The \( f \)-electron physics of rare-earth iron pnictides: influence of \( d \)-electron antiferromagnetic order on the heavy fermion phase diagram

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Some of the high \( T_c \) iron pnictides contain magnetic rare-earth elements, raising the question of how the existence and tunability of a \( d \)-electron antiferromagnetic order influences the heavy fermion behavior of the \( f \)-moments. With CeOF\(_{\text{P}}\) and CeOF\(_{\text{As}}\) in mind as prototypes, we derive an extended Anderson lattice model appropriate for these quaternary systems. We show that the Kondo screening of the \( f \)-moments are efficiently suppressed by the \( d \)-electron ordering. We also argue that, inside the \( d \)-electron ordered state (as in CeOF\(_{\text{As}}\)), the \( f \)-moments provide a rare realization of a quantum frustrated magnet with competing \( J_1-J_2-J_3 \) interactions in an effective square lattice. Implications for the heavy fermion physics in broader contexts are also discussed.

PACS numbers: 71.10.Hf, 71.27.+a, 74.70.Tx, 75.10.-b

The homologous rare-earth iron arsenides exhibit antiferromagnetic (AF) ground states in addition to the high temperature superconductivity \([1,2,3,4,5]\). The systems of interest here are the arsenides RO\(_2\)F\(_{1-x}\)FeAs, with \( R = \text{Ce, Sm, Nd, Pr...} \) being magnetic rare earths, which have superconducting transition temperatures higher \([6,7]\) than the maximal \( T_c \approx 26 \text{ K} \) of LaO\(_2\)F\(_{1-x}\)FeAs \([8]\). The parent compounds of these systems, ROFeAs, have a layered structure, with FeAs and \( RO \) layers sandwiching each other. They typically show a collinear AF order and a structure distortion, which are successively suppressed by carrier doping in favor of superconductivity \([4]\). Also of interest are the iron phosphides. LaOFe\(_{\text{P}}\) was the iron pnictide reported to show superconductivity below \( T_c \approx 4 \text{ K} \) \([9]\). This compound has the same layered structure as LaOFe\(_{\text{As}}\), but does not order magnetically \([6]\).

The distinction between the iron phosphides and arsenides becomes even more pronounced when La is replaced by Ce. CeOF\(_{\text{P}}\) is neither superconducting nor magnetically ordered, and its Ce \( f \)-electrons exhibit heavy fermion behavior with a Kondo temperature \( T_K \approx 10 \text{ K} \) \([9]\). CeOF\(_{\text{As}}\) has the \( d \)-electron collinear AF ordering below \( T_N^{(d)} \approx 130 \text{ K} \). Its \( f \)-electrons display a noticeable AF order below \( T_N^{(f)} \approx 4 \text{ K} \) \([9,10]\), but does not show any heavy fermion features. What underlies the heavy fermion behavior in CeOF\(_{\text{P}}\) and its absence in CeOF\(_{\text{As}}\)? One possibility is that this primarily reflects the very different interlayer \( 3d-4f \) couplings between CeOF\(_{\text{P}}\) and CeOF\(_{\text{As}}\), as suggested by a first-principle LDA+DMFT study \([11]\). However, a more complete theoretical estimate using a full density of states, which is strongly peaked away from the Fermi energy, suggests that the effective Kondo couplings in CeOF\(_{\text{P}}\) and CeOF\(_{\text{As}}\) may in fact be comparable \([12]\). Muon-spin relaxation and neutron scattering experiments \([13,14]\) may also be interpreted in terms of a sizable Kondo coupling in CeOF\(_{\text{As}}\).

In this Communication, we discuss the possibility that the distinction in the \( d \)-electron magnetism between CeOF\(_{\text{P}}\) and CeOF\(_{\text{As}}\) plays an important role in influencing their heavy fermion behavior. This mechanism is expected to play an especially important role when we consider not only the end materials CeOF\(_{\text{P}}\) and CeOF\(_{\text{As}}\), but also the series CeOF\(_{\text{As}}\)\(_{1-x}\)P\(_x\), which has been proposed to realize a continuously varying \( d \)-electron AF order and the associated quantum critical point \([15]\).

Studying the effect of the \( d \)-electron AF order on the heavy fermion phase diagram not only sheds new light on the properties of the iron pnictides, but also represents a new twist to the heavy fermion physics in general. Typically, AF order in heavy fermion metals is induced by the RKKY interactions among the \( f \)-moments, and the heavy fermion phase diagram involves the competition between RKKY and Kondo coupling \([16,17]\). A tunable \( d \)-electron AF order adds a new dimension to the heavy fermion phase diagram.

In the following, we will consider this effect within an extended Anderson lattice model (ALM) appropriate for the stoichiometric \( R-1111 \) compounds ROFe\(_{\text{X}}\) \((X=\text{As or P})\). The model incorporates the inter-layer hybridization between pnictogen \( X \) \( p \)-orbitals and rare earth \( R \) \( f \)-orbitals. We note in passing that the derived model takes into account the microscopic crystal structure and symmetry of the \( R-1111 \) compounds. Given that there are many materials of the same ZrCuSiAs-type structure \([18]\), with many of them containing magnetic rare-earth elements, we expect that our model will also be germane to many such related compounds \([19]\).

**General considerations.** The lattice structure of the \( R-1111 \) compound series is schematically shown in Fig. 1. Let Fe-atoms be in the \((x,y)\)-plane with the coordinate \((\vec{r},0)\), where, \( \vec{r} = (\xi_x,\xi_y)\), \( \xi_x \) and \( \xi_y \) are both integers (the nearest Fe-Fe distance is set to unity). The coordinates of \( X- \) and \( R \)-atoms are \((\vec{r}_p,\eta_2\xi_p)\) and \((\vec{r}_q,\eta_2\xi_q)\) respectively,
where \( \vec{r}_p = (i_x + 1/2, i_y + 1/2), \vec{r}_f = (i_x - 1/2, i_y + 1/2), \eta = e^{i\pi(x+y)}, \) \( z_p \) and \( z_f \) are the distances of X- and R-atoms to the Fe-plane. We denote the \( d-, p-, \) and \( f- \)electrons by \( d^{(\alpha)}(\vec{r}), p^{(\mu)}(\vec{r}, \eta_z), \) and \( f^{(m)}(\vec{r}, \eta_z) \), with orbital indices \( \alpha = d_{xy}, d_{yz}, d_{x^2-y^2}, d_{3z^2-r^2}, \mu = p_x, p_y, p_z, \) and \( m = 1, \ldots, l \).

**The model Hamiltonian.** The hybridization part of the Hamiltonian is given by \( H_{\text{hybrid}} = H_{pd} + H_{pf} \), where

\[
H_{pd} = \sum_{\vec{r}} V_{pd}(\mu)(\vec{r}_p, \eta_z \vec{p}) D_\sigma(\vec{r}) + \text{h.c.},
\]

\[
H_{pf} = \sum_{\vec{r}} V_{pf}(\mu)(\vec{r}_p, \eta_z \vec{r}) F_\sigma(\vec{r}, \eta_z) + \text{h.c.}.
\]

Here we introduce \( D_\sigma(\vec{r}) = \sum_{\alpha} d^{(\alpha)}(\vec{r}) \equiv d^{(\alpha)}(i_x, i_y) + d^{(\alpha)}(i_x + 1, i_y) + d^{(\alpha)}(i_x, i_y + 1) + d^{(\alpha)}(i_x + 1, i_y + 1) \) and \( F_\sigma(\vec{r}, \eta_z) = \sum_{m} f^{(m)}(\vec{r}, \eta_z) \equiv f^{(m)}(i_x - 1/2, i_y + 1/2, \eta_z) + f^{(m)}(i_x + 1/2, i_y + 1/2, \eta_z) + f^{(m)}(i_x + 1/2, i_y - 1/2, \eta_z) + f^{(m)}(i_x - 1/2, i_y - 1/2, \eta_z) \) as the plaquette operators for \( d-, p-, \) and \( f- \)electrons around X-atoms. (Summations over the repeated spin and channel indices are implied hereafter unless otherwise specified.)

The interaction part of the Hamiltonian, \( H_{int} = H_{int,d} + H_{int,p} + H_{int,f} \), contains the usual on-site Coulomb interactions \( (U_p, U_d, \) and \( U_f) \) and the Hund's coupling \( (J_d). \) The total Hamiltonian is then \( H = H_0 + H_{\text{hybrid}} + H_{\text{int}}, \) with \( H_0 \) containing the primitive site energies of \( d-, p-, \) and \( f- \)electrons denoted by \( \epsilon^{(\alpha)}_d, \epsilon^{(\mu)}_p, \) and \( \epsilon^{(m)}_f, \) respectively.

It is expected that \( U_p \) is small compared to the other Coulomb interactions. We will therefore set \( U_p = 0, \) in which case the \( p- \)orbitals can be readily integrated out. The obtained effective Hamiltonian \( \tilde{H} \) takes the form

\[
\tilde{H} = H_0 + H_d + H_f + H_{df} + H_{int,d} + H_{int,f}.
\]

Here

\[
H_d = \sum_{\vec{r}} V_d^{(\alpha\alpha')}(\vec{r}) D_\sigma^{(\alpha)}(\vec{r}) D_\sigma^{(\alpha')}(\vec{r}) + \text{h.c.},
\]

\[
H_f = \sum_{\vec{r}} V_f^{(m\mu)}(\vec{r}) F_\sigma^{(m)}(\vec{r}) F_\sigma^{(m)}(\vec{r}) + \text{h.c.},
\]

and \( H_{df} = \sum_{\vec{r}} V_{df}^{(\alpha\mu)}(\vec{r}) D_\sigma^{(\alpha)}(\vec{r}) F_\sigma^{(m)}(\vec{r}) + \text{h.c.}, \)

with \( V_{df}^{(\alpha\mu)} = -\sum_{\mu} V_{pd}^{(\mu)}(\vec{r}) \eta_z V_{pf}^{(\mu\alpha')}(\vec{r}, \eta_z) + \text{h.c.} \),

\[
V_{df}^{(m\mu)} = -\sum_{\mu} V_{pf}^{(\mu\alpha)}(\vec{r}) \eta_z V_{pf}^{(\mu\alpha)}(\vec{r}, \eta_z) + \text{h.c.},
\]

\[
V_{df}^{(m\mu)} = -\sum_{\mu} V_{pd}^{(\mu)}(\vec{r}) \eta_z V_{pf}^{(\mu\alpha)}(\vec{r}, \eta_z) + \text{h.c.}.
\]

In the momentum \( K \)-space (in the reduced Brillouin zone corresponding to two Fe-atoms in the conventional cell with lattice constant \( a = \sqrt{2}, \)) \( H_d = \sum_{K} V_{d}^{(\alpha\alpha')} g^{(\eta\eta')}_d(K) e^{i\eta\vec{K} \cdot \vec{r}} e^{i\eta'\vec{K} \cdot \vec{r}}, \)

\[
H_f = \sum_{K} V_{f}^{(m\mu)} g_f(K) f^{(m)}(K) f^{(m)}(K) + \text{h.c.},
\]

and \( H_{df} = \sum_{K} g^{(\eta\eta')}_{df}(K) e^{i\eta\vec{K} \cdot \vec{r}} e^{i\eta'\vec{K} \cdot \vec{r}} + \text{h.c.}, \)

\[
H^{(\eta\eta')}_{df} = \sum_{\eta} f^{(m)}(K, \eta_{K\sigma} \eta_{K\sigma}) \eta_{K\sigma} \eta_{K\sigma} + h.c.,
\]

where \( d^{(\alpha)} \) and \( f^{(m)} \) are the Fourier transform of \( d- \) and \( f- \)electron operators in the sublattices \( \eta = A \) or \( B, \) respectively. The \( K \)-dependence of the dispersions and \( d-f \) hybridization is only encoded in the form factors, given by \( g_{d}^{(AA)}(K) = g_{d}^{(BB)}(K) = 4 + 2 \cos(K_{\perp A} \cos(K_{\perp B}, \)

\[
g_{d}^{(AB)}(K) = g_{d}^{(BA)}(K) = 8 \cos(K_{\perp A}/2) \cos(K_{\perp B}/2), \)

\[
g_{f}(K) = 16 \cos^2(K_{\perp A}/2) \cos^2(K_{\perp B}/2), \)

\[
g_{d}^{(AA)}(K) = g_{d}^{(BB)}(K) = 8 \cos^2(K_{\perp A}/2) \cos(K_{\perp B}/2), \)

\[
g_{d}^{(AB)}(K) = 8 \cos(K_{\perp A}/2) \cos^2(K_{\perp B}/2). \)

**The d-electron correlations.** For moderate large \( U_d, \) we may start from the strong coupling limit yielding the frustrated \( J_1-J_2 \) Heisenberg model for the \( d- \)electrons [20].

The itinerancy of the \( d- \)electrons will further reduce the ordered moments and eventually lead to a paramagnetic phase [13]. In fact, both the weak- and strong-coupling limits suggest that the staggered magnetization \( M_d = \sum_{\alpha} m_d^{(\alpha)}/(1/N) \sum_{K}(d^{(\alpha)}_K d^{(\alpha)}_K - d^{(\alpha)}_K d^{(\alpha)}_{K+Q}) \) is a dominating order parameter with \( Q = (\pi, \pi) \) and \( N \) being the number of \( K \) points in the reduced Brillouin zone. For the purpose of demonstrating the effect of \( d- \)electron order on the Kondo effect, we treat \( m_d^{(\alpha)} \) as the mean field parameters and approximate \( H_{int,d} \) by \( J_d \sum_{K} \sigma m_d^{(\alpha)} d^{(\alpha)}_K d^{(\alpha)}_{K+Q} + h.c., \) with \( J_d \) being the effective coupling strength. The AF ordering gap, \( \Delta_{AF} = J_d M_d, \) is sizable for FeAs but vanishes for FeP.

**Kondo effect vs. d-electron ordering.** In order to understand the competition between the Kondo effect and \( d- \)electron AF order, we first neglect the \( f- \)electron ordering. We are then led to consider

\[
H_{ALM} = \sum_{K} \left[ \epsilon^{(\alpha)}_d d^{\alpha\alpha'}_d \eta_{\eta'\eta} + V_{df}^{(aa')} g_f(K) d^{(a)}_K d^{(a')}_{K+Q} \right] + \sum_{K} \left[ \epsilon^{(m)}_{f} d^{mm'}_{f} + V_{f}^{(mm')} g_f(K) f^{(m)}_K f^{(m')}_{K+Q} \right] + \sum_{K} \left[ \epsilon^{(m)}_{f} d^{mm'}_{f} + V_{f}^{(mm')} g_f(K) f^{(m)}_K f^{(m')}_{K+Q} \right] + \sum_{K} [\sigma \Delta_{AL} d^{(a)}_K d^{(a)}_{K+Q} + h.c.]
\]

\[
+ \sum_{K} \epsilon^{(m)}_{f} \sum_{f} n^{(m)}_K (\vec{r}_f, \eta_f) n^{(m)}_{K+Q} (\vec{r}_f, \eta_f) \right] + \sum_{K} \epsilon^{(m)}_{f} \sum_{f} n^{(m)}_K (\vec{r}_f, \eta_f) n^{(m)}_{K+Q} (\vec{r}_f, \eta_f) \right].
\]
In the absence of $d$-electron ordering, Eq.(4) is the ALM with weak $f$-electron dispersion and momentum dependent hybridization. (The effect of momentum dependent hybridization on the Kondo effect has recently been studied in other contexts \cite{23,24}.) For sufficiently large $U_f$, and with the $f$-levels being well below Fermi energy, we are in the Kondo limit.

To concretely demonstrate how the $d$-electron AF order influences the Kondo effect, we consider the resulting Kondo lattice model with a single $f$-electron channel and two $d$-electron bands. In the slave-boson representation this becomes

$$H_{KLM} = \sum_{\mathbf{k}} \varepsilon_d^{(\alpha\sigma)}(\mathbf{k})d_{\mathbf{k}\sigma}^{(\alpha)}d_{\mathbf{k}\sigma}^{(\alpha)^\dagger} + \lambda \left( \frac{1}{N_L} \sum_{\mathbf{k}} f_{\mathbf{k}\sigma}^\dagger f_{\mathbf{k}\sigma} - 1 \right) + \sum_{\mathbf{k}} \left[ \sigma \Delta_{\text{AF}} d_{\mathbf{k}\sigma}^{(d)} d_{\mathbf{k+Q}\sigma}^{(d)} + \text{H.c.} \right] - \frac{1}{2} J_K \sum_{\mathbf{k}} V_{df}(\mathbf{k})b_\alpha \left[ f_{\mathbf{k}\sigma}^{\dagger} d_{\mathbf{k}\sigma}^{(\alpha)} + \text{H.c.} \right], \quad (5)$$

Here, the Lagrange multiplier $\lambda$ enforces the single occupancy of $f$-electrons. The mean-field parameter $b_\alpha = \langle f_{\mathbf{k}\sigma}^{\dagger} d_{\mathbf{k}\sigma}^{(\alpha)} \rangle / 2$ describes the Kondo screening and sets the Kondo scale, $T_K \propto b^2$. The anisotropic hybridization form factor $V_{df}(\mathbf{k}) = 4 \cos k_x / 2 \cos k_y / 2$.

The energy dispersion for $d$-electrons is taken to be \cite{22}:

$$\epsilon^{(1)}(\mathbf{k}) = -2t_1 \cos k_x - 2t_2 \cos k_y - 4t_3 \cos k_x \cos k_y,$$

$$\epsilon^{(2)}(\mathbf{k}) = -2t_2 \cos k_x - 2t_1 \cos k_y - 4t_3 \cos k_x \cos k_y,$$

$$\epsilon^{(12)}(\mathbf{k}) = \epsilon^{(21)}(\mathbf{k}) = -4t_4 \sin k_x \sin k_y,$$

with $t_1 = -1, \ t_2 = 1.3, \ t_3 = t_4 = -0.85$. In our numerical study, we choose $J_K = 0.04$, temperature $T = 10^{-10}|t_1|$, and the lattice size $N_L = 3200 \times 3200$. When we vary the AF order parameter, the chemical potential is adjusted such that the $d$-electrons are fixed at the half-filling $n_d = 2.0$.

Fig. 2 shows that the $d$-electron AF order rapidly suppresses the Kondo scale. This suppression is closely related to the depression of the $d$-electron density of states (DOS) in the collinear AF state of undoped iron arsenides (see the inset of Fig. 2). The feature of low energy DOS is sensitive to the degree of nesting and the DOS minimum is not necessarily located precisely at the Fermi energy (see, e.g., the case of $M_d = 0.021$ in Fig. 2); the latter explains the effective Kondo scale first rising and then dropping with the AF order. Furthermore, the incomplete nesting of the Fermi surface keeps the depressed DOS finite (unlike, say, in the superconducting state) at the Fermi energy such that the $T = 0$ ground state has the $f$-moment always Kondo screened on the lattice.

We should stress that, for the purpose of a semi-quantitative assessment of the proposed mechanism, we have considered the upper limit for the Kondo scale in the AF state: we have coupled the $f$-moments to only the quasiparticles of the $d$-electron AF state and have also neglected the $f$-moment ordering; moreover, a genuine $f$-electron quantum phase transition will be induced by breaking the Kondo screening upon the inclusion of the standard RKKY-Kondo competition \cite{26,27,28}. We can therefore infer that the mechanism proposed here provides a viable basis to understand the distinct $f$-electron heavy fermion behaviors in CeOF eP ($M_d \approx 0$) and CeOFeAs ($M_d \approx 0.8$) \cite{10}. Our results also set the stage for understanding the evolution of the heavy fermion behavior in the CeOFeAs$_{1-x}$P$_x$ series. In general, there will be two magnetic quantum critical points $x_{c1}$ and $x_{c2}$, associated with the $d$- and $f$-electrons, respectively. The RKKY interaction would then dominate in the intermediate region of $x$, leading likely to a ferromagnetic order before the heavy fermion state is approached.

**Magnetic frustration of the $f$-electrons.** We now turn to the exchange interactions among the $f$-moments. Consider first the superexchange interaction, which can be derived by integrating out the virtual valence fluctuations of the $f$-electrons. From Eq. 4, we end up with $H_f = \sum_{\mathbf{r}} J_f^{(m,m')} \delta_F^{(m)}(\mathbf{r}_f, \eta z_f) \cdot \delta_F^{(m')}(\mathbf{r}_{f'}', \eta z_{f'})$, where $\delta_F^{(m)}(\mathbf{r}, \eta z) = \sum_\mathbf{r} \delta_F^{(m)}(\mathbf{r}, \eta z_f')$ are summations of $f$-electron spins in the corresponding plaquettes associated with $\mathbf{r}$, and $J_f^{(m,m')} \approx 2 \langle V_f^{(m,m')} \rangle^2 \left( \frac{1}{U_f + \epsilon_f^{m'}} - \frac{1}{\epsilon_f^{m'}} \right)$. This is the superexchange interaction associated with the $R$-$X$-$R$ path, which does not mix the odd and even sublattices of the $f$-sites in a single $RO$ layer (see Fig. 8(a)). There will also be a superexchange interaction from the $R$-$O$-$R$ path, due to the hybridization between the $4f$-orbitals of $X$-atoms and the 2$p$-orbitals of $O$-atoms; this superexchange mixes the odd and even sublattices (see Fig. 8(b)). In the notations of an effective square lattice of the $f$-sites (c.f. Fig. 8(c)) the $R$-$O$-$R$ path gives
rise to the nearest-neighbor (n.n.) interaction $J_1^{(O)}$ and
the next-nearest-neighbor (n.n.n.) $J_2^{(O)}$, while the R-
X-R path yields the n.n.n. $J_3^{(X)}$ and the third-nearest-
neighbor (n.n.n.n.) $J_3^{(X)}$. (Note that $J_2^{(X)}$ and $J_3^{(X)}$
are expected to turn the orders of (a) and (b) into a
helical one. The blue and brown circles label the rare-earth
surfaces, and this is consistent with the enhanced ferro-
magnetic fluctuations of the heavy fermion state observed
in CeOFeP. Still, the frustrating $J_1$-$J_2$-$J_3$ superex-
change interactions will continue to operate, helping to
suppress the tendency for AF ordering.

Discussion and summary. A number of other conse-
quences of the $p$-$f$ hybridization are relevant to the iron-
 pnictides phase diagram. First, in the heavy fermion phase,
the momentum-dependence of the induced $d$-
$f$ hybridization will generally smear the hybridization
gap (which has nodal lines along $K = \pm \pi/a$ and $K = \pm \pi/a$), and this could be visible in the optical-
conductivity spectrum. Second, the induced $d$-$f$ hy-
bridization depends on the Fe-X and X-R distances.
Increasing pressure along the c-axis will decrease the dis-
tances and increase the hybridizations, and eventually
enhance $T_K$. Finally, in light of the fact that the f-
electron ordering is further suppressed by the competing
$J_1$-$J_2$-$J_3$ interactions, the transition or crossover from
the superconducting to the heavy fermion phases may take
place at sufficiently high pressures in the carrier-doped
superconducting materials.

In summary, we have considered a mechanism for
weakening the Kondo screening effect through the ant-
iferromagnetic order of the conduction electrons, and
implemented it in an extended Anderson lattice Hamil-
tonian. For the iron pnictides, our mechanism is
semi-quantitatively viable to explain the observed ex-
istence/absence of heavy fermion behavior in CeOFeP
and CeOFeAs, respectively. More broadly, our mecha-
nism goes beyond the standard picture of heavy fermion
physics, viz. the RKKY and Kondo competition, and can
therefore shed new light on the phase diagram of heavy
fermion systems in general. Finally, we have proposed that
the $f$-electrons in the parent iron arsenides represent
a rare model system for quantum frustrated magnetism
in two dimensions.

We thank E. Abrahams, M. Aronson, G. H. Cao, X. H.
Chen, X. Dai, C. Geibel, N. L. Wang, T. Xiang, Z. A. Xu,
and H. Q. Yuan for useful discussions, and the U.S. DOE
CINT at LANL for computational support. This work
was supported by the NSF of China, the 973 Program,
and the PCSIRT (IRT-0754) of Education Ministry of
China (J.D.), the NSF Grant No. DMR-0706625 and the
IRIT-0754) of Education Ministry of
China (J.D.), the NSF Grant No. DMR-0706625 and the
Robert A. Welch Foundation (Q.S.), and by U.S. DOE
at LANL under Contract No. DE-AC52-06NA25396 (J.-
X.Z.).

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