Unconventional Superconductivity from Local Spin Fluctuations in the Kondo Lattice

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(Received 24 January 2013; published 5 April 2013)

The explanation of heavy-fermion superconductivity is a long-standing challenge to theory. It is commonly thought to be connected to nonlocal fluctuations of either spin or charge degrees of freedom and therefore of unconventional type. Here we present results for the Kondo-lattice model, a paradigmatic model to describe heavy-fermion compounds, obtained from dynamical mean-field theory which captures local correlation effects only. Unexpectedly, we find robust s-wave superconductivity in the heavy-fermion state. We argue that this novel type of pairing is tightly connected to the formation of heavy quasiparticle bands and the presence of strong *local* spin fluctuations.

DOI: 10.1103/PhysRevLett.110.146406 PACS numbers: 71.27.+a, 74.20.Mn

Heavy-fermion (HF) $4f$ and $5f$ intermetallic compounds constitute a paradigm for strong electronic correlations. Their low-temperature behavior is affected by f-shell local moments subject to antiferromagnetic (AFM) exchange coupling to the conduction electrons, resulting in Fermi liquid (FL) phases with strongly renormalized Landau parameters, most notably huge effective masses [1–4]. HF materials often display symmetry-breaking phases which occur either within the heavy FL [1] or compete with it [5]. While magnetic order in systems containing unscreened moments appears natural, HF superconductivity is conceptually nontrivial and indeed came as an unexpected discovery more than three decades ago [6]. Now, a wide variety of f-electron superconductors are known [7,8], many of them confirmed to be unconventional [1,2,9].

Superconducting (SC) transitions in HF compounds are often assumed to be driven by nonlocal fluctuations of the f-shell spin degrees of freedom. This idea finds support in the close connection between HF superconductivity and magnetic quantum phase transitions where such fluctuations are strong [3,5,10,11]. Alternatively, pairing mediated by fluctuations in the charge channel (i.e., valence fluctuations) has also been discussed [12]. Given that the basic theoretical models for HF materials, the periodic Anderson model (PAM) and Kondo-lattice model (KLM), constitute complicated interacting many-body problems which cannot be solved exactly, theoretical descriptions of HF superconductivity often employ either simple static mean-field theories or effective models of fermions coupled to spin or charge fluctuations.

A rather successful approach to study the microscopic properties of correlated-electron lattice models beyond static mean-field or effective descriptions is the dynamical mean-field theory (DMFT) together with its cluster extensions [13,14]. The lattice problem is mapped onto a self-consistent quantum impurity model at the expense of losing information on nonlocal correlation effects beyond the spatial size of the impurity cluster. Therefore, it is commonly assumed that a proper description of HF superconductivity within DMFT-based approaches requires either large enough clusters or the inclusion of a bath in the two-particle channel (which explicitly models a bosonic "glue" for superconductivity). In particular, within the conventional (single-site) DMFT only s-wave superconductivity occurs [15] and a relation to magnetic fluctuations appears highly unlikely.

In this Letter we report on the unexpected observation of a stable SC solution to the DMFT equations for the KLM without any external glue. Although the pairing symmetry is s wave, the SC state is highly unconventional: Pairing is driven by *local* spin fluctuations; it comes with a strong frequency dependence of the gap function and requires the formation of HF bands as a prerequisite.

We note that a hint of the possible occurrence of local superconductivity was found in an earlier DMFT study to the PAM [16] which, however, did not analyze the SC phase but only normal-state instabilities. Static mean-field descriptions of the KLM or the PAM can also yield solutions with local pairing [17–19], but it is difficult to assess their validity, as fluctuations beyond mean field may destroy pairing.

Model.—Within the KLM the localized f states are described by a (pseudo)spin degree of freedom which couples to the conduction (c) electrons via an exchange interaction. We use the simplest version of the model, i.e., a nearest-neighbor tight-binding conduction band with spin degeneracy only and a $S = 1/2$ spin located at each lattice site. The Hamiltonian reads

$$\mathcal{H} = -t \sum_{\langle i,j\rangle,\sigma} \hat{c}^\dagger_{i\sigma}\hat{c}_{j\sigma} + \frac{J}{2}\sum_{i,\alpha\beta}\hat{\mathbf{S}}_i \cdot \hat{c}^\dagger_{i\alpha}\boldsymbol{\tau}_{\alpha\beta}\hat{c}_{i\beta}. \quad (1)$$

Here, $\hat{c}_{i\sigma}^{(\dagger)}$ denote annihilation (creation) operators of conduction electrons with spin σ at site i, and $\langle \cdot, \cdot \rangle$ denotes nearest neighbors. $\hat{\mathbf{S}}_i$ is the operator for the localized spin, $\boldsymbol{\tau}$ the vector of Pauli matrices, and the interaction between the conduction electrons and the localized spin is modeled as an isotropic exchange coupling with $J > 0$. For simplicity, we will consider nearest-neighbor hopping t on the infinite-dimensional Bethe lattice, leading to a semicircular density of states with bandwidth W. We have checked that using different lattice types (for example, hypercubic or square lattice tight binding) does not change the results qualitatively.

Methods.—Within standard DMFT, the conduction-electron self-energy is approximated as local in space, $\Sigma(\mathbf{k}, \omega) \to \Sigma(\omega)$. Then, the KLM maps onto an effective single-impurity Kondo model (SIKM) [4] which needs to be solved within a self-consistency loop. In this work we treat the effective SIKM using Wilson's numerical renormalization group [20]. It allows one to access arbitrarily small energy scales, to calculate spectra directly on the real-frequency axis, and to work at both $T = 0$ and $T > 0$. We work with the discretization parameter $\Lambda = 2.0$, keep $N_{st} = 1000, \ldots, 2000$ states, and perform z averaging with $N_z = 2$ [20].

To allow for solutions with SC order, we generalize the DMFT equations and the impurity solver to a Nambu formulation with 2×2 matrix propagators [20,21]. This constrains our calculations to spin-singlet even-frequency s-wave superconductivity. The DMFT treatment of superconductivity is nonperturbative and thus goes beyond the standard Eliashberg theory [22]: it does not rely on any assumption about a separation of energy scales for the fermions and the bosonic glue responsible for the formation of superconductivity. At present, we restrict the calculations to SC order only, suppressing possible magnetic order. Our results below show that strong pairing occurs in a regime without magnetic order, justifying this neglect.

Results: Superconductivity at $T = 0$.—Our numerical solution of the DMFT equations yielded, for a range of model parameters, stable SC solutions whose properties we discuss in the following. The conduction-band density of states (DOS) of both the paramagnetic normal (N) and SC solutions of the KLM for fixed conduction band filling $n = 0.9$ and different J are shown in Fig. 1, left-hand panels. The only feature of the N DOS is a hybridization pseudogap above the Fermi energy, signaling the formation of heavy quasiparticles, which can be rationalized within the picture of hybridized c and f bands. The N solution is unstable against superconductivity, where the DOS exhibits two additional features. (i) A true gap Δ_{SC} with well-developed van Hove singularities is present around the Fermi energy; as a function of J, it first increases up to $J/W = 0.5$, and then slowly decreases. (ii) In addition to the SC coherence peaks, there are side resonances at positions which roughly scale with J. These structures are sharp for smaller J, but become increasingly washed out for larger J. These features are likely related to local spin fluctuations stabilizing the pairing, as discussed in the Supplemental Material [19].

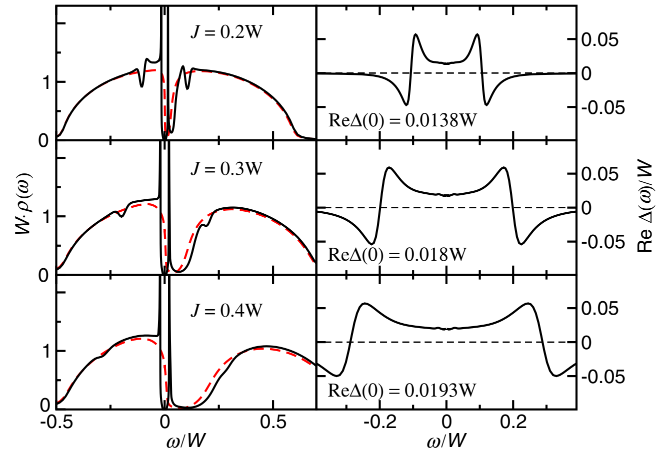

FIG. 1 (color online). Left-hand panel: N DOS (dashed red lines) and SC DOS (full lines) for $n = 0.9$ and various J. Right-hand panel: Real part of the corresponding gap functions.

As the appearance of a gap alone is not sufficient to identify the solution as a SC, one needs to look at the anomalous parts of the Nambu Green function as well as the anomalous part of the self-energy. From it, a SC gap function can be defined, as in standard Eliashberg analysis, via

$$\Delta(\omega) = \frac{\Sigma_1(\omega) + i\Sigma_2(\omega)}{1 - \Sigma_0(\omega)/\omega}, \tag{2}$$

where $\Sigma_\alpha(\omega)$ denote the components of the electronic self-energy expanded into Pauli matrices, $\Sigma = \Sigma_\alpha \tau_\alpha$ ($\alpha = 0, 1, 2, 3$), in Nambu space. The resulting real parts $\mathrm{Re}\Delta(\omega)$ are shown in the right-hand panels of Fig. 1. As expected for even-frequency pairing, $\mathrm{Re}\Delta(\omega)$ is symmetric. It shows a strong frequency dependence, with sharp features shifting to larger energies and broadening with increasing J. These structures are linked to the side resonances in the DOS: the zeros of $\mathrm{Re}\Delta$ coincide with the resonances in the DOS. The $\omega = 0$ limit, $\mathrm{Re}\Delta(0)$, provides an estimate of the gap seen in the DOS; it exhibits the same nonmonotonic behavior with J as noted above for the gap in the DOS.

To characterize the evolution of superconductivity across the phase diagram, we plot in Fig. 2 the anomalous expectation value $\Phi = \langle \hat{c}_{i\uparrow}\hat{c}_{i\downarrow} \rangle$, as a function of J and n. Superconductivity is found to be stable over wide regions of the phase diagram for $J/W > 0.1$. For a fixed $J/W = 0.2$, a finite Φ is found between $0.45 < n < 1$. For larger J/W the SC region extends to even lower fillings. A maximum of Φ appears around $J/W = 0.3$ and $n = 0.9$; the side resonances are also the most pronounced for these parameters.

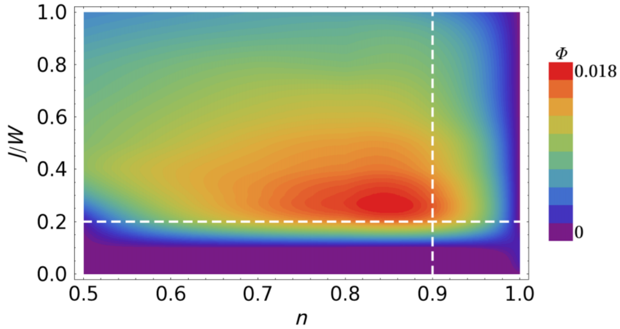

FIG. 2 (color online). Anomalous expectation value Φ as a function of J/W between quarter and half filling. The white dashed lines indicate the cuts along a fixed J and n shown in Figs. 3(a) and 3(b).

In Fig. 3 we display the evolution of Φ along two cuts along the phase diagram indicated by the white dashed lines in Fig. 2. The analysis for weak Kondo coupling, $J/W < 0.1$, is difficult as the signatures of SC become very weak and hard to distinguish from numerical noise. Thus we cannot decide whether the SC solution ceases to exist for small J, or whether it survives down to $J \to 0$ with an (exponentially) small pairing scale. (The latter would be expected in the weak-coupling limit of certain mean-field theories [19].) For $J \gtrsim W/2$, on the other hand, we observe a decay consistent with $\Phi(J) \propto 1/J$. We will comment on this behavior further down.

Normal-state Fermi liquid scale.—In the normal state, the KLM realizes a heavy FL at low temperatures for $n \neq 1$, with a FL (coherence) scale T_0. In a local self-energy approximation, T_0 can be efficiently extracted from the quasiparticle weight,

$$Z^{-1} = 1 - \frac{d\mathrm{Re}\Sigma(\omega)}{d\omega}\bigg|_{\omega=0}, \tag{3}$$

via $T_0 = WZ$, where $\Sigma(\omega) = \Sigma_0(\omega) + \Sigma_3(\omega)$. The evolution of T_0 with J and n is also depicted in Figs. 3(a) and 3(b); we recall that for $J \to 0$ the scale T_0 depends exponentially on both J and the bare c DOS, $T_0 \propto \sqrt{J/W}\exp[-\alpha(n)W/J]$ with a weakly n-dependent coefficient $\alpha(n)$ [23,24]. For large $J \gtrsim W/2$ the dependence of T_0 on J significantly deviates form this Kondo form and rather tends to saturate as $J \to \infty$. Finally, for fixed J and varying n we recover the known dependency $T_0 \propto ne^{cn}$ [24]. These different types of behavior for $T_0(n, J)$ can be seen from the lines superimposed to the data in Fig. 3.

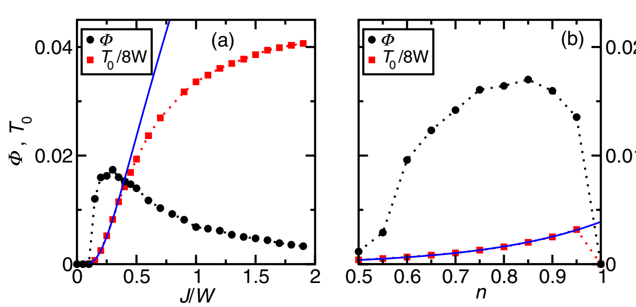

FIG. 3 (color online). Φ (circles) and T_0 (squares) as a function of J at fixed $n = 0.9$ (a), or as a function of n at fixed $J/W = 0.2$ (b). The full lines represent approximate dependencies of T_0 on J and n (see text).

Apparently, there does not exist a simple connection between T_0 and Φ. For small J, $\Phi(n, J)$ seems to scale with T_0. However, as noted before, the results for very small Φ become unreliable for numerical reasons; i.e., one cannot readily extract a simple relation between Φ and T_0 in this limit. For large J at $n = 0.9$, on the other hand, we do not see a direct relation between T_0 and Φ, but find $\Phi \propto 1/J$ instead.

Competition with magnetism.—Within the DMFT for the KLM, one also finds magnetic phases, namely, AFM close to half filling and ferromagnetism (FM) at small filling [25–27]. Note, however, that these phases have limited extent in both J and n; for example, at $n = 0.9$ we find AFM only for $J < J_c(n = 0.9) \approx 0.2W$ [26] and FM only for $n < n_c(J/W = 0.2) \approx 0.65$ [27]. Thus, the region in parameter space where we find a strong superconducting phase in Fig. 2 seems to be complementary to the regions with magnetic phases. As the boundaries seem to overlap—in particular the SC phase at $n = 1$ lies inside the AFM regime—it is surely interesting to study the competition as well as the interplay of AFM, FM, and SC in detail. This is work in progress.

Results for $T > 0$.—Figure 4 displays finite-temperature results for the DOS: With increasing temperature T the gap shrinks and the spectral side resonances are depleted. The reduced pair correlations are also reflected in a decrease of Φ (see inset of Fig. 4). Close to T_c the gap is progressively

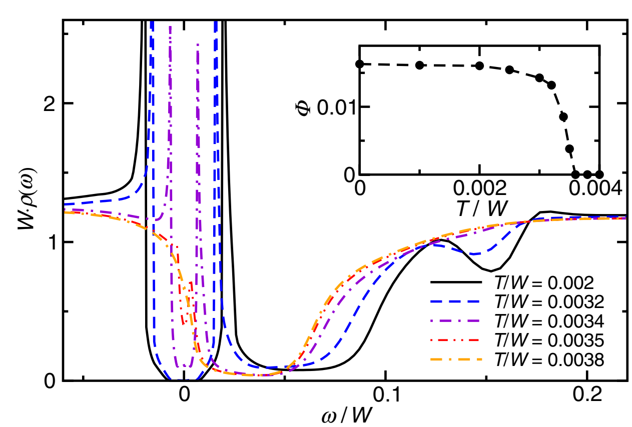

FIG. 4 (color online). Evolution of the DOS with increasing temperature for $n = 0.9$ and $J = 0.25$ W, where $T_0/W = 0.0105$ at $T \to 0$. Inset: Φ as a function of temperature. Pair correlations are fully suppressed for $T > T_c$.

TABLE I. Quantities characterizing the superconducting solution for $n = 0.9$ as a function of J in the region of optimal conditions for pairing.

J/W	0.2	0.25	0.3	0.4	0.5
T_0/W	0.0200	0.0418	0.0658	0.1139	0.1548
T_c/W	0.0027	0.0036	0.0054	0.0058	0.0054
$\Phi(T=0)$	0.0160	0.0163	0.0174	0.0153	0.0140
$\mathrm{Re}\Delta(0)/W$	0.0138	0.0165	0.0180	0.0193	0.0195
$\mathrm{Re}\Delta(0)/T_c$	5.169	4.583	3.321	3.305	3.585

filled, and both the hybridization gap and the side resonances move towards the Fermi level. Finally, in the normal-state solution for $T > T_c$, only the hybridization pseudogap is visible.

Estimating T_c from the numerical data at finite T is hard, as close to the critical temperature the signatures of SC become very small and also the convergence of the DMFT rather slow. From the data in Fig. 4 we extract $T_c \approx 0.0036$ W at $n = 0.9$ and $J = 0.25$ W, which is well below T_0. In Table I we collect the resulting estimates for T_c for fixed $n = 0.9$ and several values of J in the region where according to Fig. 3 we have optimal conditions for pairing. As a rule we observe that always $T_c < T_0$; i.e., the HF state seems to be a necessary ingredient for the appearance of the SC phase. For small J this obviously leads to a strong suppression of T_c. On the other hand, the gap $\mathrm{Re}\Delta(0)$ appears to be much less sensitive to J and T_0 in this regime. An interesting characteristic quantity is the ratio between the gap and T_c. The results are given in the last row of Table I. Obviously, the ratio exceeds the BCS value $\Delta/T_c \approx 1.74$ by a sizable factor between 2 and 3. Such values are actually observed in HF superconductors [11], although the interpretation there is usually given in terms of a weak-coupling theory for a d-wave state.

Pairing mechanism at strong coupling.—In the strong-coupling limit, $J > W$, the pairing mechanism and behavior of $\Phi(J)$ can be understood perturbatively, Fig. 5. We consider a conduction-band filling of $n = N_{\mathrm{el}}/N_s \lesssim 1$ on N_s sites. For $J/W \to \infty$ there are N_{el} Kondo singlets and $(N_s - N_{\mathrm{el}})$ uncompensated local moments. The c electrons are mobile, such that the uncompensated moments can alternatively be interpreted as spinful c holes with density

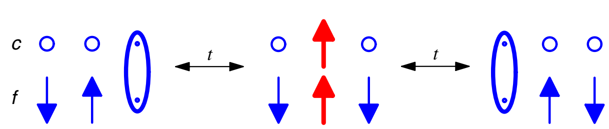

FIG. 5 (color online). Second-order process responsible for pairing in the limit of large J/W. Shown are the c electron and local-moment (f) configurations on adjacent lattice sites; the ellipses denote a singlet bond of two electrons. Here, two hoppings effectively move a down-spin hole by two sites across an up-spin hole, with a triplet intermediate state (bold red arrows).

$(1 - n)$ and a hard-core repulsion, forming the Fermi liquid with a coherence scale $T_0 \propto W$ (whereas the impurity Kondo scale simply diverges $\propto J$).

For large but finite J/W additional excitations out of this manifold are allowed. The lowest one consists of converting a singlet into a triplet and can be created by c (hole) hopping. Interestingly, the excited triplet may decay via a different neighboring hole provided that its spin is opposite to the first one. Together, this second-order virtual process leads to correlated hopping, with an energy gain $\propto W^2/J$, which binds two holes into a singlet state, Fig. 5. The pairing is local—it occurs *on* the site of the virtual triplet—but the holes share this site only in the virtual triplet state, so that the pairing is strongly retarded.

It is plausible that this pairing mechanism continues to operate at smaller J. Importantly, the existence of the virtual state, whose energy may now be approximated by the Kondo binding energy T_0, requires Kondo screening to be intact—this naturally explains the limitation $T_c < T_0$. The picture also makes clear that superconductivity is more favorable close to half filling: Here, Kondo screening is done by c electrons whereas the paired carriers are c holes (for $n < 1$). In contrast, in the opposite (exhaustion) limit of small n, Kondo screening becomes strongly nonlocal, and it is the same c electrons doing the screening that need to be paired; thus, the pairing is weak.

Summary.—We have identified a novel mechanism for superconductivity in heavy-fermion materials: Local spin fluctuations due to the Kondo exchange coupling can act as a retarded paring interaction and drive s-wave superconductivity in the heavy FL. A particularly interesting feature is the appearance of structures in the tunneling DOS at scales related to the spin fluctuation spectrum, i.e., well separated from the coherence peaks at the gap edges. Such structures have been observed, for example, in recent scanning tunneling spectroscopy (STS) measurements on iron pnictides [28]. It would be interesting to extend such STS experiments to systematically study HF superconductors.

For model parameters relevant to HF systems, T_c can be as large as several kelvin, a typical T_c scale for existing HF superconductors. However, given that pairing in our theory is s wave, it can be ruled out for those materials where the existence of gap nodes has been established experimentally. More generally, it is an interesting question to what extent this mechanism can cooperate or will actually compete with SC driven by, e.g., nonlocal magnetic fluctuations. To address this point numerical studies beyond DMFT will be required. Work along these lines is in progress.

This work was supported by the DFG through PR293/13-1 (O. B., T. P.), the BMBF through IND 10/067 (T. P.), as well as FOR 960 and GRK 1621 (M. V). R. Ž. acknowledges the support of ARRS through Program P1-0044. Numerical renormalization group calculations have been done using a modification of the *NRG Ljubljana* package. Computer support was provided by the Jožef Stefan Institute Ljubljana and the Gesellschaft für wissenschaftliche Datenverarbeitung Göttingen (GWDG).

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