Heavy electrons and the symplectic symmetry of spin

REBECCA FLINT, M. DZERO AND P. COLEMAN*

Center for Materials Theory, Rutgers University, Piscataway, New Jersey 08854, USA *e-mail: coleman@physics.rutgers.edu

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The recent discovery of two heavy-fermion materials $PuCoGa_5$ and $NpPd_5Al_2$, which transform directly from Curie paramagnets into superconductors, reveals a new class of superconductors where local moments quench directly into the superconducting condensate. Unlike other heavy-electron superconductors, where Cooper pairing is thought to be driven by spin fluctuations, these higher-transition-temperature materials do not seem to be close to a magnetic instability. Large-N expansions have been invaluable in describing heavy-fermion metals, but so far cannot treat superconductivity. Here, we introduce a new class of large-N expansion that uses symplectic symmetry to protect the odd time-reversal parity of spin and sustain Cooper pairs as well-defined singlets. We show that when a lattice of magnetic ions exchange spin with their metallic environment in two distinct symmetry channels, they can simultaneously satisfy both channels by forming a condensate of composite pairs between local moments and electrons. In the tetragonal crystalline environment relevant to $PuCoGa_5$ and $NpPd_5Al_2$, the lattice structure selects a natural pair of spin exchange channels, predicting a unique anisotropic paired state with either d- or g-wave symmetry. This pairing mechanism also predicts a large upturn in the NMR relaxation rate above T_c and strong enhancement of Andreev reflection in tunnelling measurements.

A powerful tool in the description of heavy-fermion metals is the large-N expansion, which expands the physics in powers of 1/Nabout a solvable limit where particles carry a large number (N) of spin components. Large-N approximations for interacting electron systems have provided an invaluable tool for understanding heavy-fermion materials¹⁻³ and low-dimensional magnetism^{4,5}, yet so far, they do not encompass heavy-fermion superconductivity. We are motivated to return to this unsolved problem by the discovery of two new singlet heavy-electron superconductors PuCoGa₅ and NpPd₅Al₂ (refs 6,7), which transform from Curie paramagnets into superconductors without first developing a Fermi liquid. In contrast with other heavy-electron superconductors where pairing is thought to be mediated by antiferromagnetic fluctuations⁸⁻¹⁴, these materials do not seem to be close to a magnetic instability. Moreover, the condensation entropy lost on formation of these superconductors is between a quarter and a third of the unquenched spin entropy Rln2 of the Curie paramagnet, indicating that the spin-quenching normally associated with the Kondo effect is an integral part of the development of superconductivity.

The difficulty in developing a large-N description of heavy-fermion superconductivity is that the odd time-reversal parity of the electron spin, $\mathbf{S} \stackrel{\theta}{\longrightarrow} -\mathbf{S}$, is not preserved by SU(N) spins. The inversion of spins under time reversal protects singlet superconductivity, guaranteeing that an electron paired with its time-reversed twin is a singlet. However, if we extend the theory so that the number of spin components exceeds two, the resulting SU(N) spin operators do not all invert under time reversal, so that time-reversed pairs of particles cease to be spin singlets. Here, we show how the time inversion of spins is restored in a new class of symplectic large-N expansions that link time-reversal symmetry to a symplectic property of the electron spin. Our approach enables us to model superconductivity on an equal footing with the Kondo effect and leads us to propose that the

superconductivity in PuCoGa₅ and NpPd₅Al₂ represents a new kind of lattice coherence associated with the Kondo effect, with several observable consequences.

We begin by demonstrating the link between time reversal and symplectic symmetry. Time reversal is an anti-unitary operator θ that acts on an electron wavefunction $\psi_{\sigma}(x,t)$ as $\theta\psi(\mathbf{x},t)=\hat{\epsilon}\psi^*(\mathbf{x},-t)$, where ψ^* is the complex conjugate of the wavefunction and $\hat{\epsilon}_{\alpha\beta}=\tilde{\alpha}\delta_{\alpha,-\beta}$ is the skew-symmetric matrix that interchanges 'up' and 'down' amplitudes, where $\tilde{\alpha}=\mathrm{sgn}(\alpha)$. θ is normally written as a product $\theta=\hat{\epsilon}K$ (ref. 15) with the operator K, which conjugates quantum amplitudes, $K\psi=\psi^*K$. Now consistency of time reversal requires that it commutes with spin rotations, $U\theta=\theta U$, or

$$U \in K U^{\dagger} = \in K$$
.

where U is the unitary rotation operator. But the conjugate of U^{\dagger} is its transpose, $(U^{\dagger})^* = U^{\mathsf{T}}$, so K converts U^{\dagger} into its transpose $KU^{\dagger} = U^{\mathsf{T}}K$, and consistency requires that

$$U\hat{\epsilon}U^{\mathrm{T}} = \hat{\epsilon}.\tag{1}$$

This is the symplectic symmetry of spin: a symmetry that must hold if the spin rotation group is to remain consistent with the concept of time reversal. The unusual appearance of the transpose U^{T} , rather than a Hermitian conjugate U^{\dagger} reflects the anti-unitary nature of time reversal. Replacing U by an infinitesimal rotation, $U=1+i\alpha\cdot\mathbf{S}$, we obtain the inversion of spins under time reversal $\mathbf{S} \overset{\beta}{\to} \theta \mathbf{S} \theta^{-1} = \hat{\mathbf{c}} \ \mathbf{S}^{\mathsf{T}} \hat{\mathbf{c}}^{\mathsf{T}} = -\mathbf{S}$.

At first sight, this entire set of reasoning immediately extends to the case of fields with any even number N=2k of components, $\psi_{\alpha} \equiv (\psi_1 \quad \psi_{-1} \quad \dots \quad \psi_k \quad \psi_{-k})$. Unfortunately, for N>2, the SU(N) group does not satisfy condition (1) for any choice of the

matrix ϵ . To preserve a consistent definition of both time reversal and spin rotation for N>2, we have to choose the symplectic subgroup SP(N), the elements of which are actually defined by the consistency condition (1) with $\hat{\epsilon}_{\alpha\beta} = \tilde{\alpha}\delta_{\alpha,-\beta}$. Seventeen years ago, Read and Sachdev^{16,17} made the observation that the symplectic group SP(N) allows spin operators that form singlet pairs. Their approach has been extensively applied to frustrated magnetism^{18,19}, the t-J model²⁰ and most recently, to paired Fermi gases^{16,17,21,22}. From this discussion, symplectic spins assume an extra importance as the only consistent way to sustain time-inversion symmetry in the large-N limit.

The spin operators of the SU(N) group are written as

$$\mathcal{T}_{\alpha\beta} = \psi_{\alpha}^{\dagger} \psi_{\beta} - \left(\frac{n_{\psi}}{N}\right) \delta_{\alpha\beta},$$

where $n_{\psi} = \sum_{\alpha} \psi_{\alpha}^{\dagger} \psi_{\alpha}$ is the number of particles that make up the spin. Under time reversal, $\mathcal{T}_{\alpha\beta} \stackrel{\theta}{\to} \tilde{\alpha} \tilde{\beta} \mathcal{T}_{-\beta,-\alpha}$, SU(N) spins have no well-defined parity. When ψ_{α} is a Fermi operator, we can also define a charge conjugation operator \mathcal{C} that converts particles into holes $\psi_{\alpha} \stackrel{\mathcal{S}}{\to} \tilde{\alpha} \psi_{-\alpha}^{\dagger}$. By taking antisymmetric or symmetric combinations of the SU(N) spins with their time-reversed version, we may divide them into two sets:

'magnetic' moments,
$$S_{\alpha\beta} = \psi_{\alpha}^{\dagger} \psi_{\beta} - \tilde{\alpha} \tilde{\beta} \psi_{-\beta}^{\dagger} \psi_{-\alpha}$$
, $(\theta, \mathcal{C}) = (-, +)$,

which invert under time reversal but are neutral under charge conjugation, and

'electric' dipoles,
$$\mathcal{P}_{\alpha\beta} = \psi_{\alpha}^{\dagger}\psi_{\beta} + \tilde{\alpha}\tilde{\beta}\psi_{-\beta}^{\dagger}\psi_{-\alpha},$$
 $(\theta, \mathcal{C}) = (+, -),$

which are invariant under time reversal, but change sign under charge conjugation²³. There are D = (1/2)N(N+1) magnetic moments that form the generators of SP(N). For the physical case of N=2, there are no dipoles, but as N becomes large, the SU(N) group contains approximately equal numbers of 'moments' and 'dipoles'.

If we are to sustain the time-reversal properties of spin in a large-N expansion, we must use Hamiltonians that do not contain the unwanted dipole moment operators. This requirement differentiates the new approach from earlier SP(N) treatments of spin systems. As the magnetic moments form a closed SP(N) algebra, their spin dynamics (dS/dt) = -i[H[S], S] will remain closed and decoupled from the electric dipoles provided the Hamiltonian H contains no dipole moments \mathcal{P} that spoil the closure. When the local moments are built out of fermions, this closure gives rise to a particularly important gauge symmetry. SU(N) spins commute with the number operator $n_{j\psi}$ at each site, giving rise to a U(1) gauge invariance that features heavily in many analyses of correlated electron physics. However, symplectic spins also commute with the fermion pair operator $\Psi_j = \sum_{\alpha} \tilde{\alpha} \psi_{j,-\alpha} \psi_{j\alpha}$ because it is an SP(N) singlet,

$$[S_{\alpha\beta}, \Psi_i] = [S_{\alpha\beta}, \Psi_i^{\dagger}] = [S_{\alpha\beta}, n_{ijl}] = 0.$$

In a lattice, these symmetries apply independently at every spin site j, giving rise to an SU(2) local gauge invariance. This symmetry was first identified for spin-1/2 by Affleck $et\,al.^{24}$, who argued for its central role in defining the neutrality of spin. Symplectic spins allow us to extend this gauge symmetry to large N, provided we exclude dipole moment operators from the Hamiltonian. This is a more stringent requirement than enforcing global SP(N) symmetry,

and to delineate it from earlier SP(N) approaches, we refer to it as 'symplectic-N'.

A key distinction between earlier SP(N) approaches and symplectic-N lies in the way spin interactions are decoupled. The 'dot product' of symplectic spins $S_i \cdot S_j = (1/2)S_{\alpha\beta}(i)S_{\beta\alpha}(j)$ has a unique decoupling in terms of both particle—hole and singlet pairs (see the Supplementary Information):

$$\mathbf{S}_1 \cdot \mathbf{S}_2 = -B_{21}^{\dagger} B_{21} + \eta_{\psi} A_{21}^{\dagger} A_{21}, \qquad (\eta_{\psi} = \pm),$$

where $B_{21}^\dagger = \sum_\sigma \tilde{\sigma} \psi_{2\sigma}^\dagger \psi_{1-\sigma}^\dagger$ creates a valence bond of spins between sites one and two, whereas $A_{21} = \sum_\sigma \psi_{2\sigma}^\dagger \psi_{1\sigma}$ 'resonates' valence bonds between sites. The signature η_ψ depends on whether the field ψ is a boson $(\eta_\psi = +1)$ or fermion $(\eta_\psi = -1)$. In the large-N limit, these bond variables acquire expectation values. For N=2, this kind of decoupling²⁵ is one of many alternative schemes considered earlier by authors in the context of frustrated magnetism. The standard SP(N) decoupling procedure omits the last term and is equivalent to a Hamiltonian containing a mixture of spins and dipoles, $H=J\sum_{i,j}(S_i\cdot S_j-\mathcal{P}_i\cdot \mathcal{P}_j)$. Removing the dipoles from the Hamiltonian gives rise to a much broader class of mean-field theories, with potential applications in the realm of both frustrated magnetism and strongly correlated metals and superconductors.

Here however, we focus on the application of symplectic-N to heavy-fermion superconductivity in $PuCoGa_5$ and $NpPd_5Al_2$ (refs 6,7), where the appearance of an SU(2) gauge symmetry in the fermionic symplectic-N approach has marked physical consequences. These materials contain a lattice of local moments, immersed in a sea of electrons to form a 'Kondo lattice'. We assume that at low temperatures, the Pu and Np ions in these materials behave as Kramers's doublets. The exchange of spin with its environment involves virtual valence fluctuations into ionic configurations with one more, or one less f-electron: $f^n = f^{n\pm 1} = e^-$, where n=3 and 5 for $NpPd_5Al_2$ and $PuCoGa_5$ respectively. In complex magnetic ions, the partial-wave symmetries of these two virtual processes are distinct²⁶ (see the Methods section), giving rise to two independent Kondo screening channels, $\Gamma = 1, 2$, described by the following model (see the Supplementary Information)

$$\hat{H} = \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}} c_{\mathbf{k}\sigma}^{\dagger} c_{\mathbf{k}\sigma} + \frac{1}{N} \sum_{j} \left[J_{1} \psi_{1\mathbf{k}\alpha}^{\dagger} \psi_{1\mathbf{k}'\beta} + J_{2} \psi_{2\mathbf{k}\alpha}^{\dagger} \psi_{2\mathbf{k}'\beta} \right] \times S_{\beta\alpha}(j) e^{i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{R}_{j}}.$$
(2)

To develop a solvable mean-field theory, we examine the family of models where

$$\hat{S}_{\alpha\beta}(j) = f_{j\alpha}^{\dagger} f_{j\beta} - \tilde{\alpha} \, \tilde{\beta} f_{j-\beta}^{\dagger} f_{j-\alpha}$$

are the N component symplectic representation of the magnetic moment at each site j. The physical system corresponds to the limit N=2. The quantities J_1 and J_2 describe two unequal antiferromagnetic exchange couplings in the two symmetry channels and the operators $\psi^{\dagger}_{\Gamma \mathbf{k}\alpha} = [\Phi_{\Gamma}(\mathbf{k})]_{\alpha\sigma}c^{\dagger}_{\mathbf{k}\sigma}$ create electrons in a partial-wave state with symmetry $\Gamma=1,2$, where the matrices $\Phi_{1\mathbf{k}}$ and $\Phi_{2\mathbf{k}}$ are set by the crystal field symmetry.

In the tetragonal crystal symmetry environment of NpPd₅Al₂ and PuCoGa₅, the magnetic moments are surrounded by a cage of Pd and Ga ligand atoms, respectively. Electrons that interact with the magnetic moment must scatter in one of three tetragonal crystal field doublets, labelled $\{\Gamma_7^+, \Gamma_7^-, \Gamma_6\}$. The Γ_7^\pm states connect selectively with the in-plane and out-of-plane ligand atoms, as shown in Fig. 1. The Γ_6 crystal field state is aligned along the c axis, overlapping weakly with the nearby ligand ions. This leads us to propose a model in which the two Γ_7^\pm channels dominate the spin exchange with the f-electrons.

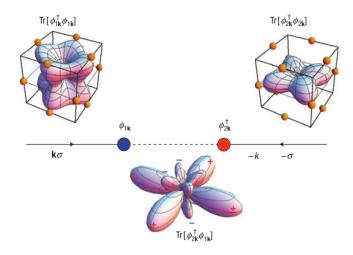


Figure 1 Composite pairing in two crystal field screening channels. Top: Angular dependence of the hybridization functions $|\Phi_{\Gamma k}|^2$ for the two tetragonal crystal field scattering states, Γ_7^+ and Γ_7^- for j=5/2. The figures show the crystal field configurations with maximum overlap with the out-of-plane and in-plane ligand atoms. Bottom: Formation of a composite pair leads to Andreev scattering of conduction electrons between the two channels, generating a superconducting gap with symmetry given by the trace of the overlap between the two hybridization functions (see Methods, equation (8)). The transfer of four units of orbital angular momentum that accompanies this process leads to a gap with l=4, 'q-wave' symmetry.

The presence of two distinct scattering channels plays a central role in our model. For a single magnetic Kondo ion, the strongest spin-screening channel always dominates, forming a local Fermi liquid of the corresponding symmetry. At $J_1 = J_2$, the two channels are perfectly balanced, giving rise to a critical state in which the spin screening fluctuates between the channels. Many groups have speculated that in a lattice environment, the spins will attempt to avoid this critical state through the development of superconductivity (refs 26,27,29, and ref. 28 and references therein). Symplectic-N enables us to develop the first controlled realization of this conjecture, which we apply to the new superconductors PuCoGa₅ and NpPd₅Al₂.

When we expand the Kondo interaction in equation (2), we obtain $H_1 = \sum_{\Gamma=1,2} H_{\Gamma}(j)$, where

$$H_{\Gamma}(j) = -\frac{J_{\Gamma}}{N} \left[(\psi_{j\Gamma}^{\dagger} f_j) (f_j^{\dagger} \psi_{j\Gamma}) + (\psi_{j\Gamma}^{\dagger} \epsilon^{\dagger} f_j^{\dagger}) (f_j \epsilon \psi_{j\Gamma}) \right]$$
(3)

describes the spin exchange at site j in channels $\Gamma=1,2$ and $\psi_{j\Gamma}^{\dagger}=\sum_{\bf k}\psi_{{\bf k}\Gamma}^{\dagger}{\rm e}^{-i{\bf k}\cdot{\bf R}_j}$ creates an electron in a Wannier state of symmetry Γ at site j. This interaction exhibits the local SU(2) gauge symmetry $f_{\sigma}\to\cos\theta f_{\sigma}+\sin\theta\tilde{\sigma}f_{-\sigma}^{\dagger}$. The important point here is that this symmetry survives for all even N. Earlier efforts have been made to develop SU(2) gauge theories of heavy-electron systems²⁹ and high-temperature superconductors³⁰, but were not justified in terms of a controlled expansion.

When the Kondo interaction (3) is factorized, it decouples into a Kondo hybridization V and pairing field Δ as follows

$$H_{\Gamma}(j) \rightarrow \sum_{\sigma} \left[\left(f_{\sigma}^{\dagger} V_{\Gamma} + \tilde{\sigma} f_{-\sigma} \Delta_{\Gamma} \right) \psi_{\Gamma \sigma} + \text{h.c.} \right] + N \left(\frac{|V_{\Gamma}|^{2} + |\Delta_{\Gamma}|^{2}}{J_{\Gamma}} \right), \tag{4}$$

where we have suppressed the site indices j. The mean-field Hamiltonian defined by this decoupling becomes exact in the large-N limit. Despite the appearance of 'pairing terms' in the Kondo interaction, the formation of Kondo singlets in a single channel does not lead to superconductivity; an SU(2) gauge transformation on the f-electron can always absorb the pairing term Δ into a redefinition of the f-electron: $(f_\sigma^\dagger V_\Gamma + \tilde{\sigma} f_{-\sigma} \Delta_\Gamma) \to V_0 \tilde{f}^\dagger_{\ \sigma}$. For a one-channel Kondo lattice, the symplectic-N and SU(N) large-N limits are thus identical.

For two channels, this is no longer the case. Here, it is convenient to define a Nambu spinor for the f-electrons and two corresponding matrix SU(2) order parameters

$$\tilde{f}_{j\sigma} = \begin{pmatrix} f_{\sigma} \\ \tilde{\sigma} f_{-\sigma}^{\dagger} \end{pmatrix}_{i}, \qquad \mathcal{V}_{\Gamma j} = \begin{pmatrix} V_{\Gamma} & \bar{\Delta}_{\Gamma} \\ \Delta_{\Gamma} & -\bar{V}_{\Gamma} \end{pmatrix}_{i},$$

which transform identically under an SU(2) gauge transformation g_j , $\tilde{f}_j \to g_j \tilde{f}_j$, $\mathcal{V}_{\Gamma j} \to g_j \mathcal{V}_{\Gamma j}$. The product $\mathcal{V}_{2j}^+ \mathcal{V}_{1j}$ forms an SU(2) invariant quantity with off-diagonal component $\Psi_j = (V_{1j} \Delta_{2j} - V_{2j} \Delta_{1j})$: this quantity preserves the local SU(2) invariance, but it breaks the global U(1) gauge invariance associated with physical charge, and plays the role of a superconducting order parameter. If we carry out an SU(2) gauge transformation that removes the Δ_{1j} , the composite order parameter $\Psi_j = V_{1j} \Delta_{2j}$. These terms lead to Andreev reflection off the screened Kondo impurity, as shown in Fig. 1, bottom.

Physically, we may understand this phenomenon as the consequence of the formation of a condensate of composite pairs: pairs of electrons, bound to magnetic impurities. When an electron scatters off a magnetic impurity, the quantity Ψ determines the amplitude for emitting an Andreev hole, leaving behind a composite pair. Detailed analysis (see the Methods section) confirms this insight and demonstrates the formation of composite order with expectation value

$$\langle N_e - 2|\psi_{\perp}^1(j)\psi_{\perp}^2(j)S_f^+(j)|N_e\rangle \propto (V_{1j}\Delta_{2j} - V_{2j}\Delta_{1j}),$$

where $|N_e\rangle$ and $|N_e-2\rangle$ refer to the ground states containing N_e and N_e-2 electrons respectively. In a single-impurity model, this order parameter is forbidden, because electrons cannot change scattering channels, but in the lattice, electrons travelling between sites no longer conserve the channel index, which permits composite order. Once composite order develops, the Kondo singlets resonate between the two screening channels, and the resulting resonance energy stabilizes the coherent state.

This basic mechanism was first proposed in ref. 29; our large-N analysis provides the first solvable limit in which this mechanism can be rigorously validated, while at the same time incorporating the detailed spin–orbit physics of the crystal-field-split screening channels. When we evaluate the resonant Andreev scattering from the Feynman diagram shown in Fig. 1, we find that the conduction electrons acquire a gap that is proportional to the overlap of the two hybridization functions $\Delta_{\rm e}({\bf k}) \propto {\rm Tr}[\varPhi_{2k}^{\dagger} \varPhi_{1k}]$, as shown in Fig. 1. One of the fascinating features of this gap is that the orthogonality of \varPhi_{2k} and \varPhi_{1k} guarantees the presence of line nodes in the gap, despite their absence in the underlying hybridization functions.

We have computed the superconducting transition temperature T_c as a function of the ratio J_2/J_1 using symplectic-N (see the Methods section). Figure 2 shows the results of a model calculation for a two-dimensional Kondo lattice, assuming uniform expectation values for \mathcal{V}_{Γ} . It is instructive to contrast the phase diagrams of the SU(N) and symplectic large-N limits. In the former, there is a single quantum phase transition that separates the heavy-electron Fermi liquids formed via a Kondo effect about the strongest channel. In the symplectic treatment, this quantum critical point is immersed beneath a superconducting 'dome'. The

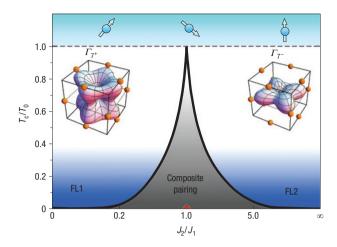


Figure 2 Phase diagram for a two-channel Kondo lattice, computed in the symplectic large-N limit for tetragonal symmetry, where spin is exchanged via channels $\Gamma_1 \equiv \Gamma_7^+$ and $\Gamma_2 \equiv \Gamma_7^-$. The X-axis coordinate is the parametric variable $X = 2(J_2/J_2)/(1+J_2/J_1)$ running from X=0 to 2, corresponding to J_2/J_1 running from zero to infinity, as labelled. Temperature is measured in units of the maximum Kondo temperature $T_0 = \max(T_{K1}, T_{K2})$. Fermi liquids of different symmetry develop for small, or large J_2/J_1 , separated by a region of composite pairing, delineated in grey. The red circle indicates the single-impurity quantum critical point that develops when the two channels are degenerate, which is avoided in the lattice through the development of composite pairing.

Cooper channel in the heavy-electron normal state guarantees that the secondary screening channel is always marginally relevant in the lattice, as first speculated in ref. 29. This is, to our knowledge, the first controlled mean-field theory in which the phenomenon of 'avoided criticality' gives rise to superconductivity.

There are three concrete consequences of composite pairing that permit our ideas to be compared to experiment.

- (1) Crystal fields determine the gap symmetry. When an electron Andreev scatters between the $\Gamma = \Gamma_7^\pm$ channels, it scatters between the $|\mp 3/2\rangle \leftrightarrow |\pm 5/2\rangle$ states, and in so doing picks up $|\Delta m_z| = 4$ units of angular momentum via the spin-orbit interaction. $\Gamma_7^+ \otimes \Gamma_7^-$ composite pairs thus carry l=4 units of angular momentum, giving rise to a g-wave gap symmetry with eight nodal surfaces, as shown in Fig. 1. In contrast, if the second channel had Γ_6 symmetry then the $|\mp 3/2\rangle \leftrightarrow |\pm 1/2\rangle$ scattering involves transfer of $|\Delta m_z| = 2$ units of angular momentum, so $\Gamma_7^+ \otimes \Gamma_6$ composite pairs have d-wave symmetry, with four nodal surfaces.
- (2) Upturn in the NMR. In the approach to the composite ordering transition, the local moments must correlate between sites, and this manifests itself through the development of an enhancement of the NMR relaxation rate. For those systems with maximal T_c , the NMR relaxation rate in the normal state is predicted to contain a term derived from the interference between the two screening channels, proportional to the product $(1/T_1T) \propto J_1(T)J_2(T)$ of the temperature-renormalized Kondo coupling constants (see the Supplementary Information). This gives rise to an upturn in the NMR relaxation rate $(1/T_1T) \propto [\ln^2(T/T_c) + \pi^2]^{-1}$ (see Fig. 3), a result in accord with recent measurements on PuCoGa₅ (ref. 31), but which has yet to be tested in NpPd₅Al₂.
- (3) Andreev reflection. The main driver for this mechanism of heavy-fermion superconductivity is Andreev reflection off quenched magnetic moments. Conventional Andreev reflection involves the direct transfer of an electron from

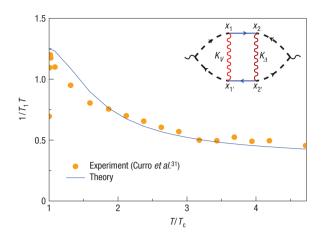


Figure 3 Upturn in the NMR relaxation rate created by the cooperative interference of the Kondo effect in two channels at different sites, for the extreme case of maximum T_c , where $J_1=J_2$ (blue line), compared with measured NMR relaxation rate in $PuCoGa_5$ (ref. 31) (yellow circles). Inset: The Feynman diagram used to compute this contribution, where dotted lines describe the f-fermions, blue lines describe conduction electrons propagating between sites and the wavy lines describe the Kondo interaction in the particle—hole (red) and particle—particle (blue) channels at different sites. Temperature is measured in units of the transition temperature T_c .

the probe into the pair condensate of the conduction sea. The Blonder–Tinkham–Klapwijk theory of Andreev reflection³² predicts such processes are severely suppressed by the large mismatch between the probe and heavy-electron group velocities. However, in a heavy-electron system, an electron can also 'cotunnel' into the Kondo lattice—a process well known in magnetic quantum dots, whereby the electron flips a localized spin as it tunnels into the material³³. In a composite paired superconductor, these processes will result in the direct absorption of the electron into the condensate of composite pairs, providing an enhanced contribution to the Andreev tunnel current with a Fano resonant structure³⁴.

(4) Internal proximity effect. When the Kondo ions in the superconductor are substituted by Kondo ions with a larger Kondo temperature, provided the gap symmetry is unchanged, this is expected to lead to an internal proximity effect, where the Andreev reflection off the substituted impurities enhances the superconducting T_c . This effect requires an overlap between the gap functions of the two different ions, but this is guaranteed by crystal symmetry, providing the same screening channels are operative for both ions. For instance, Pu doping of NpPd₅Al₂ may lead to an enhancement of the superconducting T_c .

Our work leaves open the question of the relationship between superconductivity in NpPd₅Al₂ and PuCoGa₅ and other 115 superconductors with lower transition temperatures, such as PuRhGa₅ (ref. 35) and CeXIn₅ (X = Rh, Ir, Co) (refs 36–38). PuRhGa₅ is a 9 K superconductor where the local moments seem to quench before superconductivity develops. This system fits naturally into our picture as a mixed-valent partner of PuCoGa₅ where a more asymmetric coupling ratio $J_1/J_2 > 1$ causes a Kondo effect to develop in the strongest channel before composite pair formation. In contrast, superconductivity in the cerium 115 materials seems to develop in the vicinity of an antiferromagnetic quantum critical point. Conventional wisdom attributes the superconductivity to Cooper pairing between intinerant f-electrons, mediated by the exchange of soft magnetic

fluctuations^{11–13}. Yet in CeCoIn₅ and CeIrIn₅, the observation of crystal field excitations³⁹ and the presence of a sharp upturn in magnetic susceptibility before superconductivity^{37,38} suggest the presence of local moments at the superconducting transition, a situation tending to favour composite, rather than Cooper pair formation. But could antiferromagnetism play a role in the formation of composite pairs? In fact, critical magnetic interactions have long been known to induce extra spin-screening channels into the Kondo effect^{40,41}. This suggests the intriguing possibility that antiferromagnetic fluctuations in the cerium-115 materials may drive superconductivity by opening up secondary screening channels for composite pairing with the local moments.

METHODS

TWO-CHANNEL KONDO MODEL

Our two-channel Kondo lattice model assumes that the ground state of an isolated magnetic ion is a Kramers's doublet $|\Gamma_1\sigma\rangle$ containing an odd number n of f-electrons. For example, the Pu³+ ion in PuCoGa⁵ is a single f-hole in a filled j=5/2 atomic shell, forming a $|5f^5\rangle$ Kramers's doublet with n=5. We assume that the dominant spin fluctuations occur via valence fluctuations into singlet states

$$\begin{array}{ccc}
|0\rangle & \rightleftharpoons & |\Gamma_1 \sigma\rangle & \rightleftharpoons & |\phi\rangle \\
f^{n+1} & f^n & f^{n-1}
\end{array}$$

To illustrate the situation, consider PuCoGa₅, where $|0\rangle \equiv |f^6\rangle$ is an empty j=5/2 f-shell. In a tetragonal crystal environment, the j=5/2 multiplet of f-electrons splits into three Kramers's doublets: $\{\Gamma_7^+, \Gamma_7^-, \Gamma_6\}$. The f^5 Kramers's doublet can be written as $|\Gamma_1\sigma\rangle = f_{\Gamma_1\sigma}^{\dagger}|0\rangle$, where $f_{\Gamma\sigma}^{\dagger}$ creates an f-hole in one of these three crystal field states. To form a low-energy f^4 singlet, the strong Coulomb interaction between f-electrons forces us to add a second f-hole in a different crystal field channel Γ_2 . We assume that this state is a singlet,

$$|\phi\rangle \equiv |\Gamma_2 \otimes \Gamma_1\rangle_s = \frac{1}{\sqrt{2}} \sum_{\sigma = \pm 1} \operatorname{sgn}(\sigma) f_{\Gamma_2 \sigma}^{\dagger} f_{\Gamma_1 - \sigma}^{\dagger} |0\rangle.$$

In practice, there are many other excited states, but these are the most relevant, because they are the only states that generate antiferromagnetic Kondo interactions and scale to strong coupling. In a conventional Anderson model, Γ_2 and Γ_1 are the same channel, but Hund's coupling forces Γ_1 and Γ_2 to be different, and it is this physics that introduces new symmetry channels into the charge fluctuations.

COMPOSITE PAIRING

The SU(2) invariant order parameter of the composite paired state is $(\mathcal{V}_2^{\dagger}\mathcal{V}_1)$. We shall show that this matrix is equal to the amplitudes for composite pairing and hybridization using a path integral approach. It proves useful to use the following matrix representation for the conduction and f-fields at each site j

$$\begin{split} \hat{F}_{j} &= \begin{pmatrix} f_{j1} & f_{j-1} & \dots & f_{jk} & f_{j-k} \\ f_{j-1}^{\dagger} & -f_{j1}^{\dagger} & \dots & f_{j-k}^{\dagger} & -f_{jk}^{\dagger} \end{pmatrix}, \\ \hat{\Psi}_{\varGamma j} &= \begin{pmatrix} \psi_{\varGamma j1} & \psi_{\varGamma j-1} & \dots & \psi_{\varGamma jk} & \psi_{\varGamma j-k} \\ \psi_{\varGamma j-1}^{\dagger} & -\psi_{\varGamma j1}^{\dagger} & \dots & \psi_{\varGamma j-k}^{\dagger} & -\psi_{\varGamma jk}^{\dagger} \end{pmatrix}. \end{split}$$

Here k = N/2, so these are N by 2 matrices with columns that are Nambu spinors. In the following, we drop the site index, j. This notation can be used to recast the hybridization terms in a more compact form.

$$\frac{1}{2} \sum_{\alpha} \tilde{\psi}_{\Gamma \alpha} \mathcal{V}_{\Gamma} \tilde{f}_{\alpha} = \frac{1}{2} \text{Tr} \left[\hat{\Psi}_{\Gamma}^{\dagger} \mathcal{V}_{\Gamma}^{\dagger} \hat{F} \right] = -\frac{1}{2} \text{Tr} \left[U_{\Gamma} \mathcal{V}_{\Gamma}^{\dagger} \right],$$

where $U_\Gamma \equiv \hat{F}\,\hat{\Psi}_T^\dagger$ is a two-dimensional matrix operator. The decoupled interaction Hamiltonian can now be compactly written as

$$H_{K} = -\frac{1}{2} \sum_{r} \left(\text{Tr} \left[U_{\Gamma} V_{\Gamma}^{\dagger} \right] + \text{h.c.} \right) + \frac{N}{2I_{\Gamma}} \text{Tr} \left[V_{\Gamma} V_{\Gamma}^{\dagger} \right]. \tag{5}$$

To evaluate $(\mathcal{M}) \equiv (\mathcal{V}_2^{\dagger} \mathcal{V}_1)$, we add a source term $\text{Tr} [\eta \mathcal{M}] + \text{h.c.}$ to the Hamiltonian. By varying η , we can then read off $\mathcal{M}_{ab} = \delta H/\delta \eta_{ab}$. Combining the source term with the final trace in equation (5), we obtain

$$H_K[\eta] = -\frac{1}{2} \sum_{\Gamma} \left(\text{Tr} \left[U_{\Gamma} \mathcal{V}_{\Gamma}^{\dagger} \right] + \text{h.c.} \right) + \frac{1}{2} \text{Tr} \left[\mathcal{V}_{\Gamma} \begin{bmatrix} N/J_1 & 2\eta \\ 2\bar{\eta} & N/J_2 \end{bmatrix} \mathcal{V}_{\Gamma'}^{\dagger} \right].$$

After carrying out the Gaussian integral over V_{Γ} , we expand $H_K[\eta]$ to first order in η ,

$$H_K[\eta] = -\sum_{\Gamma} \left(\frac{J_{\Gamma}}{2N}\right) \text{Tr}\left(U_{\Gamma} U_{\Gamma}^{\dagger}\right) + \frac{J_1 J_2}{N^2} \text{Tr}\left[\eta U_2^{\dagger} U_1 + \text{h.c.}\right].$$

Differentiating with respect to η then gives $(\mathcal{V}_2^{\dagger}\mathcal{V}_1) = (J_1J_2/N^2)U_2^{\dagger}U_1 = (J_1J_2/N^2):[\hat{\Psi}_2\hat{F}^{\dagger}\hat{F}\,\hat{\Psi}_1^{\dagger}]:$, where colons denote normal ordering. Using the identity: $\hat{F}^{\dagger}\hat{F}:=\mathbf{S}\cdot\boldsymbol{\sigma}_N^T$, which can be found by expanding the symplectic spins, we obtain

$$\langle \mathcal{V}_{2}^{\dagger} \mathcal{V}_{1} \rangle = -\frac{J_{1}J_{2}}{N^{2}} \begin{bmatrix} \langle \psi_{1}^{\dagger}(\boldsymbol{\sigma}_{N} \cdot \mathbf{S})\psi_{2} \rangle & \langle \psi_{1}^{\dagger}(\boldsymbol{\sigma}_{N} \cdot \mathbf{S})\epsilon\psi_{2}^{\dagger} \rangle \\ \langle \psi_{1}\epsilon^{T}(\boldsymbol{\sigma}_{N} \cdot \mathbf{S})\psi_{2} \rangle & \langle \psi_{1}\epsilon^{T}(\boldsymbol{\sigma}_{N} \cdot \mathbf{S})\epsilon\psi_{2}^{\dagger} \rangle \end{bmatrix}. \tag{6}$$

In our mean-field theory, we chose $V_1 = v_1$, $V_2 = \Delta_2 \tau_2$, so that the composite order parameter is given by the off-diagonal term in equation (6) $\langle \psi_1^{\dagger}(\boldsymbol{\sigma}_N \cdot \mathbf{S}) \epsilon \psi_2^{\dagger} \rangle = (N^2/J_1J_2)v_1\Delta_2$.

NODAL STRUCTURE OF THE GAP

Composite pairing manifests itself as the development of an Andreev reflection component to the resonant scattering off magnetic impurities. We capture this scattering in the mean-field theory by integrating out the f-electrons, which leads to a conduction electron Green's function of the form

$$G(\kappa)^{-1} = \omega - \epsilon_{\mathbf{k}} \tau_3 - \Sigma(\kappa), \qquad \Sigma(\kappa) = \mathcal{V}_{\mathbf{k}}^{\dagger} (\omega - \lambda \tau_3)^{-1} \mathcal{V}_{\mathbf{k}},$$

where $\kappa \equiv (\mathbf{k}, \omega)$. The hybridization matrices are written as $\mathcal{V}_{\mathbf{k}} = \mathcal{V}_{\mathbf{l}} \hat{\Phi}_{\mathbf{l}\mathbf{k}} + \mathcal{V}_{\mathbf{l}} \hat{\Phi}_{\mathbf{2}\mathbf{k}}$ where we will take $\mathcal{V}_{\mathbf{l}} = i \nu_{\mathbf{l}}$ and $\mathcal{V}_{\mathbf{l}} = \Delta_{\mathbf{l}} \tau_{\mathbf{l}}$.

The matrix $\hat{\Phi}_{\Gamma k}$ takes the form $\phi_{\Gamma k}\mathcal{U}_{\Gamma k}$, where $\phi_{\Gamma k}$ is a scalar and $\mathcal{U}_{\Gamma k}$ is a two-dimensional unitary matrix defining the interconversion between Bloch states and spin—orbit-coupled Wannier states. When an electron 'enters' the Kondo singlet, its spin quantization axis is rotated according to the matrix $\mathcal{U}_{\Gamma k}$. When it leaves the ion in the same channel, this rotation process is undone, and the net hybridization matrix $\hat{\Phi}_{\Gamma k}^{\dagger}\hat{\Phi}_{\Gamma k} = (\phi_{\Gamma k})^2$ 1 is spin-diagonal. However, in the presence of composite pairing, an incoming electron in channel 1 can Andreev scatter from the ion as a hole in channel 2. This leads to a net rotation of the spin quantization axis through an angle ζ_k about an axis \mathbf{n}_k that both depend on the location on the Fermi surface, as follows

$$\hat{\Phi}_{2\mathbf{k}}^{\dagger} \cdot \hat{\Phi}_{1\mathbf{k}} = \phi_{2\mathbf{k}} \phi_{1\mathbf{k}} \Big[c_{\mathbf{k}} + i s_{\mathbf{k}} (\mathbf{n}_{\mathbf{k}} \cdot \boldsymbol{\sigma}) \Big],$$

where $c_k = \cos(\zeta_k/2)$ and $s_k = \sin(\zeta_k/2)$.

When we expand the self-energy, we obtain both normal and Andreev components, $\Sigma = \Sigma_N + \Sigma_A$, where the Andreev terms are given by

$$\Sigma_{A}(\kappa) = \left(\frac{i\omega\tau_{2} - \lambda\tau_{1}}{(\omega^{2} - \lambda^{2})} \Phi_{2\mathbf{k}}^{\dagger} \Phi_{1\mathbf{k}} + \text{h.c.}\right)$$

$$= \frac{2\nu_{1\mathbf{k}}\nu_{2\mathbf{k}}}{(\omega^{2} - \lambda^{2})} \left[-\lambda c_{\mathbf{k}}\tau_{1} + \omega s_{\mathbf{k}}(\mathbf{n}_{\mathbf{k}} \cdot \boldsymbol{\sigma})\tau_{2}\right], \tag{7}$$

defining $v_{1k} = v_1 \phi_{1k}$ and $v_{2k} = \Delta_2 \phi_{2k}$. Notice how the Andreev scattering equation (7) contains two terms: (1) a scalar term $(2v_{1k}v_{2k}/(\lambda^2 - \omega^2))\lambda c_k \tau_1$ that is finite at the Fermi energy $(\omega = 0)$, with gap symmetry of the form

$$\Delta_{\mathbf{k}} \propto \operatorname{Tr} \Phi_{2\mathbf{k}}^{\dagger} \Phi_{1\mathbf{k}} \sim \phi_{1\mathbf{k}} \phi_{2\mathbf{k}} c_{\mathbf{k}}$$
 (8)

and (2) a 'triplet' term $-\omega(2\nu_{1k}\nu_{2k}/(\lambda^2-\omega^2))[s_k(\mathbf{n_k}\cdot\boldsymbol{\sigma})\tau_2]$ that is odd in frequency and vanishes on the Fermi surface.

In a tetragonal crystal field environment, the Kramers's doublets are given by $|\Gamma\pm\rangle = |jm\rangle\langle jm|\Gamma\pm\rangle$, (j=5/2), where

$$\begin{split} &\Gamma_6: \quad f_{\Gamma_6\pm}^\dagger = |\pm 1/2\rangle, \\ &\Gamma_7^+: \quad f_{\Gamma_7^+\pm}^\dagger = \cos\beta |\mp 3/2\rangle + \sin\beta |\pm 5/2\rangle, \\ &\Gamma_7^-: \quad f_{\Gamma_7^-\pm}^\dagger = \sin\beta |\mp 3/2\rangle - \cos\beta |\pm 5/2\rangle, \end{split}$$

where β is determined by the crystal field configuration. The overlap of the Bloch states with the spin–orbit-coupled Wannier states $|jm\rangle$ is given by $\langle jm|\mathbf{k}\sigma\rangle=\langle jm|lm-\sigma,(1/2)\sigma\rangle Y_{m-\sigma}^3(\hat{\mathbf{k}})$.

The form factors of the Wannier states are thus $[\Phi_{\Gamma \mathbf{k}}]_{\alpha\sigma} = \sum_{m} \langle \Gamma \alpha | jm \rangle \langle jm | \mathbf{k} \sigma \rangle$.

The scalar form factors $\phi_{\Gamma\mathbf{k}}$ are given by $(\phi_{\Gamma\mathbf{k}})^2=(1/2)\mathrm{Tr}[\Phi_{\Gamma\mathbf{k}}^\dagger\Phi_{\Gamma\mathbf{k}}]$. Generically these can have point, but not line nodes, and the nodes of the pair wavefunction (8) are determined by the function $c_\mathbf{k}$. When an electron Andreev reflects through one hybridization channel into the other, it acquires orbital angular momentum. For example, the 'up' states of the $\Gamma_7^+\sim|-3/2\rangle$ and $\Gamma_7^-\sim|+5/2\rangle$ differ by $\Delta m=4$, so the resulting gap has g-wave symmetry. In contrast, the up states of the $\Gamma_7^+\sim|-3/2\rangle$ and $\Gamma_6\sim|+1/2\rangle$ differ by $\Delta m=2$, leading to d-wave symmetry. These are the two symmetry classes of superconductor possible in our model, one formed from $\Gamma_7^+\otimes\Gamma_7^-$, the other from $\Gamma_7\otimes\Gamma_6$. In our model, we choose $\Gamma_1\equiv\Gamma_7^+$, which maximizes the overlap with the out-of-plane ligand atoms and $\Gamma_2\equiv\Gamma_7^-$ corresponding to the in-plane atoms. In this case, we find the gap has the form

$$\phi_{1\mathbf{k}}\phi_{2\mathbf{k}}c_{\mathbf{k}} = \cos(2\beta)\Delta_{g1}(\mathbf{k}) - \sin(2\beta)\Delta_{g2}(\mathbf{k}),$$

where Δ_{g1} and Δ_{g2} are g-wave gap functions of the form

$$\Delta_{g1}(\mathbf{k}) = \frac{\sqrt{5}}{16\pi}\cos(4\phi)\sin^2[\theta],$$

$$\Delta_{g2}(\mathbf{k}) = \frac{3}{16\pi} \sin^2(\theta) (3 + \cos(2\theta)).$$

COMPUTING THE SUPERCONDUCTING TRANSITION TEMPERATURE

To compute the composite pairing transition, we solve the mean-field equations for the temperature at which the second channel turns on. To obtain the mean-field equations, we first find and diagonalize the mean-field Hamiltonian, $H_{\rm MF}$, which is found by inserting the decoupling (4) in the Kondo Hamiltonian (2). Next, we add a Lagrange multiplier, λ , to constrain the average number of f-electrons at each site, $\lambda(\sum_{\alpha}f_{j\alpha}^{\dagger}f_{j\alpha}-(N/2))$. $H_{\rm MF}$ is then Fourier transformed into \hat{k} space and diagonalized. We have used the gauge choice of $\mathcal V$ from the previous section and assume uniform expectation values.

The eigenvalues of H_{MF} are then given by $\omega = (\omega_{\mathbf{k}\pm}, -\omega_{\mathbf{k}\pm})$;

$$\omega_{\mathbf{k}\pm} = \sqrt{\alpha_{\mathbf{k}} \pm (\alpha_{\mathbf{k}}^2 - \gamma_{\mathbf{k}}^2)^{1/2}},$$

where $\alpha_{\mathbf{k}} = v_{\mathbf{k}+}^2 + (1/2)(\epsilon_{\mathbf{k}}^2 + \lambda^2)$, $\gamma_{\mathbf{k}}^2 = (\epsilon_{\mathbf{k}}\lambda - v_{\mathbf{k}-}^2)^2 + 4(v_{1\mathbf{k}}v_{2\mathbf{k}}\epsilon_{\mathbf{k}})^2$ and $v_{\mathbf{k}+}^2 = v_{1\mathbf{k}}^2 \pm v_{2\mathbf{k}}^2$.

The mean-field free energy is then

$$\mathcal{F} = -NT \sum_{\mathbf{k},\pm} \log[2\cosh(\beta\omega_{\mathbf{k}\pm}/2)] + N\mathcal{N}_{s} \sum_{\Gamma=1,2} \frac{v_{\Gamma}^{2}}{J_{\Gamma}}.$$

Minimizing \mathcal{F} with respect to λ , ν_1 and Δ_2 leads to three mean-field equations, which are then solved for the temperature when Δ_2 goes to zero, yielding the curve shown in Fig. 2.

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Author information

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