\parallel c \rightarrow \perp c$										
CeRuPO (tetra.) [13,37–39]		41 ^c	15 ^a			15	<i>c</i>	<i>ab</i>	0.3	–
Crossing $\perp c \rightarrow \parallel c$										
Yb(Rh _{0.75} Co _{0.25}) ₂ Si ₂ (tetra.)		47 ^c	1.3 ^a			1.3	<i>c</i>	<i>ab</i>	0.1	850
Crossing $\perp c \rightarrow \parallel c$ [13,40–42,44]										
YbIr ₃ Ge ₇ (rhomb.) [13,47]		7 ^c	2.9			2.6	<i>ab</i>	<i>c</i>	0.05	–
Crossing $\parallel c \rightarrow \perp c$										
UGe ₂ (ortho.) [4]		70	0 ^d			52	<i>a</i>	<i>a</i>	1.48	34

TABLE I. (Continued.)

Compound	P (GPa)	T_{coh}^* (K)	T_{cross} (K)	$T_{\text{max}}^{\text{hard}}$ (K)	T_N (K)	T_C (K)	Orien.	HTEA	μ (μ_B)	γ (mJ mol $^{-1}$ K $^{-2}$)
UNiSb $_2$ (tetra.) [43,44]		209	0 ^d		175		c	c	—	—
UCu $_2$ Si $_2$ (tetra.) [45]		254	0 ^d		106	100	c	c	1.75	20
USb $_2$ (tetra.) [46]		80	0 ^d		200		c	c	1.9	27

^aThe crossing occurs less than 1 K above the Curie temperature.

^b T_{cross} determined from mCW fits.

^cNo coherence maximum observed.

^dNo clear susceptibility crossing observed. For EuCu $_2$ (Ge $_{1-x}$ Si $_x$) $_2$ with $x = 0.7$, the crossing is just about discernible, but the HTEA cannot be resolved.

^eThe coherence temperatures were determined from linear interpolations of resistivity measurements at different pressure values.

^fAn almost isotropic and constant term has been removed from the susceptibilities.

the insulating limit, and unless specified otherwise, just below half filling.

A. Large- N treatment

We write the $S = 1$ local moments in the Schwinger fermion representation [12,48],

$$S^\eta(\mathbf{r}_i) = \sum_{a=1,2} S_a^\eta(\mathbf{r}_i) = \frac{1}{2} \sum_{\sigma\sigma'} f_{a\sigma}^\dagger(\mathbf{r}_i) \sigma_{\sigma\sigma'}^\eta f_{a\sigma'}(\mathbf{r}_i), \quad (2)$$

where $a = 1, 2$ index the two spin-1/2 fermions, σ, σ' denote the up/down spin indices, σ^η are Pauli matrices with $\eta \in \{x, y, z\}$, and $f_{a\sigma}(\mathbf{r}_i)$ obey the usual fermionic anticommutation relations. We will also promote the $SU(2)$ spin degrees of freedom to $SU(N)$. This can be done in a number of ways, and we will choose to have $N/2$ replicas of spin-up states and an equal number of spin-down states on each lattice site, with half of them occupied, so that we are connected most straightforwardly to the physical $N = 2$ limit. N will be the control parameter for the large- N theory. The Hamiltonian can be written (up to a constant) as

$$H_N = \frac{J_K}{N} \sum_{\alpha\alpha\sigma\beta\sigma'} f_{\alpha\alpha\sigma}^\dagger(\mathbf{r}_i) f_{\alpha\beta\sigma'}(\mathbf{r}_i) c_{\beta\sigma'}^\dagger(\mathbf{r}_i) c_{\alpha\sigma}(\mathbf{r}_i) + \sum_{\mathbf{k}\alpha\sigma} \epsilon_{\mathbf{k}} c_{\alpha\sigma}^\dagger(\mathbf{k}) c_{\alpha\sigma}(\mathbf{k}) + \sum_{i\alpha\eta} D^\eta \left(\sum_a S_a^\eta(\mathbf{r}_i) \right)^2 - \sum_{i\eta\alpha} h_i^\eta \left(\sum_a S_a^\eta(\mathbf{r}_i) + s_\alpha^\eta(\mathbf{r}_i) \right),$$

where

$$S_{a\alpha}^\eta(\mathbf{r}_i) = \frac{1}{2} \sum_{\sigma\sigma'} f_{a\alpha\sigma}^\dagger(\mathbf{r}_i) \sigma_{\sigma\sigma'}^\eta f_{a\alpha\sigma'}(\mathbf{r}_i), \quad (3)$$

and α, β index the spin up/down replicas. The enlargement of the Hilbert space through the Schwinger fermion representation comes with several constraints and associated gauge symmetries:

$$\hat{n}_i^f := \sum_{a=1,2} \hat{n}_{ai}^f = \sum_{\alpha\alpha\sigma} f_{\alpha\alpha\sigma}^\dagger(\mathbf{r}_i) f_{\alpha\alpha\sigma}(\mathbf{r}_i) = N, \\ \hat{T}_i^\eta := \frac{1}{2} \sum_{\alpha\alpha'\sigma\sigma'} f_{\alpha\alpha\sigma}^\dagger(\mathbf{r}_i) \tau_{\alpha\alpha'}^\eta f_{\alpha'\sigma}(\mathbf{r}_i) = 0, \quad (4)$$

where $\tau^{x,y,z}$ are Pauli matrices, which now act on the two-dimensional a subspace. We can understand the constraints on $\hat{n}_i^f$ and $\hat{T}_i^z$ as together ensuring that there are $\hat{n}_{1i}^f = \frac{N}{2} f_1$ fermions and $\hat{n}_{2i}^f = \frac{N}{2} f_2$ fermions on each lattice site. The constraints $\hat{T}^{x,y}$ couple the $SU(N)$ f_1 and f_2 spins, which for $N = 2$ project out the singlet state and correspond to an infinite Hund's coupling between f_1 and f_2 moments. The constraints generate the $U(2) \sim U(1) \times SU(2)$ group of local gauge transformations.

Read-Newns large- N theory introduces the hybridization field

$$\begin{pmatrix} \hat{V}_{1i} \\ \hat{V}_{2i} \end{pmatrix} = \frac{1}{N} \sum_{\alpha\sigma} \begin{pmatrix} f_{1\alpha\sigma}^\dagger(\mathbf{r}_i) c_{\alpha\sigma}(\mathbf{r}_i) \\ f_{2\alpha\sigma}^\dagger(\mathbf{r}_i) c_{\alpha\sigma}(\mathbf{r}_i) \end{pmatrix}. \quad (5)$$

In the saddle-point approximation, the local $U(2)$ gauge symmetry can be used to set $\langle \hat{V}_{2i} \rangle = 0$ and $\langle \hat{V}_{1i} \rangle > 0$. f_2 moments thus decouple from conduction electrons at the level of large- N mean-field theory [48].

B. Effective Hamiltonian for the residual local moments

$\mathcal{O}(1/N)$ corrections to the saddle-point approximation break the macroscopic degeneracy of the f_2 moment subspace [48]. This can be captured with the following variational ansatz for the low-temperature state:

$$|\Psi\rangle = \prod_{\mathbf{k}\alpha\sigma\nu} (u_{\mathbf{k}\sigma}^\nu c_{\alpha\sigma}^\dagger(\mathbf{k}) + v_{\mathbf{k}\sigma}^\nu f_{1\alpha\sigma}^\dagger(\mathbf{k})) |0\rangle \otimes \prod_{\alpha i} |\psi_{2\alpha i}\rangle, \quad (6)$$

where ν indexes two heavy-fermion bands and $|\psi_{2\alpha i}\rangle$ is the state of one of the f_2 moments. By employing the usual static approximation for the f_2 moments, calculating $\langle \Psi | H_N | \Psi \rangle$ with $\{u_{\mathbf{k}\sigma}^\nu, v_{\mathbf{k}\sigma}^\nu\}$ chosen to minimize the expectation value subject to global constraints on the number of conduction and f_1 fermions, we can derive the effective RKKY interaction between f_2 moments that is mediated by the heavy fermions. Expanding to quadratic order in $1/N$ and D^η and linear order in an external magnetic field h_i^η , we obtain

$$H_{\text{eff}} = -\frac{1}{2} \sum_{i \neq j, \eta} \mathcal{J}^\eta(\mathbf{r}_i - \mathbf{r}_j) S_2^\eta(\mathbf{r}_i) S_2^\eta(\mathbf{r}_j) - \sum_{\mathbf{q}\eta} \tilde{h}^\eta(\mathbf{q}) S_2^\eta(\mathbf{q}), \quad (7)$$

where $S_2^\eta(\mathbf{q}) = \frac{1}{\sqrt{N_s}} \sum_i S_2^\eta(\mathbf{r}_i) e^{-i\mathbf{q} \cdot \mathbf{r}_i}$, $\mathcal{J}^\eta(\mathbf{q}) = \sum_i \mathcal{J}^\eta(\mathbf{r}_i) e^{-i\mathbf{q} \cdot \mathbf{r}_i}$, N_s is the number of lattice sites, and

$$\mathcal{J}^\eta(\mathbf{q}) = \frac{4J_K^2}{N^2} \chi_{cc}(\mathbf{q}) + \frac{8J_K}{N} D^\eta \chi_{cf}(\mathbf{q}) + (2D^\eta)^2 \chi_{ff}(\mathbf{q}),$$

$$\frac{\tilde{h}(\mathbf{q})}{h(\mathbf{q})} = 1 - \frac{2J_K}{N} (\chi_{cc}(\mathbf{q}) + \chi_{cf}(\mathbf{q})) - 2D^\eta (\chi_{ff}(\mathbf{q}) + \chi_{cf}(\mathbf{q})), \quad (8)$$

and we have dropped the replica index in H_{eff} , reverting to the physical case $N = 2$, so that $S_2^\eta(\mathbf{r}_i)$ are $SU(2)$ spin operators. The susceptibilities of the $N \rightarrow \infty$ ground state (f_2 moments completely decouple in this limit) are given by

$$\begin{aligned} \chi_{cc}(\mathbf{q}) &= \frac{1}{2N_s} \sum_{\mathbf{k}\nu\nu'} |u_{\mathbf{k}}^\nu|^2 |u_{\mathbf{k}+\mathbf{q}}^{\nu'}|^2 \frac{n(E_{\mathbf{k}+\mathbf{q}}^{\nu'}) - n(E_{\mathbf{k}}^\nu)}{E_{\mathbf{k}}^\nu - E_{\mathbf{k}+\mathbf{q}}^{\nu'}}, \\ \chi_{cf}(\mathbf{q}) &= \frac{1}{2N_s} \sum_{\mathbf{k}\nu\nu'} \bar{v}_{\mathbf{k}}^\nu v_{\mathbf{k}+\mathbf{q}}^{\nu'} u_{\mathbf{k}}^\nu \bar{u}_{\mathbf{k}+\mathbf{q}}^{\nu'} \frac{n(E_{\mathbf{k}+\mathbf{q}}^{\nu'}) - n(E_{\mathbf{k}}^\nu)}{E_{\mathbf{k}}^\nu - E_{\mathbf{k}+\mathbf{q}}^{\nu'}}, \\ \chi_{ff}(\mathbf{q}) &= \frac{1}{2N_s} \sum_{\mathbf{k}\nu\nu'} |v_{\mathbf{k}}^\nu|^2 |v_{\mathbf{k}+\mathbf{q}}^{\nu'}|^2 \frac{n(E_{\mathbf{k}+\mathbf{q}}^{\nu'}) - n(E_{\mathbf{k}}^\nu)}{E_{\mathbf{k}}^\nu - E_{\mathbf{k}+\mathbf{q}}^{\nu'}}, \end{aligned} \quad (9)$$

where $\chi_{cc}(\mathbf{q})$ ($\chi_{cf}(\mathbf{q})$) gives the magnetic susceptibility of the conduction electrons (f_1 local moments) when a $\mathbf{q}$ -modulated magnetic field is applied isothermally to conduction electrons only. Similarly, $\chi_{ff}(\mathbf{q})$ gives the magnetic susceptibility of the f_1 local moments, when the applied field only couples to them. $E_{\mathbf{k}}^\nu$ is the energy dispersion of the heavy-fermion quasiparticles in band ν and $n(E_{\mathbf{k}}^\nu)$ the corresponding Fermi occupation number. (Expressions for $u_{\mathbf{k}}^\nu$, $v_{\mathbf{k}}^\nu$, and $E_{\mathbf{k}}^\nu$ and an outline of the derivation of the effective Hamiltonian can be found in Appendix A.) In the isotropic limit, our effective Hamiltonian agrees with the one derived in Ref. [48] for an underscreened spin-1 Kondo chain.

III. EASY-AXIS REORIENTATION

Clearly, at temperatures much greater than T_K and T_{RKKY} , where the spin-1 moments are unhybridized and RKKY interactions can be neglected, a hard-axis anisotropy $D^\eta > 0$ along η leads to a smaller magnetic response along this axis. Let us now consider a magnetic modulation with wave vector $\mathbf{q}$ at temperatures below T_K . Assuming for now (see later discussion) that $\chi(\mathbf{q})$ and $\chi(\mathbf{q}) - \bar{\chi}$ are all positive for $T \ll T_K$ (the bar denotes an average of the respective function over the Brillouin zone), the anisotropy of the effective field, $\tilde{h}^\eta(\mathbf{q})$, and the anisotropy of the RKKY exchange $\mathcal{J}^\eta(\mathbf{q}) - \bar{\mathcal{J}}^\eta$ are of opposite sign to first order in D^η . For example, a hard-axis anisotropy $D^\eta > 0$ along η leads to a reduction of the residual effective moment but a reinforcement of RKKY exchange along this direction. We can understand this by looking at how the residual moment polarizes the surrounding heavy Fermi sea. The greater the D^η the stronger the local antialignment induced by the anisotropy interaction between the f_2 and the now delocalized f_1 moments (see the bottom panel of Fig. 1). Locally, this gives a lower effective moment along the direction with higher D^η , and therefore lower paramagnetic response at high temperature, but globally, the long-range RKKY forces are stronger along this direction. The collective

fluctuations generated by RKKY dominate at low temperatures and the higher the D^η the stronger the magnetic response. In particular, a magnetically ordered collinear ground state would have moments along the direction with the highest D^η .

The local and the exchange (for $\mathbf{q} = \mathbf{0}$) anisotropies, respectively, correspond to the anisotropy in the effective moments μ_{eff}^η and Curie-Weiss constants Θ_{CW}^η . Indeed, in agreement with H_{eff} , many of the U-based compounds in Table I have $(\Delta\mu_{\text{eff}}^\eta)/\mu_{\text{eff}} \sim -(\Delta\Theta_{\text{CW}}^\eta)/T_{\text{cross}}$, where Δ takes the difference between the two directions involved in uniform susceptibility crossing and μ_{eff} is the average effective moment for these two directions. The competition between the two anisotropies can lead to a crossing of the uniform susceptibilities at a temperature of

$$\begin{aligned} T_{\text{cross}} &= \frac{J_K^2}{N^2} (\chi_{cc}(\mathbf{0}) - \bar{\chi}_{cc}) + \frac{J_K}{N} \frac{\chi_{cf}(\mathbf{0}) - \bar{\chi}_{cf}}{\chi_{ff}(\mathbf{0}) + \chi_{cf}(\mathbf{0})} \\ &\times \left(1 - \frac{2J_K}{N} (\chi_{cc}(\mathbf{0}) + \chi_{cf}(\mathbf{0})) \right), \end{aligned} \quad (10)$$

in the Weiss mean-field approximation, and to quadratic order in $1/N$ and first nonvanishing order in D^η .

We identify two regimes. In the strong-coupling limit $J_K/W \gtrsim 1/2$, we have to first nonvanishing order in D^η and $1/N$:

$$T_{\text{cross}} \approx \frac{T_K}{N}, \quad (11)$$

where T_K is the Kondo energy scale [49]. Extrapolating to the physical case $N = 2$, we find that the crossing temperature is proportional to the strength of the Kondo interaction; i.e., we should have $T_{\text{cross}} \propto T_{\text{coh}}^*$, which indeed shows excellent agreement with experimental data in a wide range of heavy-fermion compounds as shown in Fig. 1.

In the weak-coupling limit, for any fixed N , as J_K is reduced, the crossing temperature approaches the RKKY energy scale, which we define as $T_{\text{RKKY}} = |\Theta_{\text{CW}}| = (J_K/N)^2 |\chi_{cc}(\mathbf{0}) - \bar{\chi}_{cc}|$, before vanishing entirely. We also find that the sign of the $D^\eta = 0$ Curie-Weiss constant Θ_{CW} changes from positive to negative around $J/W \sim 1/2$ as we transition from the strong to weak coupling regime, suggesting that the ground state is ferromagnetic at strong coupling and antiferromagnetic at weak coupling. The fact that we see this for a constant density of states approximation, where there is no specific underlying lattice, shows that this is a universal phenomenon, which we indeed observe for other lattices (see our later discussion). Figure 2 illustrates the two regimes for both theory and experimentally measured values for $\text{UCo}_{1-x}\text{Ir}_x\text{Ge}$, which spans the range from strongly to weakly coupled as a function of the doping x .

IV. MAGNETIC GROUND STATES

We apply our effective Hamiltonian to the representative case of the simple cubic lattice with nearest-neighbor hopping (similar behavior observed for other orthorhombic lattices) and determine its classical $T = 0$ phases in the weak anisotropy limit $D^\eta \rightarrow 0$ for a range of conduction electron lattice fillings $0.75 < n_c < 1$ and coupling constants $1/6 < J_K/W < 3$. To simplify the discussion of easy-axis reorienta-

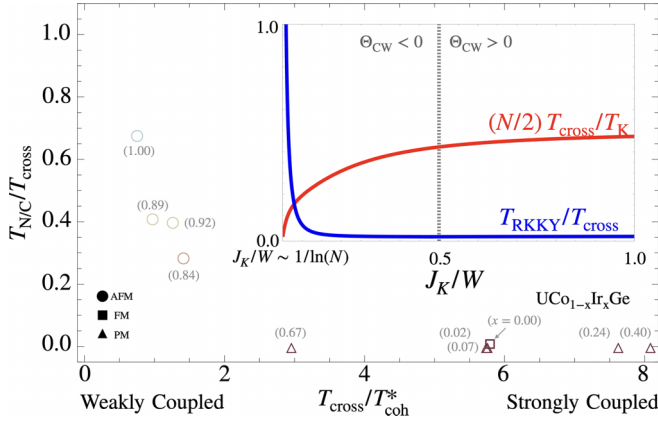


FIG. 2. The ratios $T_{N/C}/T_{\text{cross}}$ and $T_{\text{cross}}/T_{\text{coh}}^*$ plotted for $\text{UCo}_{1-x}\text{Ir}_x\text{Ge}$, where T_{cross} is the uniform susceptibility crossing temperature, T_{coh}^* is the coherence temperature, and $T_{N/C}$ is the Neel (circles) or Curie (squares) temperature depending on the type of magnetic order. Triangles denote lack of magnetic order. Inset: The ratios $T_{\text{RKKY}}/T_{\text{cross}}$ and $(N/2)T_{\text{cross}}/T_K$ plotted against the Kondo coupling strength, J_K/W . There are two regimes: weak coupling at $J_K/W \lesssim 1/2$, where the Curie-Weiss constant is negative and T_{cross} approaches T_{RKKY} before vanishing entirely around $J_K/W \sim 1/\ln N$, and strong coupling at $J_K/W \gtrsim 1/2$, where the Curie-Weiss constant is positive and $T_{\text{RKKY}} \ll T_{\text{cross}} \approx T_K/N$.

tion, we restrict ourselves to simple collinear phases, where the location of the maximum of $\chi_{cc}(\mathbf{q})$ determines the magnetic ordering wave vector $\mathbf{q}$, and if $\chi_{cf}(\mathbf{q}) - \bar{\chi}_{cf}$ is positive (negative) at this wave vector, the magnetic order is along the high-temperature hard (easy) axis.

Our results are shown in Fig. 3 for two representative fillings. We can see that order takes place along the high-temperature hard axis at strong enough Kondo coupling, when the excitation energy for interband particle-hole fluctuations is sufficiently large and intraband fluctuations dominate. $\chi_{cf}(\mathbf{q})$ is manifestly positive in this limit (and $\chi_{cf}(\mathbf{q}) - \bar{\chi}_{cf}$ at the ordering wave vector if $\chi_{cf}(\mathbf{q})$ and $\chi_{cc}(\mathbf{q})$ are maximized at similar wave vectors). Stronger Kondo coupling (at fixed $N \gg 1$ in our theory) corresponds to a greater dominance of the Kondo energy scale over the RKKY energy scale and thus more itinerant magnetism. From Table I, we can indeed see that compounds with higher ratios of $T_{\text{coh}}^*/T_{N/C}$ are more likely to exhibit order along the high-temperature hard axis. A perhaps related phenomenon has been observed in recent DFT + U studies on UAu_2 [50], where more localized 5f moments favored an easy-plane anisotropy, whereas more delocalized ones favored an easy-axis anisotropy.

At weak coupling, interband particle-hole fluctuations dominate. They enforce the antiferromagnetic correlation between conduction and f_1 electrons, and $\chi_{cf}(\mathbf{q}) < 0$ (and $\chi_{cf}(\mathbf{q}) - \bar{\chi}_{cf} < 0$ at the ordering wave vector if $\chi_{cf}(\mathbf{q})$ and $\chi_{cc}(\mathbf{q})$ are extremized at similar wave vectors). Because the indirect band gap is smaller than the direct one, the antiferromagnetic $\mathbf{q} = (\pi/a, \pi/a, \pi/a)$ phase also dominates in this limit. Interband fluctuations are the only allowed ones at $T = 0$ in a Kondo insulator. Here, applying a small field to conduction electrons cannot change the occupancy of up or down heavy fermions and only makes the singlet state

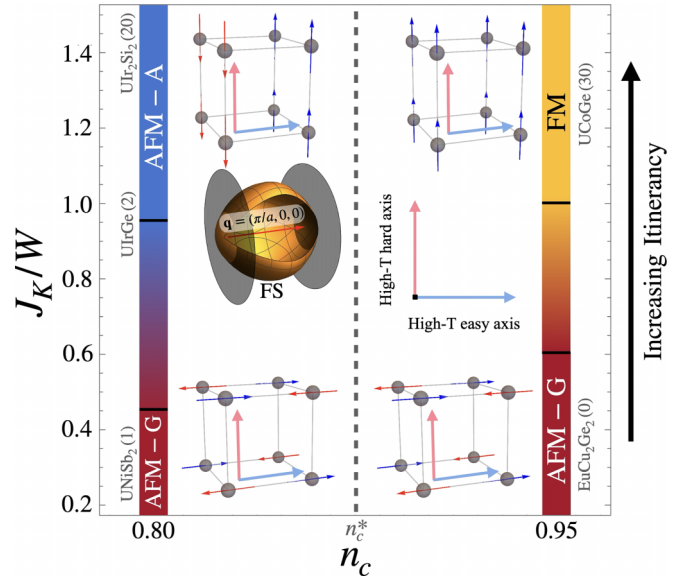


FIG. 3. The ground-state phase diagram of the $S = 1$ simple cubic Kondo lattice in the weak anisotropy limit $D^{\eta} \rightarrow 0$ for two representative values of the electron filling n_c as a function of the Kondo coupling J_K normalized by the bandwidth W . There is an AFM-G phase with spins along the high-temperature easy axis at low coupling and an AFM-A or FM phase with spins along the high-temperature hard axis at large J_K . For each phase, we give a representative compound from Table I and state the ratio of $T_{\text{coh}}^*/T_{N/C}$, where T_{coh}^* is the coherence temperature and $T_{N/C}$ the Neel or Curie temperature. Higher ratios signal more itinerant magnetism. The AFM-A phase relies on nesting. The Fermi surface and the locus of points that are connected by the AFM wave vector to a point of equal energy have been shown. n_c^* denotes the region where the crossing from AFM-A to FM occurs at large J_K . The gaps between phases indicate regions where energy differences between competing phases were smaller than the numerical error and/or there is noncollinear magnetic order.

slightly asymmetric, leading to opposing magnetizations of c and f_1 moments and $\chi_{cf}(\mathbf{q}) < 0$. Kondo insulators' lack of compressibility thus precludes easy-axis reorientation in these systems via our proposed mechanism.

On the other hand, when intraband particle-hole fluctuations close to the Fermi surface dominate, the overriding effect from an external field applied only to conduction electrons is to increase (decrease) the occupancy of up (down) heavy fermions. Because f_1 and c moments share the same z -angular momentum in the heavy-fermion quasiparticle, they acquire the same sign of magnetization and $\chi_{cf}(\mathbf{q}) > 0$.

We have observed that either ferromagnetic or antiferromagnetic magnetic order can take place along the high-temperature hard axis. The latter is favored when the Fermi surface is well nested (see Fig. 3), and is thus lattice and filling specific by nature, and perhaps therefore less common.

V. DISCUSSION

In this section, we discuss the key limitations of our theoretical model and list some of the possible alternative mechanisms that have so far been proposed.

The underscreened spin-1 Kondo lattice model is particularly suited for compounds where two valence f electrons are coupled by a strong Hund's interaction that generates a local spin-1 moment. This might be the situation in uranium-based compounds with a U $5f^2$ valency, and Ref. [51] shows that UTe_2 might actually be close to it. Further, we have assumed that the spin-1 moment is underscreened and a robust residual moment remains after the heavy-fermion state forms. Other compounds will have different valence f -shell configurations (e.g., somewhere between $4f^6$ and $4f^7$ in $\text{EuCu}_2(\text{Ge}_{1-x}\text{Si}_x)_2$) and/or spin-orbit coupling schemes. For example, Ce- and Yb-based compounds with strong spin-orbit coupling are described by an entirely different microscopic Hamiltonian, the Coqblin-Schrieffer model. We leave the detailed analyses of these models to future work, but it poses the following conundrum. Although derived for a specific Kondo lattice model, the mechanism proposed in this work captures the easy-axis reorientation phenomenology seen across a wide variety of heavy-fermion systems. The key ingredient appears to be the two-fluid description of the heavy-fermion system, which models it as a combination of a coherent heavy-fermion fluid and residual local moments. This empirical model has had a lot of success in capturing much heavy-fermion data [7,9]. Although we have shown how such an effective two-fluid model arises for the spin-1 underscreened Kondo lattice, given its wide empirical success, the other microscopic models are likely to give rise to similar effective descriptions.

Easy-axis reorientation can be viewed as yet another manifestation of the two-fluid heavy-fermion state, and we now show how the phenomenon arises naturally within the two-fluid framework. The antiferromagnetic Kondo exchange can lead to a heavy-fermion paramagnet via Kondo hybridization but also to magnetic order of the local moments by generating an RKKY interaction between them. The competition between the two gives rise to the Doniach phase diagram [52]. Kondo hybridization, being an angular momentum conserving process, favors alignment between f moments and conduction electrons during a weak polarization of the heavy-fermion sea. The RKKY interaction, on the other hand, favors their local antialignment. In the two-fluid state, where the residual local moments and the heavy-fermion liquid coexist, f moments are frustrated as they try to simultaneously align and antialign with conduction electrons. This frustration is relieved along the hard axis by a single-ion anisotropy that favors antialignment of the hybridized and local components of the f moment (respectively, f_1 and f_2 in our notation), which, respectively, align and antialign with the conduction electrons. A weak single-ion anisotropy thus stabilizes the coexistence of the heavy-fermion liquid and magnetic order along the hard axis and lowers its ground-state energy with respect to magnetic order along the easy axis.

Another limitation of our model is its restriction to the weak anisotropy regime. Once the anisotropy becomes too strong, it competes with Kondo hybridization and the stabilization mechanism mentioned above does not take place. Indeed, easy-axis reorientation has not been observed in Ce or Yb compounds where anisotropy is too strong [13]. Although the large- N saddle-point approximation is only valid for weak anisotropy, we can increase D^η in the mean-field Hamiltonian in Eq. (A2) and solve the self-consistency equations. The

hybridization field that is responsible for the reduction of the effective moment and the reinforcement of the RKKY exchange collapses once D^η for a given direction reaches a critical value. We revert to free-moment behavior, where the higher the D^η , the lower the susceptibility. This can happen before the hybridization field collapses for the two directions with lower D^η , so there might still be an easy-axis reorientation for these directions. Indeed, for the orthorhombic systems listed in Table I, we find that the two higher susceptibilities cross but not the lowest one (i.e., with highest D^η). YbRhSb [53] at high pressures is the only example of an orthorhombic system that we could find where magnetic order is along the direction with the lowest susceptibility at high temperatures. We note that, although the collapse signals the return of the free-moment state, it is beyond the validity of our perturbative model and we leave the detailed study of the strong anisotropy limit to future work. We should also mention here that Refs. [13,47,54] have highlighted the difficulty with overcoming large CEF anisotropy by an opposite exchange anisotropy and suggested fluctuation-based mechanisms as an alternative explanation for easy-axis reorientation, particularly in compounds with strong CEF anisotropy.

The conundrum of why in so many heavy-fermion systems the anisotropy of the RKKY interaction opposes the CEF anisotropy has been a puzzle for some time now. Spin Hamiltonians with opposing anisotropies in the effective moments (or Landé g factors) and Curie-Weiss constants have been postulated previously and showed good agreement with experimental data (see, for example, Ref. [15]). However, we believe that this is the first time that such a spin Hamiltonian is derived microscopically from a Kondo lattice model. A very recent survey [16], focusing on Ce compounds, also came to the conclusion that easy-axis reorientation (or hard-axis order in the terminology of that work) is the effect of competition between local CEF anisotropy and exchange interaction anisotropy. Without detailed calculations, two possible origins were put forward for the splitting of the Curie-Weiss constants: (1) a difference in the distance between neighboring magnetic ions along different crystallographic directions and (2) the anisotropic Kondo effect (see [55] for an example of the latter). Our proposed mechanism is effectively equivalent to the latter since CEF anisotropy (D^η) renormalizes the effective Kondo coupling making it anisotropic: $J_K^\eta \rightarrow J_K + 2D^\eta$ (neglecting $\mathbf{q}$ dependence). (A similar renormalization was found in the perturbative Renormalisation group (RG) treatment of a single anisotropic Kondo impurity [56].) Importantly, however, our mechanism also explains why local and exchange anisotropies tend to oppose each other in many heavy-fermion systems. It also shows that how their competition leads to easy-axis reorientation at a temperature that tracks the coherence energy scale in the strongly coupled regime. Incidentally, Ref. [16] did not find a strong correlation with the strength of the Kondo effect. However, the size of the local moment entropy contribution was used to ascertain the latter, whereas we show that the key correlation is with the coherence maximum in transport measurements. The difference could be due to the fact that the entropy of the local moments is also affected by single-ion Kondo screening, whereas the resistivity maximum marks the development of coherence

between individual scattering events, which is necessary for the development of opposing exchange anisotropy.

In the strong coupling regime, the direct proportionality between T_{cross} and T_{coh}^* provides direct evidence of the latter being the energy scale that drives the reorientation. In the weak coupling regime, there exists a difficulty in ascribing easy-axis reorientation to the development of coherence, as the coherence and reorientation energy scales can be vastly different. How can we be sure that the reorientation is not just a CEF effect? We propose three universal features that strongly suggest that the reorientation is due to the development of coherence rather than a CEF effect: (1) the reorientation is preceded by a clear coherence maximum in the resistivity at a significantly higher temperature; (2) there are opposing anisotropies in the Curie-Weiss constants and in the effective moments; and (3) the uniform susceptibilities cross just above the magnetic ordering temperature. We should note here that Ce- and Yb-based ferromagnets, with magnetic order along the high-temperature hard axis, always appear to be in the weak coupling regime, with the uniform susceptibilities crossing just before the onset of order. We surmise that this is because in the Coqblin-Schrieffer model, which describes these systems, the local moments are fully screened, giving rise to a vanishing residual moment when the coherence energy scale is much greater than the RKKY energy scale, precluding the strong coupling regime.

VI. CONCLUSION

We have shown that the development of flat bands in heavy-fermion metals can destabilize the single-ion magnetic anisotropy of the local moments, which sets the easy axis at high temperatures, by generating an opposing exchange anisotropy. The destabilization leads to a reorientation of the easy axis with cooling and magnetic order (if it takes place) at 90° to the high-temperature easy axis. The destabilization is mostly likely to occur in more itinerant heavy-fermion compounds that have a clear resistivity maximum at a temperature significantly higher than the magnetic ordering temperature. In agreement with our theory, a wide range of heavy-fermion compounds exhibit a crossing of uniform susceptibilities at a temperature that tracks the coherence energy scale, a trend not previously noticed. Our theoretical analysis follows an extension of the Read-Newns theory to the spin-1 underscreened Kondo lattice, which naturally gives rise to the coexistence of local moment order and Kondo hybridization, even when the energy scale of the latter is much greater.

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crystals grown and studied with the support of the program of Czech Research Infrastructures (Project No. LM2023065).

APPENDIX: DERIVATION OF THE EFFECTIVE HAMILTONIAN IN EQ. (7)

The variational parameters $\{u_{\mathbf{k}\alpha}, v_{\mathbf{k}\alpha}\}$ are chosen to minimize $\langle \Psi | H_N | \Psi \rangle$ subject to the constraints on $\langle \hat{n}_{i\alpha}^f \rangle$ and the total number of conduction electrons, which corresponds to solving

$$\frac{\delta}{\delta u_{\mathbf{k}\alpha}} \left(\langle H_N \rangle - \bar{\lambda}_1 \sum_i \langle \hat{n}_{i1}^f \rangle - \mu \sum_i \langle \hat{n}_i^c \rangle \right) = 0, \quad (\text{A1})$$

where the Lagrange multipliers $\bar{\lambda}_1$ and μ are adjusted to give the expectation values $\langle \hat{n}_{i1}^f \rangle = \frac{N}{2}$ and $\langle \hat{n}_i^c \rangle = \sum_{\alpha\sigma} \langle c_{\alpha\sigma}^\dagger(\mathbf{r}_i) c_{\alpha\sigma}(\mathbf{r}_i) \rangle = \frac{1}{2} n_c N$. n_c is the electron filling. The expectation value to be minimized can be computed using Wick's theorem and is equal to $\langle \Psi | H_{\text{MF}} | \Psi \rangle$, where, up to a constant, $H_{\text{MF}} = H_0 + H_1$ is an appropriately chosen mean-field Hamiltonian with

$$\begin{aligned} H_0 &= \sum_{\mathbf{k}\alpha\sigma} \Psi_{\alpha\sigma\mathbf{k}}^\dagger \begin{pmatrix} \epsilon_{\mathbf{k}} - \mu & -J_K \bar{V}_1 \\ -J_K \bar{V}_1 & \bar{\lambda}_1 \end{pmatrix} \Psi_{\alpha\sigma\mathbf{k}} \\ &\quad + N N_s \left(J_K \bar{V}_1^2 + \frac{1}{2} \mu n_c + \frac{1}{2} \bar{\lambda}_1 \right) - \sum_{i\alpha\eta} h_i^\eta \bar{S}_{2\alpha i}^\eta, \\ H_1 &= \sum_{i\alpha\eta} \left[(J_M \bar{S}_{\alpha i}^\eta + 2D^\eta \bar{S}_{2\alpha i}^\eta - h_i^\eta) \hat{S}_{1\alpha}^\eta(\mathbf{r}_i) \right. \\ &\quad \left. + (J_M \bar{S}_{1\alpha i}^\eta + J_M \bar{S}_{2\alpha i}^\eta - h_i^\eta) \hat{S}_\alpha^\eta(\mathbf{r}_i) - J_M \bar{S}_{\alpha i}^\eta \bar{S}_{1\alpha i}^\eta \right] \\ &\approx \sum_{i\alpha\eta} \left[(2D^\eta \bar{S}_{2\alpha i}^\eta - h_i^\eta) \hat{S}_{1\alpha}^\eta(\mathbf{r}_i) + (J_M \bar{S}_{2\alpha i}^\eta - h_i^\eta) \hat{S}_\alpha^\eta(\mathbf{r}_i) \right], \end{aligned} \quad (\text{A2})$$

where $\Psi_{\alpha\sigma\mathbf{k}}^\dagger = (c_{\alpha\sigma}^\dagger(\mathbf{k}), f_{1\alpha\sigma}^\dagger(\mathbf{k}))$, $\bar{V}_1 = \langle \hat{V}_1 \rangle$, $\bar{S}_{\alpha i}^\eta = \langle \hat{S}_{\alpha i}^\eta(\mathbf{r}_i) \rangle$, $\bar{S}_{\alpha i}^\eta = \frac{1}{2} \sum_{\sigma\sigma'} \langle c_{\alpha\sigma}^\dagger(\mathbf{r}_i) \sigma_{\sigma\sigma'}^\eta c_{\alpha\sigma'}(\mathbf{r}_i) \rangle$, and $J_M = 2J_K/N$.

$|\Psi\rangle$ that minimizes $\langle \Psi | H_N | \Psi \rangle$ is the ground state of H_{MF} . Working to quadratic order in J_M, D^η, h^η , we can treat H_1 perturbatively and use linear response theory to compute the ground-state energy of H_{MF} . Since $\bar{S}_{\alpha i}^\eta$ and $\bar{S}_{1\alpha i}^\eta$ are both on the order of J_M, D^η, h^η , we have further approximated H_1 . The change in the order parameters $\bar{V}_1, \bar{\lambda}_1$, and μ due to the perturbation is of second order in J_M, D^η, h^η , and because they mark the stationary point of $\langle \Psi | H_0 | \Psi \rangle$, it can be neglected. We can now straightforwardly find the effective Hamiltonian

$$\begin{aligned} H_{\text{eff}} &= -\frac{1}{2} \int_0^\beta \langle e^{H_0\tau} H_1 e^{-H_0\tau} H_1 \rangle_{H_0} d\tau - \sum_{i\alpha\eta} h_i^\eta \hat{S}_{2\alpha i}^\eta \\ &= -\frac{1}{2} \sum_{\eta\alpha, i \neq j} \hat{S}_{2\alpha i}^\eta \hat{S}_{2\alpha j}^\eta [\chi_{ff}(\mathbf{r}_i - \mathbf{r}_j) (2D^\eta)^2 \\ &\quad + 2\chi_{fc}(\mathbf{r}_i - \mathbf{r}_j) 2D^\eta J_M + \chi_{cc}(\mathbf{r}_i - \mathbf{r}_j) J_M^2] \\ &\quad - \sum_{ij\alpha\eta} \hat{S}_{2\alpha i}^\eta [\delta_{ij} - J_M (\chi_{cc}(\mathbf{r}_i - \mathbf{r}_j) + \chi_{cf}(\mathbf{r}_i - \mathbf{r}_j)) \\ &\quad - 2D^\eta (\chi_{ff}(\mathbf{r}_i - \mathbf{r}_j) + \chi_{cf}(\mathbf{r}_i - \mathbf{r}_j))] h_j^\eta, \end{aligned} \quad (\text{A3})$$

where $\bar{S}_{2\alpha i}^\eta$ have been promoted to spin-1/2 operators, constant terms have been dropped, $\chi_{ff}(\mathbf{r}) = \int_0^\beta \langle e^{H_0\tau} \hat{S}_{1\alpha}^\eta(\mathbf{r}) e^{-H_0\tau} \hat{S}_{1\alpha}^\eta(\mathbf{0}) \rangle_{H_0}$, $\chi_{cf}(\mathbf{r}) = \int_0^\beta \langle e^{H_0\tau} \hat{S}_\alpha^\eta(\mathbf{r}) e^{-H_0\tau} \hat{S}_{1\alpha}^\eta(\mathbf{0}) \rangle_{H_0}$, $\chi_{cc}(\mathbf{r}) = \int_0^\beta \langle e^{H_0\tau} \hat{S}_\alpha^\eta(\mathbf{r}) e^{-H_0\tau} \hat{S}_\alpha^\eta(\mathbf{0}) \rangle_{H_0}$, and $\chi(\mathbf{q}) = \sum_i \chi(\mathbf{r}) e^{-i\mathbf{q}\cdot\mathbf{r}_i}$. These susceptibilities can be easily evaluated to obtain the expressions given in the main text, once H_0 is expressed in terms of the heavy-fermion quasiparticle creation/annihilation operators $d_{v\alpha\sigma}^\dagger(\mathbf{k})$ and $d_{v\alpha\sigma}(\mathbf{k})$:

$$H_0 = \sum_{v\mathbf{k}\alpha\sigma} E_{\mathbf{k}}^v d_{v\alpha\sigma}^\dagger(\mathbf{k}) d_{v\alpha\sigma}(\mathbf{k}) - \sum_{i\alpha\eta} h_i^\eta \bar{S}_{2\alpha i}^\eta, \quad (\text{A4})$$

where we have dropped the constant terms, $v = \pm$ indexes the heavy-fermion bands, $d_{v\alpha\sigma}(\mathbf{k}) = u_{\mathbf{k}}^v c_{\sigma\alpha}(\mathbf{k}) + v_{\mathbf{k}}^v f_{1\sigma\alpha}$, and

$$E_{\mathbf{k}}^\pm = \frac{\epsilon_{\mathbf{k}} - \mu - \bar{\lambda}_1 \pm \sqrt{(\epsilon_{\mathbf{k}} - \mu + \bar{\lambda}_1)^2 + 4J_K^2 \bar{V}_1^2}}{2},$$

$$u_{\mathbf{k}}^\pm = \frac{E_{\mathbf{k}\alpha}^\pm + \bar{\lambda}_1}{\sqrt{(E_{\mathbf{k}}^\pm + \bar{\lambda}_1)^2 + J_K^2 \bar{V}_1^2}},$$

$$v_{\mathbf{k}}^\pm = \frac{-J_K \bar{V}_1}{\sqrt{(E_{\mathbf{k}}^\pm + \bar{\lambda}_1)^2 + J_K^2 \bar{V}_1^2}}. \quad (\text{A5})$$

For example, using Wick's theorem, we obtain the following for $\chi_{ff}(\mathbf{r})$:

$$\begin{aligned} \chi_{ff}(\mathbf{r}) &= \frac{1}{2} \int_0^\beta \langle e^{H_0\tau} f_{1\alpha\uparrow}^\dagger(\mathbf{r}) e^{-H_0\tau} f_{1\alpha\uparrow}(\mathbf{0}) \rangle_{H_0} \\ &\quad \times \langle e^{H_0\tau} f_{1\alpha\uparrow}(\mathbf{r}) e^{-H_0\tau} f_{1\alpha\uparrow}^\dagger(\mathbf{0}) \rangle_{H_0} d\tau \\ &= \frac{1}{2N_s^2} \sum_{\mathbf{k}\mathbf{q}\mathbf{v}\mathbf{v}'} e^{i(\mathbf{q}-\mathbf{k})\cdot\mathbf{r}} |v_{\mathbf{k}}^v|^2 \langle d_{v\alpha\uparrow}^\dagger(\mathbf{k}) d_{v\alpha\uparrow}(\mathbf{k}) \rangle_{H_0} \\ &\quad \times |v_{\mathbf{q}}^{v'}|^2 \langle d_{v'\alpha\uparrow}(\mathbf{q}) d_{v'\alpha\uparrow}^\dagger(\mathbf{q}) \rangle_{H_0} \frac{e^{\beta(E_{\mathbf{k}}^v - E_{\mathbf{q}}^{v'})} - 1}{E_{\mathbf{k}}^v - E_{\mathbf{q}}^{v'}}, \quad (\text{A6}) \end{aligned}$$

which gives us the desired result once we express the expectation values in terms of the Fermi function.

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