Superconductivity from emerging magnetic moments

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A growing body of theoretical work shows that the Hund coupling in materials with multiple active orbitals leads to interesting correlation effects \cite{1} and that this physics is important for understanding the properties of ruthenates \cite{2,3} and iron pnictides \cite{4-6}. These so-called Hund metals exhibit large mass enhancements without Hubbard bands, and many show characteristic non-Fermi liquid properties \cite{2,7-9}. The underlying phenomenon is spin-freezing \cite{2}: In a rather narrowly defined range of fillings and interaction strengths, long-lived magnetic moments appear in the metal phase. Here, we show that the fluctuating local moments at the border of the spin-frozen regime induce spin-triplet superconductivity at low temperature. The resulting phase diagram features a superconducting dome below a non-Fermi liquid metallic region and next to a magnetically ordered phase. We suggest that this type of fluctuating-moment induced superconductivity, which is not related to a quantum critical point, may be realized in spin-triplet superconductors such as Sr\textsubscript{2}RuO\textsubscript{4} and the uranium compounds UGe\textsubscript{2}, UCoGe and URhGe.

Spin-triplet superconductivity, in the sense of equal-spin pairing, is believed to occur in a number of correlated materials. The best candidate is the layered compound Sr\textsubscript{2}RuO\textsubscript{4}, where the Knight shift remains unchanged across the superconducting phase boundary, in stark contrast with the behavior expected for spin-singlet pairing \cite{10}. In the iron pnictides, where a spin-triplet superconducting phase has been proposed in early theoretical works \cite{11}, the experimental evidence points toward spin-singlet pairing, although in LiFeAs a spin-triplet scenario is still being debated \cite{12-15}. The uranium based superconductors are also possible candidates for spin-triplet pairing. In compounds such as UGe\textsubscript{2}, UCoGe and URhGe, the superconducting state is found near a non-Fermi liquid metallic region, and iron pnictides \cite{4-6}. These so-called Hund metals exhibit large mass enhancements without Hubbard bands, and many show characteristic non-Fermi liquid properties \cite{2,7-9}. The underlying phenomenon is spin-freezing \cite{2}: In a rather narrowly defined range of fillings and interaction strengths, long-lived magnetic moments appear in the metal phase. Here, we show that the fluctuating local moments at the border of the spin-frozen regime induce spin-triplet superconductivity at low temperature. The resulting phase diagram features a superconducting dome below a non-Fermi liquid metallic region and next to a magnetically ordered phase. We suggest that this type of fluctuating-moment induced superconductivity, which is not related to a quantum critical point, may be realized in spin-triplet superconductors such as Sr\textsubscript{2}RuO\textsubscript{4} and the uranium compounds UGe\textsubscript{2}, UCoGe and URhGe.

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While a p-wave symmetry is usually assumed for the pairing state in spin-triplet superconductors, an s-wave spin-triplet pairing is also possible by taking into account the orbital degrees of freedom. The mechanism of this unconventional superconductivity can be easily understood \cite{11,14,23}: same-spin electrons tend to occupy the same site due to the Hund coupling which favors high-spin states. A new insight in this paper is that the s-wave spin-triplet pairing is closely connected to the emergence of local magnetic moments in the spin-freezing crossover regime.

Spin-freezing is a consequence of the formation of a large composite spin in multi-band systems with Hund coupling (right panel of Fig. 1b). In the absence of long-range order, the emerging local moments will be screened at sufficiently low temperature, so that there is no quantum phase transition associated with the spin freezing phenomenon. However, screening large local moments is difficult, and the Fermi liquid coherence temperature becomes very low \cite{20}. Hence, as demonstrated here, a spontaneous symmetry breaking pre-empts the screening of the moments. While deep in the spin-frozen regime the long-lived local moments order magnetically at low temperatures, the emerging and still fluctuating local moments in the spin-freezing crossover regime generate spin-triplet pairing. This leads to the formation of a superconducting dome separating the Fermi liquid metal from the magnetically ordered region, resulting in phase diagrams which closely resemble those of unconventional superconductors.

While the phenomena discussed in this paper are generic features of multi-orbital systems with non-zero Hund coupling parameter J, we will show results for the 3-orbital case. As explained in the Methods section, the model contains intra-orbital (U) and inter-orbital (U') Coulomb repulsions, and we choose U' = U - 2J and J/U = 1/4. To study the effect of spin anisotropy, which may originate from spin-orbit coupling, we introduce a parameter α that allows to interpolate between the Ising anisotropic (α = 0) and SU(2) symmetric (α = 1) interaction in spin space. The model is solved using the dynamical mean field theory (DMFT) \cite{27}, combined with a numerically exact continuous-time Monte Carlo method \cite{28}, which captures local correlation effects. For simplicity, we consider a semi-circular density of states with bandwidth W = 1.

To illustrate the spin-freezing phenomenon \cite{2}, we compute the dynamic contribution to the local magnetic susceptibility

\[
\Delta \chi_{\text{loc}} = \int_0^\beta d\tau \left( \langle S_i(\tau) S_i \rangle - \langle S_i(\beta/2) S_i \rangle \right), \tag{1}
\]

where \( \beta = 1/T \) is the inverse temperature and \( O(\tau) = e^{i\tau H} O e^{-i\tau H} \). The operator \( S_i = \frac{1}{2M} \sum_{\gamma=1}^M (c^\dagger_{\gamma i} c_{\gamma i} - c^\dagger_{\gamma i} c_{\gamma i}) \), with \( M = 3 \) the number of orbitals, measures the local spin. The first term on the right-hand
FIG. 1: Spin freezing phenomenon. a, Illustration of possible local configurations with two electrons in three orbitals, and the corresponding energies $E$. The Hund coupling $J$ favors the right-most configuration with a large composite spin. b, Filling dependence of the local magnetic susceptibilities for $U = 1$ and $T = 0.005$. The black curve shows the local magnetic susceptibility $\chi_{\text{loc}}$, and the blue curve the contribution $\beta \langle S_i (\beta/2) S_i \rangle$ reflecting long-lived (frozen) moments. The difference $\Delta \chi_{\text{loc}} = \chi_{\text{loc}} - \beta \langle S_i (\beta/2) S_i \rangle$ (red curve) quantifies the fluctuations of the local moments and is used here to define the spin-freezing crossover regime (magenta), as well as the crossover point (location of the maximum). $\alpha = 0$ and $\alpha = 1$ correspond to the Ising-anistropic and spin-isotropic system, respectively. c, Filling dependence of the renormalization factor $z$ for the same parameters.

side yields the local magnetic susceptibility ($\chi_{\text{loc}}$). In Eq. (1) we subtract the long-time correlator $\langle S_i (\beta/2) S_i \rangle$, which reflects the magnitude of long-lived frozen moments. Hence, the quantity $\Delta \chi_{\text{loc}}$ measures the fluctuations of the moments. Figure 1b shows the filling dependence of these quantities for $\alpha = 0$. While the local susceptibility $\chi_{\text{loc}}$ monotonically increases with increasing $n$, the fluctuation $\Delta \chi_{\text{loc}}$ reaches a maximum at $n \approx 1.9$. This peak indicates the crossover between the Fermi liquid and spin-frozen regimes, and we use the location of the maximum as our definition of the “spin-freezing crossover”. The spin-freezing is also reflected in the renormalization factor $z$, or mass-enhancement factor $1/z$, of the quasi-particles (see Method section for more details). Figure 1c exhibits a drop of $z$ in the spin-freezing crossover region. Qualitatively similar results are obtained in the SU(2) symmetric case ($\alpha = 1$, dashed lines).

To study the stability regions of ordered phases, we calculated the susceptibilities

$$
\chi_{\text{loc}} = \frac{1}{N} \sum_i \langle S_i \rangle \langle \sigma \rangle \langle \sigma \rangle^\dagger 
$$

where $N$ is the total number of sites. The operator $\sigma$ is given by

$$
\sigma = \begin{cases} 
\sum_i S_i & \text{(FM)} \\
\sum_i \lambda_i S_i & \text{(AFM)} \\
\sum_i c^\dagger_{\gamma\gamma'} c^\dagger_{\gamma'\gamma} & \text{for } \gamma \neq \gamma' \text{ (SC)}
\end{cases}
$$

for ferromagnetic order (FM), anti-ferromagnetic order (AFM) and s-wave inter-orbital spin-triplet superconductivity (SC). $\lambda_i$ is a sign which depends on the sublattice. A divergence in $\chi_{\text{loc}}$ (or equivalently a sign-change in $1/\chi_{\text{loc}}$) indicates a possible transition into a long-range ordered phase. Figure 2a shows the inverse susceptibilities for $T = 0.005$, $U = 0.75$, $\alpha = 0$ and different fillings. Symmetry broken phases exist in the regions where $1/\chi_{\text{loc}} < 0$. For example, at $1.9 \lesssim n < 2.3$ (2.4 $\lesssim n \lesssim 3$) the SC (AFM) phase is stable. On the basis of these susceptibility calculations, it is not possible to determine the transition point from the SC to the AFM phase, and whether or not there may be a coexistence of both orders.

Repeating this analysis for different $U$, we obtain the $T = 0.005$ phase diagram shown in Fig. 3a. At $U > 1.25$, a FM phase appears, while near half-filling the AFM phase is stable. A new result is the existence of an extended SC region connecting the FM and AFM phases. This spin-triplet pairing is clearly associated with the spin-freezing crossover, which is indicated by the black dashed line. We also show the phase diagram at a lower $T = 0.0025$ in Fig. 3b, where the SC region expands.

Next, we discuss the temperature-filling phase diagram shown in Fig. 3c. With hole doping from half-filling ($n = 3$), the AFM transition temperature decreases and becomes zero at $n \approx 2.3$, which is close to the spin-freezing line. By further doping with holes, we find the spin-triplet superconducting phase with a dome-shaped $T_c$. If we fix the filling to $n = 2$ and change $U$, we obtain the phase diagram shown in Fig. 3d. With decreasing $U$ (increasing pressure, experimentally), the FM order is destroyed and again a superconducting dome appears next to the magnetic region. Superconducting domes are usually understood as a manifestation of fluctuations associated with magnetic quantum critical points. However, the superconductivity revealed in this paper is induced by local magnetic fluctuations in the spin-freezing crossover regime. Nevertheless, the superconducting order naturally appears in the vicinity of a magnetic phase, since the strengthening of the magnetic moments deeper.
FIG. 2: Stability region of ordered phases. a, Filling dependence of the inverse susceptibilities for FM, AFM and SC orders for $U = 0.75$, $T = 0.005$ in the system with Ising spin anisotropy ($\alpha = 0$). A negative $1/\chi_O$ indicates a long-range ordered phase with order parameter $O$. b,c, Interaction-filling phase diagrams at $T = 0.005$ and $T = 0.0025$, respectively. The black dashed line shows the location of the spin-freezing crossover in the system without long-range order.

FIG. 3: Critical temperatures of the ordered phases. a, Temperature-filling phase diagram at $U = 0.75$ (horizontal dashed line in Fig. 2b,c). b, Temperature-interaction phase diagram at $n = 2$ (vertical dashed line in Fig. 2b,c). Experimentally, a decrease of $U$ means an increase of pressure. The black dashed line indicates the spin-freezing crossover in the system with suppressed long-range order. In the spin-freezing crossover regime, the normal phase exhibits strong deviations from Fermi-liquid behavior.

inside the spin-frozen regime causes magnetic ordering. Furthermore, the normal state above the superconducting dome is a non-Fermi liquid whose properties are influenced by the spin-freezing crossover.

So far we have shown results for the system with Ising anisotropy. We now clarify how the superconductivity is affected by the spin-flip term in the model with $\alpha \neq 0$ (see Eq. 4). Figure 4 shows $T_c$ for $U = 0.875$ and filling $n = 2$. As $\alpha$ is increased from 0, the transition temperature decreases and drops below the lowest accessible temperature at $\alpha = 1$. The destabilization of the electron pairs by the spin-flip term can be intuitively understood by looking at Fig. 1a. Since the spin-flip term exchanges $\uparrow$ and $\downarrow$ spin electrons residing in different orbitals, the configuration shown in the middle panel is favored. As a result, the probability for the equal-spin state (right panel) decreases compared to the spin-anisotropic case.

Finally, let us comment on the potential implications of these findings for unconventional multi-band superconductors. Because an Ising-type spin-freezing is un-
derlying the fluctuating-moment induced spin-triplet superconductivity, it may be expected to occur in electron systems with strong spin-orbit coupling. Promising candidates are the uranium-based superconductors UGe$_2$ [14], URhGe [13] and UCoGe [16], which exhibit a superconducting phase bordering a FM phase. In these compounds a strong Ising spin-anisotropy is observed: the magnetization along the easy axis is several times larger than along the hard axis [17]. The $5f$ electrons in the uranium ions, which play a central role in the low-temperature behavior, have a relatively itinerant nature and are strongly correlated. Hence our mechanism could be realized in these uranium-based superconductors.

We furthermore believe that Sr$_2$RuO$_4$ is a candidate compound which might exhibit a fluctuating-moment induced superconductivity. Here, the spin-orbit coupling is nearly 100 meV [10], and as shown in Fig. 4 the spin anisotropy need not be very large to realize superconductivity at low temperatures. Also, the estimated $U \simeq 0.8$ [3] and the filling $n = 4$ (same as $n = 2$ due to particle-hole symmetry) place this material in the paramagnetic phase (Figures 2b and c). A related compound, SrRuO$_3$, with a larger $U$, becomes a ferromagnet [1] and exhibits the non-Fermi liquid behavior associated with spin freezing in the high-temperature phase [7, 8].

In the iron pnictides, the Coulomb interactions and fillings on the Fe sites can also be close to the spin-freezing crossover values [3, 6], and for LiFeAs, in particular, the experimental signatures fully support this interpretation [28]. On the other hand, the 3$d$ electrons have weak spin-orbit coupling, so that the spin anisotropy may not be sufficient for spin-triplet superconductivity.

Apart from insights into the mechanism of superconductivity in the above families of compounds, our work provides a guiding principle in the search for new unconventional multi-band superconductors, namely the combination of emerging local moments in the spin freezing crossover regime and spin anisotropy in heavy elements.

**Methods**

We consider a three-orbital Hubbard model whose Hamiltonian is given by

$$
\mathcal{H} = \sum_{k \sigma} (\varepsilon_k - \mu) c_{k \sigma}^\dagger c_{k \sigma} + U \sum_{i \gamma} n_{i \gamma \uparrow} n_{i \gamma \downarrow} + U' \sum_{i \gamma < \gamma'} n_{i \gamma \sigma} n_{i \gamma' \sigma} + (U' - J) \sum_{i \gamma < \gamma'} n_{i \gamma \sigma} n_{i \gamma' \downarrow} - \alpha J \sum_{i, \gamma < \gamma'} (c_{i \gamma \uparrow} c_{i \gamma' \downarrow} c_{i \gamma' \uparrow} + c_{i \gamma \downarrow} c_{i \gamma' \uparrow} c_{i \gamma' \downarrow} + \text{H.c.}),
$$

(4)

where $i$ is the site index, $\gamma = 1, 2, 3$ the orbital index, $\sigma = \uparrow, \downarrow$ the spin index, and $\bar{\sigma}$ represents the complementary spin ($\uparrow = \downarrow$). $\varepsilon_k$ is the dispersion of electrons on the lattice, and $\mu$ is the chemical potential. The interaction terms contain the intra-orbital ($U$) and inter-orbital ($U'$) Coulomb repulsions, and the Hund coupling $J$. The parameter $\alpha$ controls the anisotropy in spin space, i.e. $\alpha = 1$ corresponds to a spin-rotationally invariant system and $\alpha = 0$ to the Ising anisotropic case where the interactions are only of density-density type. A spin anisotropy may originate from spin-orbit coupling, and the parameter $\alpha$ allows us to incorporate this effect in a simple manner. We use the relation $U' = U - 2J$ which is valid in a spherically symmetric system.

The first and second terms in the last line of Eq. (4) are known as spin-flip and pair-hopping terms, respectively. An anisotropic coupling in spin space should in principle only change the prefactor of the spin-flip term. However, for $J > 0$ the pair-hopping term, which transfers two electrons in the same orbital to another orbital, is not important. This is because the pair-hopping process favors the state shown in the left panel of Fig. 4, which is hardly realized due to the presence of the intra-site Coulomb interaction $U$ which is larger than $J$. We therefore consider it more convenient to put the anisotropy factor in front of both terms, so that $\alpha$ interpolates between the familiar Ising and rotationally invariant limits.

In the DMFT approximation [27], the multi-orbital lattice model (4) is mapped to a self-consistently determined multi-orbital impurity model, whose solution yields an estimate of the local lattice Green’s function and other local observables. If the hopping is orbital-diagonal, this impurity model is described by the action

$$
\mathcal{S} = \sum_{\gamma \sigma} \int d\tau d\tau' c_{i \gamma \sigma}^\dagger(\tau) \Delta_{\gamma \sigma}(\tau - \tau') c_{i \gamma \sigma}(\tau') + \int d\tau \mathcal{H}_{\text{loc}}(\tau),
$$

(5)

where $\Delta_{\gamma \sigma}$ is the hybridization function, and $\mathcal{H}_{\text{loc}}$ corresponds to the local part of the Hamiltonian. Enabling
such a mapping to a single-site effective model is the approximation of a $k$-independent self-energy. This approximation becomes exact in the limit of large dimension or coordination number \[ [30]. \] The only information about the lattice which enters the DMFT calculation is the density of states, and for the semi-circular density of states with bandwidth $W$ used in this study, the self-consistency condition can be expressed as \[ \Delta_{\sigma} = (W/4)^2 G_{\sigma\sigma}, \] with $G_{\sigma\sigma}$ the impurity Green’s function.

Since the local problem (representing a single atom in its hybridized environment) is solved exactly, the DMFT method is suitable for treating strong local correlations. The main task in a DMFT calculation is to obtain the numerical solution of the impurity model \[ [30]. \] We use a continuous-time quantum Monte Carlo (CTQMC) method based on the hybridization expansion \[ [28], \] which samples a formal perturbation expansion of the partition function in powers of $\Delta$. This method can handle arbitrary local interaction terms.

The local self-energy $\Sigma$ obtained by the CTQMC calculation is used to evaluate the lattice Green’s function $G_k$ via the Dyson equation

\[
G_k = G_k^0 + G_k^0 \Sigma G_k, \tag{6}
\]

where the spin/orbital $(\sigma, \gamma)$ and frequency $(\omega)$ indices have been omitted. Here, $G_k^0$ is the non-interacting lattice Green’s function. For the estimation of the renormalization factor, we use the ansatz $\Sigma(\omega \to 0) = a + b\omega$ and determine the coefficients by fitting the numerical data \[ [8]. \] Specifically, we fit the self energy by the Padé approximation using the lowest two Matsubara frequencies, and compute the renormalization factor by the relation

\[
z = (1 - b)^{-1}.
\]

In a similar manner, the vertex part $\Gamma$ of the two-particle Green’s function, which can be regarded as the two-particle version of the self-energy, is also wave-vector-$q$ independent \[ [27]. \] This vertex part can be calculated from the local impurity problem, and the lattice two-particle Green’s function $\chi_q$ is obtained by solving the Bethe-Salpeter equation

\[
\chi_q = \chi_q^0 + \chi_q^0 \Gamma \chi_q. \tag{7}
\]

These quantities should be regarded as matrices with respect to spin/orbital indices and frequency. The susceptibilities in Eq. \[ [9] \] can be derived from $\chi_q$. While we have also calculated the susceptibilities for other types of orders, such as orbital ordering, only the quantities listed in Eq. \[ [30] \] diverge in the parameter regions considered in this paper. In our calculation, the number of Matsubara frequencies is typically taken as 150. Although the value of the susceptibility varies as a function of this cutoff, the divergent points (phase boundaries) are insensitive to it. On the other hand, the local susceptibility in Eq. \[ [1] \] is measured directly in the impurity model.

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