Robust *d*-Wave Pairing Correlations in the Heisenberg Kondo Lattice Model

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The Kondo lattice model enlarged by an antiferromagnetic coupling J_{AF} between the localized spins is here investigated using computational techniques. Our results suggest the existence of a *d*-wave superconducting phase close to half-filling mediated by antiferromagnetic fluctuations. This establishes a closer connection between theory and heavy fermion experiments than currently provided by the standard Kondo lattice model with $J_{AF} = 0$.

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Introduction.—Heavy fermions (HF) continue attracting the interest of the condensed matter community [1]. These materials are phenomenologically described by Doniach's scenario, where localized spins interact with conduction electrons via an exchance interaction J [2]. At small J, the indirect Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism is expected to induce an antiferromagnetic (AF) state, whereas for large J a paramagnetic spin-liquid state emerges [2]. The discovery of superconductivity in $CeCu_2Si_2$ [3], and subsequently in several other HF compounds, unveiled the rich variety of phenomena present in these strongly correlated electronic systems. Currently, HF superconductors are widely considered as examples of superconductivity not mediated by lattice vibrations. In fact, in $CeIn_3$, $CePd_2Si_2$, and UPd_2Al_3 there is evidence that the superconducting (SC) pairing is caused by spin fluctuations [4,5].

Several formal theories of HF materials start with the Kondo lattice model (KLM) [6]. The development of powerful many-body numerical techniques, and the continuous growth in computer power, have allowed for non mean-field and free of noncontrolled parameters investigations of the KLM, at least in low dimensional systems. These investigations have revealed two potentially important problems in establishing a connection between KLM and HF phenomenology: (1) Including hole carriers away from half-filling, and at large J, the KLM leads to a robust ferromagnetic state that is not obviously connected with states found experimentally [7]; (2) more importantly, there is evidence that the standard one-dimensional (1D) KLM does *not* present SC tendencies close to half-filling [8]. Although these anomalies may be caused by the low dimensionality of the Kondo lattices studied, they still raise doubts about the full validity of the simple KLM to describe heavy fermions. Moreover, it would be advantageous for theoretical investigations to identify a simple HF model with clear SC tendencies, even in low dimensions. The growing number of SC HF compounds clearly requires a model paradigm involving not only AF and spin-gapped phases, as in the past, but including a SC phase as well.

To address these concerns, and better capture the physics of HF systems, we must move beyond the standard KLM. In this Letter, using computational techniques it is shown that the addition of a direct AF coupling J_{AF} between the localized spins considerably alleviates the problems mentioned above. Previous investigations had already suggested that for UPd_2Al_3 the AF coupling between localized spins is important to understand the SC phase [9]. Moreover, other authors already remarked the importance of J_{AF} when focusing on the magnetism of the HF systems [10–12]. However, our present effort goes beyond these previous studies by revealing the formation of *d*-wave symmetric hole-pairs and the development of robust SC correlations when J_{AF} is incorporated to the two-dimensional KLM. This establishes a better connection theory-experiment in the HF context than provided by the plain KLM.

Model and methods.—The antiferromagnetic Heisenberg Kondo Lattice Model (HKLM) is given by

$$H = -\sum_{\langle i,j\rangle,\sigma}(c^\dagger_{i,\sigma}c_{j,\sigma} + \text{H.c.}) + J\sum_j \mathbf{S}_j\cdot\mathbf{s}_j + J_{AF}\sum_{\langle i,j\rangle}\mathbf{S}_i\cdot\mathbf{S}_j,$$

where $J > 0$ is the Kondo coupling between the conduction electrons and the local moments, J_{AF} is the antiferromagnetic interaction between the localized spins $1/2$, and the hopping amplitude was set to unity to fix the energy scale. The rest of the notation is standard. The HKLM on a $N \times L$ cluster was here investigated using the Lanczos technique [13], and the DMRG method [14] under open boundary conditions (OBC) [15,16].

Note that the same HKLM also describes the manganites if $J < 0$ and the $\mathbf{S}_j$'s are assumed classical [17]. Investigations of manganite models have shown that J_{AF} is crucial for the numerical stabilization of experimentally known phases that otherwise become unstable due to the strong ferromagnetic tendencies [17,18]. Thus, our effort also provides a unifying view of HF and manganite research regarding the relevance of the J_{AF} coupling.

Binding energies.—Our investigations mainly focused on pairing and SC correlations. To observe indications of pairing in the HKLM ground state, we measure the pair-binding energy defined as $\Delta_B = E(0) + E(2) - 2E(1)$, where $E(l)$ is the ground state energy in the subspace with $(NL - l)$ conduction electrons. If the holes do not form a bound state, in a *finite system* the binding energy is positive $\Delta_B > 0$ [13], while in the thermodynamic limit it vanishes. On the other hand, if holes do form a bound state then $\Delta_B < 0$, and this is indicative that attractive effective forces are present, as widely discussed before in the context of high-T_c investigations [13].

In Fig. 1, we show the region in the $J - J_{AF}$ plane where Δ_B is less than -0.1, for the cluster sizes 1×10, 2×6 and for a square lattice 3×3 with OBC. We choose to present the region where $\Delta_B < -0.1$, as opposed to $\Delta_B = 0$, since previous experience in the cuprate context [13] suggests that this procedure takes better into account the size effects. The limitation of using small clusters originates in the huge Hilbert space of the HKLM, with 8 states per site, and in the need to calculate hundreds of points to extract comprehensive phase diagrams. However, for some selected couplings much larger clusters were considered and a thermodynamic limit extrapolation was carried out (see Fig. 2 and discussion below). Note that the binding regions in Fig. 1 are qualitatively similar, although the clusters have different shapes. Thus, the formation of hole pairs is a robust effect, suggesting that pair formation might also appear in the three-dimensional phase diagram (currently unreachable numerically). Moreover, the tendency toward increasing pairing strength found in Fig. 1 moving from chains to ladders or square clusters is similar to tendencies found in the $t - J$ model for cuprates [13]. It is interesting also to note that a previous study by Sikkeman *et al.* [11] have found that J_{AF} induces a spin gap phase with no charge gap close to half-filling for the one-dimensional HKLM, in agreement with the phase diagram present in Fig. 1. Finally, note that values of J_{AF} are expected to be small due to the small overlap between nearest-neighbor $4f/5f$ orbitals, and in Fig. 1(c) it appears that hole binding can occur for J_{AF} as small as 0.2. Only a careful two-dimensional size scaling, beyond our capabilities at present, can answer how small J_{AF} can be to still induce hole pairing.

In Figs. 2(a)–2(c), Δ_B vs $1/L$ is shown for the chain, two-leg ladder and square clusters, respectively. There are clearly two distinct behaviors. For the couplings that present hole binding for the small clusters presented in Figs. 1, Δ_B tends to negative values when $L \to \infty$, while for the other parameters $\Delta_B \to 0$. Regarding the 6×6 cluster, even working with up to $m = 4000$ states the energies did not converge well. However, as presented in Fig. 2(d), Δ_B tends to a negative value for the intermediate coupling range, similar as the results obtained for the other cluster shapes. All these results indicate that close to half-filling, pairing tendencies exist for intermediate values of J and J_{AF} in the bulk of the HKLM. It is also interesting to observe the behavior of Δ_B vs J for a fixed J_{AF} (or fixing J and varying J_{AF}). As present in Fig. 3(a), $-\Delta_B$ reaches a maximum value at intermediate J, important detail to compare with experiments, as discussed later. We found that the hole binding is robust in the same region where the nearest-neighbor spin-spin correlations are robust, as shown in the inset of Fig. 3(a). There, we also plot the nearest-neighbor hole-hole correlation [19]. The fact that spin and hole correlations behave similarly supports our claim that antiferromagnetism and hole binding are related. At very large J, both quantities are suppressed together. This suggests that the origin of the hole-pair attraction is connected with AF fluctuations. We believe mechanisms similar to those identified in cuprates [13], such as hole

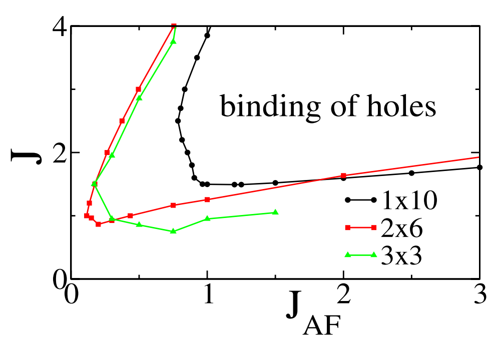

FIG. 1 (color online). The region with binding of holes in the $J - J_{AF}$ plane, for different cluster shapes with OBC. The solid line corresponds to $\Delta_B = -0.1$.

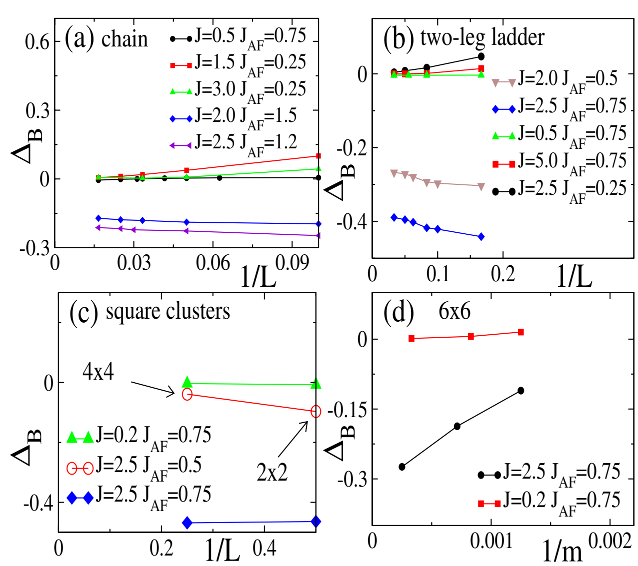

FIG. 2 (color online). Pair-binding energies Δ_B vs $1/L$ for a chain (a), a two-leg ladder (b), and square clusters (c). (d) Δ_B vs $1/m$ for the cluster 6×6, at couplings indicated.

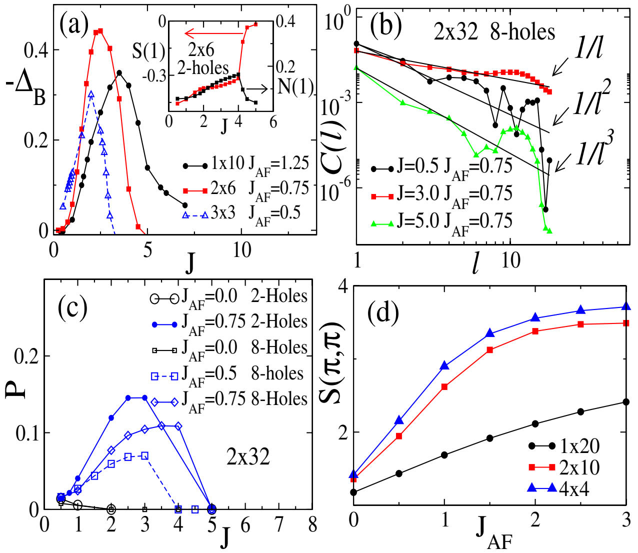

FIG. 3 (color online). (a) $-\Delta_B$ vs J for the clusters 1×10, 2×6, and 3×3 with OBC. Inset: The nearest-neighbor spin-spin ($S(1)$) and hole-hole ($N(1)$) correlations, measured in the center of the 2×6 cluster with 2-holes and $J_{\mathrm{AF}} = 0.75$, as function of J. (b) The pair-pair correlation function $C(l)$ vs l for a system size 2×32 and $J_{\mathrm{AF}} = 0.75$ with 8 holes, for some values of J. The straight lines are data fits with the powers indicated by the arrows. (c) P vs J for the cluster 2×32 with 2 and 8 holes and some values of J_{AF}. (d) The spin structure factor $S(\pi, \pi)$ vs J_{AF} for the clusters 1×20, 2×10 and 4×4, at $J = 2$ and half-filling.

attraction to minimize the "damage" to the AF background, are in action in the HKLM as well.

Pair symmetry.—Our exact diagonalization results for the clusters 2×2 and 3×3 show that the ground state with 0 (2) holes has s-wave (d-wave) symmetry under rotations. Thus, these ground states are connected via a pair-creation operator with d-wave symmetry, similarly as observed in the Hubbard and $t - J$ models [13]. Then, the HKLM predicts d-wave superconductivity, compatible with previous results for HF systems [5].

Pairing correlations.—To confirm that hole-pair tendencies are concomitant with robust superconducting correlations, for the two-leg ladder we measured the rung-rung pair correlation functions $C(l) = \langle \Delta_{i+l} \Delta_i^\dagger \rangle$ where the pair operator is defined as $\Delta_l^\dagger = c_{l1,\uparrow}^\dagger c_{l2,\downarrow} - c_{l1,\downarrow}^\dagger c_{l2,\uparrow}$, and $c_{l\lambda,\sigma}$ annihilates a conduction electron on rung l and leg $\lambda = 1$, 2 with spins $\sigma = \uparrow, \downarrow$ [20]. In Fig. 3(b), $C(l)$ is shown vs distance l, for a fixed J_{AF} and some values of J, using a cluster 2×32 with 8 holes. Similar results are obtained with 2 holes. In order to obtain the *slope* of $C(l)$ we fit our data with the function a/L^n (n an integer). Clearly from this figure, the pair-pair correlations $C(l)$ are enhanced at large distances for intermediate coupling values, i.e., $J = 3$ and $J_{\mathrm{AF}} = 0.75$, with a robust power-law decay $\sim 1/l$. It is interesting to note that the well known two-leg $t - J$ model also has the same slope close to half-filling [21,22]. The origin of this effect may be due to the fact that the HKLM can be mapped, in a limiting case, into the $t - J$ model [11]. Similar SC tendencies are also observed in Fig. 3(c), where $P = \sum_{l=5}^{18} C(l)$ is shown vs J, at a fixed J_{AF}. P is enhanced *only* for nonzero values of J_{AF}, indicating that superconductivity appears only when this interaction is active. Although there is no true long-range order in quasi-1D systems, our results provide strong evidence that superconductivity dominates at intermediate J and J_{AF} in the HKLM close to half-filling. Similar conclusions were reached for the $t - J$ model on chains and ladders [13,21,22].

Magnetic properties.—We also investigated the effect of J_{AF} in the magnetic properties of the HKLM. At small values of J, due to the the RKKY interaction, antiferromagnetic long-range order is expected, whereas for large J a paramagnetic state must emerge. The competition between these two states leads to a quantum critical point at $J_c \sim 1.45$ for the two-dimensional KLM at half-filling [2,8,23,24]. If the AF coupling between the localized spins is added, we favor antiferromagnetism even more. For this reason, J_c is expected to increase with J_{AF}. Here, we do not attempt to provide the location of $J_c(J_{\mathrm{AF}})$ for the two-dimensional HKLM, which is a formidable task, but only show the dominant tendencies in the problem. In Fig. 3(d), we present the intensity of the spin structure factor $S(\vec{q})$ at $\vec{q} = (\pi, \pi)$ vs J_{AF}, for several clusters at $J = 2.0$ and half-filling Clearly from this figure, $S(\pi, \pi)$ increases with J_{AF}, suggesting that J_c will increase as well, as anticipated. As in the two-leg ladder [25], we also have found no evidence of ferromagnetism close to half-filling for the 4×4 cluster. Note also that it is expected that the AF-phase will survive away from half-filling, for the three-dimensional KLM [7]. We believe this phase will also exist in the two-dimensional KLM close to half-filling.

Discussion.—Based on the numerical results described in this investigation, in Fig. 4(a) a qualitative phase diagram for the HKLM close to half-filling is presented. The robustness of our results with respect to the shape of the

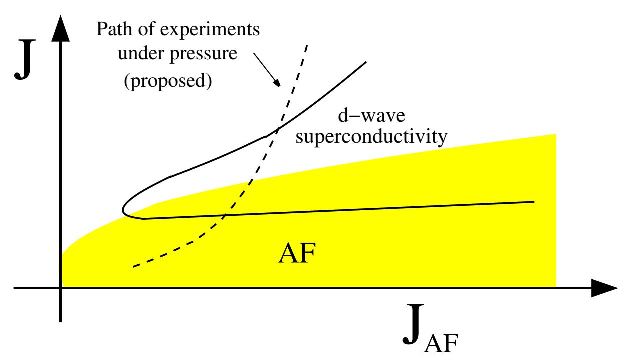

FIG. 4 (color online). Schematic phase diagram of the HKLM close to half-filling. The solid line defines the region where d-wave SC should exist based on our numerical calculations. The yellow region is the long-range AF phase. The dashed line describes a trajectory with increasing pressure that is compatible with HF experiments (see text).

cluster used, suggest our conclusions may be qualitatively valid even in higher dimensions. The only mild assumption in Fig. 4 is the existence of an overlap between the AF and d-wave superconducting regions. At a fixed J and with increasing J_{AF} this overlap is to be expected. In the SC phase not overlapping with AF, strong short-range AF fluctuations are also to be expected.

The phase diagram in Fig. 4 is in qualitative agreement with experimental results reported for some HF materials, such as $CeIn_3$ and $CePd_2Si_2$, at low temperatures. A possible experimental trajectory is shown as a dashed line in Fig. 4. At ambient pressure these compounds are known to be AF [4]. As observed in the figure, for small values of J and J_{AF} (corresponding to low pressures in the experiments) there is no binding of holes (see Fig. 1) and superconductivity is not expected. However, with increasing pressure at a critical value p_{c_1}, and within a narrow pressure range, superconductivity develops and coexists with long-range AF order [4]. This is compatible with our results, since at intermediate values of J and J_{AF} (intermediate pressures) a tendency to d-wave superconductivity was numerically observed. At pressures even higher $p_{c_2} > p_{c_1}$, the real HF system first stabilizes a SC state without AF long-range order, and then finally a transition to a non-SC paramagnetic phase is reached. Again, from the theory perspective this is reasonable since for even larger values of J and J_{AF} (larger pressures) the system eventually transitions into a phase without hole binding (see Figs. 1 and 2). Note that the positive slope of the proposed "path" in Fig. 4 is qualitatively correct since under pressure both J and J_{AF} increase, due to the increasing overlaps of wave functions. Note also that theoretically the maximum binding energy is reached at intermediate couplings in the dashed line path, suggesting that the SC critical temperature first increases under pressure, reaches a maximum, and then decreases, as in experiments [4,26].

Summarizing, we have investigated the HKLM with emphasis on hole-pair formation and SC correlations. A region with a robust tendency toward a SC state was identified at intermediate values of J and J_{AF} close to half-filling. The binding of holes only appears when the AF interaction between the localized spins is considered, showing its critical importance for a proper theoretical description of heavy fermion materials.

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