Inverse indirect magnetic exchange

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Magnetic moments strongly coupled to the spins of conduction electrons in a nanostructure can confine the conduction-electron motion due to scattering at almost localized Kondo singlets. We show that this may lead to local-moment formation in the conduction-electron system and to a novel magnetic exchange coupling mediated by the Kondo singlets. Its distance dependence is oscillatory and induces robust ferro- or antiferromagnetic order in multi-impurity systems.

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Introduction. The appearance of magnetic order in condensed-matter systems requires (i) the existence of the formation of local magnetic moments, (ii) a coupling mechanism favoring a certain alignment of the moments, e.g. ferro- or antiferromagnetically, and (iii) the stability of long-range magnetic order against different types of thermal or quantum fluctuations and against competing ordering phenomena as superconductivity, charge or orbital order.

Local-moment formation typically results from incompletely filled localized orbitals or from strong local correlations and can be described by Hubbard-, Anderson- or Kondo-type models. Among the different known coupling mechanisms, such as the direct Heisenberg exchange or the indirect Anderson super exchange, the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction provides a mechanism for longer-ranged coupling between magnetic impurities in a D-dimensional metallic system which can be either ferro-or antiferromagnetic, depending on the distance. It originates from a local exchange coupling \( J \) which, for weak \( J \), mediates an indirect interaction \( J_{\text{RKKY}} \propto J^2 \).

The RKKY exchange has gained much interest recently in the context of nanostructures, e.g. in double-dot semiconductor quantum devices with tunable RKKY-mediated control on spin degrees of freedom. Nanostructures with tailored magnetic properties can be engineered using scanning-tunnelling techniques by positioning magnetic atoms on non-magnetic metallic surfaces at certain distances where RKKY is ferro- or antiferromagnetic. Furthermore, the Kondo effect, i.e., the screening of the local magnetic moments due to non-local antiferromagnetic correlations induced by \( J \), competes with RKKY-mediated magnetism below a characteristic temperature scale \( T_K \). In nanosystems with strong electron-confinement effects, a subtle Kondo-vs-RKKY competition takes place at weak \( J \).

Here, we propose a novel exchange mechanism where the roles of conduction electrons and impurities are “inverted”. We show that the Kondo effect helps to form local moments, (ii) to couple the moments and (iii) leads to magnetic order in certain nano-structured geometries: For strong \( J \), almost local Kondo singlets are formed which act as hard scattering centers for the itinerant conduction electrons and may confine their motion, depending on the impurity positions. In certain geometries, this tends to localize the conduction electrons and leads to the formation of local magnetic moments in the a priori uncorrelated conduction-electron system. These moments are found to couple magnetically via virtual excitations of the Kondo singlets.

We study the resulting “inverse indirect magnetic exchange” (IME) by means of strong-coupling perturbation theory and different numerical techniques: The IME shows an oscillatory distance dependence. For extended systems, it triggers long-range magnetic order that smoothly evolves from standard RKKY-induced magnetism with increasing \( J \). The IME-induced ferromagnetic ground state in a one- and in a two-dimensional diluted Kondo lattice is studied in detail. It is found to be robust against charge fluctuations on the impurities but sensitively depends on the quantum confinement of the conduction electrons, e.g., on the geometry of magnetic adatoms in an experimental setup using scanning-tunnelling techniques. The proposed IME mechanism is an ideal tool for tailoring magnetism in nanostructures.

From RKKY to inverse exchange. We consider a system with \( R \) spins \( S_r \), with spin-quantum numbers \( 1/2 \), which are coupled locally via an antiferromagnetic exchange \( J > 0 \) to the local spins \( s_i \) of a system of \( N \) itinerant and non-interacting conduction electrons. The conduction electrons hop with amplitude \( t \equiv 1 \) between non-degenerate orbitals on neighboring sites of a D-dimensional lattice of \( L \) sites:

\[
\mathcal{H} = -t \sum_{\langle \langle i,j \rangle \rangle, \sigma} c_{i\sigma}^\dagger c_{j\sigma} + J \sum_{r=1}^R s_i, S_r .
\]  

Here, \( c_{i\sigma} \) annihilates an electron at site \( i = 1, \ldots, L \) with spin projection \( \sigma = \uparrow, \downarrow \), and \( s_i = \frac{1}{2} \sum_{\sigma, \sigma'} c_{i, \sigma}^\dagger \sigma_{\sigma, \sigma'} c_{i, \sigma'} \) is the local conduction-electron spin at \( i \), where \( \sigma \) is the vector of Pauli matrices. Impurity spins couple to the local conduction-electron spins at the sites \( \iota_r \). We investigate the half-filled system with \( N = L \) electrons.

To illustrate the crossover from conventional RKKY indirect magnetic exchange at weak \( J \) to the inverse indirect exchange at strong \( J \), we first analyze a simple model with
a small number of $L = 8$ sites and $R = 2$ spins at $i_1 = 3$ and $i_2 = 5$ using exact diagonalization (see Fig. 1). In the RKKY regime for $J \to 0$, the low-energy sector of $\mathcal{H}$ is exactly described by an effective RKKY two-spin model $H_{\text{RKKY}} = -J_{12} S_i S_j$ with $J_{12} \propto (-1)^{|i_1-i_2|}J^2/|i_1-i_2|$. For a “ferromagnetic distance” $i_1-i_2 = 2$ the two impurity spins form a non-local triplet in the ground state.

As is seen in Fig. 1, the ground state is unique (apart from the spin degeneracy) for any $J \neq 0$, $J \neq \infty$. The absence of a ground-state level crossing at half-filling and for a bipartite lattice is in fact enforced by analytical results [20, 21] based on Lieb’s concept of reflection positivity in spin space [22]. Consequently, the ground-state results [20, 21] based on Lieb’s concept of reflection positivity in spin space [22]. Therefore, the ground state must be a triplet. The numerical analysis of spin-correlation functions shows that this triplet and dominates for $J \to 0$.

For $J \to 0$, standard RKKY theory would predict $S_{\text{tot},0} = S_{\text{max}} = R/2$. In the present case, exactly one of the impurity spins, however, is Kondo screened by the single electron occupying the spin-degenerate one-particle energy level at the Fermi wave vector of the non-interacting conduction-band system (see Ref. [19]). This results in $S_{\text{tot},0} = (R-1)/2$. For all (finite but large) systems studied here, the ground state turns out to be a smooth function of $J$. This explains that $S_{\text{tot},0} = (R-1)/2$ must be the same in both limits. For $J \to 0$, this large spin must then result from a ferromagnetic coupling of local magnetic moments at the sites $i = 2, 4, \ldots, L - 1$ (A-sites, see Fig. 2, inset) which are formed as a result of the increasing confinement of electrons due to the formation of local Kondo singlets at the B sites. The DMRG calculations indeed yield strong antiferromagnetic local spin correlations $\langle S_i S_r \rangle \to -3/4$, vanishing RKKY correlations $\langle S_i S_r \rangle \to 0$, and local-moment formation $\langle s_i^z \rangle \to 3/4$ at A-sites for $J \to \infty$.

Fig. 2 shows the ordered magnetic moments at the central impurity $m_{\text{imp}} \equiv \langle S_{r,z} \rangle$, at the “sub-impurity” B site $m_{\text{B}} \equiv \langle n_{i_1^+} - n_{i_1^-} \rangle$ and a neighboring A-site $m_A \equiv \langle n_{i_1^+} - n_{i_2^-} \rangle$, as obtained from the ground state with maximum $M_{\text{tot}} = S_{\text{tot},0}$. With increasing $J$, there is a clear crossover from the RKKY regime, with $m_{\text{imp}} \to 1$, $m_A$, $m_B \to 0$, to the IIME regime for $J \to \infty$, where the magnetization of the system results from ordered moments at A-sites. The results are characteristic for the infinite system as is obvious by comparing results for $L = 49$ and $L = 89$ (see $J = 5$ in Fig. 2).

The ferromagnetic state is metallic in the entire J range: A vanishing charge gap is found by extrapolation to the $L \to \infty$ limit. This is contrary to a “dense” Kondo lattice (with $R = L$) at half-filling which is a spin-singlet Kondo insulator for any $J$, see Refs. [26, 27].

**Low-energy model.** To analyze the mechanism generating a ferromagnetic coupling between magnetic moments at next-nearest neighboring A-sites $i$ and $j$, we treat the hopping term $\propto t$ in Eq. 1 perturbatively. The starting point is the highly degenerate ground state of the $\mathcal{H} = 0$ model consisting of local Kondo singlets and an arbitrary electron configuration. A non-trivial effective...
FIG. 2: (Color online) Crossover from the RKKY regime at weak coupling to the IIME regime at strong coupling. Calculated ordered magnetic moments on different sites of a tightly correlated chain with spin-1/2 Kondo impurities as functions of $J$ (solid lines, filled symbols) and with Anderson impurities as functions of $8V^2/U$ at Hubbard $U = 8$ (dashed, open) – and of the effective model as well. The effective model capturing the low-energy sector of $H$ in the limit $0 < t \ll J$ is obtained at fourth order in $t$ processes where e.g. an electron hops from $i \in A$ via the neighboring B site to $j \in A$ and, again via B, back to $i$. Here, the local Kondo singlet at B must be excited at an energy cost $\propto J$ first and restored again on the way back. Calculations are lengthy but straightforward and will be published elsewhere [28]. For $J > 0$ and keeping terms up to $O(t^4/J^3)$ we find:

$$H_{\text{eff}}/\alpha = -\sum_{i<j \in A} (s_i s_j - t_i t_j) + \sum_{i \in A} (n_i \uparrow - \frac{1}{2}) (n_i \downarrow - \frac{1}{2}) - \frac{1}{2} \sum_{i < j \in A} \sigma \cdot (e_i^\dagger \sigma e_j + \text{H.c.})(1 - n_{i-\sigma} - n_{j-\sigma}) \ . (2)$$

This effective model describes spin and charge degrees of freedom on the A sites only and is governed by a single energy scale $\alpha \equiv 64t^4/3J^3$. The first term represents a Heisenberg-type ferromagnetic spin interaction and indeed explains the ferromagnetic IIME through a local Kondo singlet. Ferromagnetism due to the IIME competes with formation of a charge-density wave or $\eta$ pairing [20] as favored by the second term. This includes the local isospin $t_i = \frac{1}{2} (c_i^\dagger \uparrow (-1)^i c_{i\downarrow}) \cdot \sigma \cdot (c_i^\dagger \downarrow (-1)^i c_{i\uparrow})^T$. Note that the total isospin $T_{\text{tot}} = \sum_i t_i$ and the total spin $S_{\text{tot}}$ are the generators of the SO(4) symmetry group of the half-filled Kondo model on the bipartite lattice [26] – and of the effective model as well. The effective isospin interaction is “antiferromagnetic”. Analogous to the Mermin-Wagner theorem [29], and opposed to the ferromagnetic spin order, antiferromagnetic (staggered) isospin order would be suppressed by quantum fluctuations of the order parameter for $D = 1$. The necessary formation of local isospin moments in the ground state is suppressed anyway by the repulsive Hubbard term (third term in Eq. (2)). On the contrary, the Hubbard interaction favors formation of local magnetic moments. Finally, there is a correlated hopping term in $H_{\text{eff}}$ which, however, is only active between a spin at $i$ and an isospin at $j$ or vice versa. Exact diagonalization of $H_{\text{eff}}$ for systems with a few A-sites is easily done and in fact yields a ferromagnetic ground state with $S_{\text{tot},0} = (R - 1)/2$ while $(t_i^\dagger i) = 0$.

Charge fluctuations. The IIME mechanism is robust against charge fluctuations on the impurities. This is demonstrated by DMRG calculations where the spin-1/2 Kondo impurities are replaced by Anderson impurities, i.e. the coupling term in the Hamiltonian, Eq. (1), is replaced by $U \sum_{i=1}^n (n_i \uparrow - 1/2) (n_i \downarrow - 1/2) + V \sum_{r \sigma} d_{r\sigma}^\dagger c_{i,\sigma} + \text{H.c.}$. Here, $d_{r\sigma}$ creates an electron at the $r$-th impurity site, namely a correlated site with Hubbard interaction $U$ coupled to the conduction electrons by a hybridization $V$. The weak-$V$ limit is the Kondo limit of the resulting periodic Anderson model with diluted impurities at “ferromagnetic distances”. Indeed, as is seen in Fig. 2 for $8V^2/U = J \ll t$ and as it is prescribed by the Schrieffer-Wolff transformation [30], the results for the two models (filled and open symbols) agree. Deviations are seen beyond the Kondo limit and grow with increasing $V$. However, we again find a crossover from RKKY-coupled magnetic moments at the impurity sites for weak $V$ to IIME-coupled moments formed at the A-sites for strong $V$, producing a ferromagnetic ground state in both limits. For Kondo impurities the crossover roughly takes place between $J/t = 2$ and $J/t = 4$ while in the Anderson case it is located around $V/t = 2$ for the respective $D = 1$ models. In the Anderson case for strong $V$, confinement of A-site electrons is due to the formation of local and strongly bound states at the B sites.

**Dynamical mean-field theory.** IIME-induced ferromagnetism is accessible to a mean-field description. The solid and dashed lines in Fig. 2 show the results of DMFT calculations for Kondo and Anderson impurities, respectively. We employ a standard implementation [25] based on the Lanczos algorithm as an impurity solver [31]. In case of the diluted Kondo-lattice model, Eq. (1), the effective self-consistently determined impurity problem consists of the local spin $S_r$, the corresponding B site and up to 8 bath sites [32, 33]. Up to 9 bath sites are used in the Anderson case. An almost perfect agreement with the DMRG data is found, see Fig. 2.

Let us discuss this finding by referring to the extreme limits: For weak hybridization $V$, the non-local correlations due to the RKKY interaction in the metastable paramagnetic state are not accessible to DMFT [25].
DMFT is able, however, to describe the symmetry-broken ferromagnetic state which is weakly correlated for $V \to 0$. For strong $V$, the essential physics is dominated by local correlations. While the effective spin and isospin interactions in $H_{\text{ eff}}$ are non-local, they take place between degrees of freedom at the $a$ priori uncorrelated A-sites and are therefore expected to have a small feedback on the electron self-energy which can be non-zero at the impurity sites only. This results in a weakly $k$-independent, i.e., almost local self-energy accessible to DMFT.

Higher dimensions. Metallic ferromagnetism caused by the IIME is not restricted to one-dimensional models and, as the agreement with DMRG for the symmetry-broken state in $D = 1$ has shown, can be described quantitatively within DMFT. Nanostructures of magnetic atoms on a $D = 2$ metallic surface layer (isolated from the substrate by a spacer) can be created to confine, with the help of the Kondo effect, conduction electrons in certain geometries.

Fig. 3 displays an example for a $D = 2$ layer with several magnetic impurities. Calculations are performed using the real-space generalization of DMFT [34] for the corresponding diluted Kondo lattice model in the symmetry-broken ground state. Electrons in the chain of A-sites 1, 2, ..., 8 with relative distance $\delta = 2$ are confined. Their moments in fact order ferromagnetically. A-site 9 resides at a distance $\delta = 4$ and is coupled ferromagnetically while the local moments at sites 10 and 11 (with $\delta = 3$ and $\delta = 5$) couple antiferromagnetically to the chain. Note that a tiny Weiss field produced by the chain is sufficient to almost fully polarize the isolated moment if this is weakly coupled to the rest of the system. $|m_z|$ is in fact found to slightly increase with increasing distance from the chain. We conclude that the IIME is oscillatory and decreasing with distance – similar to the RKKY case. Opposed to RKKY, however, the distance dependence of the IIME through a set of $d$ Kondo singlets is expected to be decreasing exponentially with $d$.

There are more characteristic features of the IIME (see Fig. 3): Neighboring A-sites with higher effective coordination mutually support magnetic polarization. This explains the slightly enhanced $m_A$ at and around A-site 12. Confinement of an odd number of electrons is important: There is almost no moment at sites 13 and 14 while the structure 15, 16, 17 is polarized. Confinement as such (with respect to all dimensions) is essential: Electrons at 18 or 19 are not confined, no local moments are formed and thus no polarization found. There is a proximity effect, however, as can be seen at 20, 21, or 22. Furthermore, spin-dependent multiple scattering of conduction electrons at the magnetic structures causes an interference pattern, see sites 23, 24, for example. The real-space DMFT is expected to give quantitatively reliable results: Summing up the local magnetic moments for the 396 uncorrelated sites and the 57 impurities, we find a ground-state spin moment of $m_{\text{tot}} = 2S_{\text{tot},z} = 15.13$ which is, within numerical uncertainties, equal to the exact value $m_{\text{tot}} = 36 - 21 = 15$ which can be obtained analytically [20, 21, 22] by counting the number of impurities on B (filled) and on A sites (open dots).

Conclusions. We have proposed an indirect magnetic exchange mechanism where confinement of conduction electrons due to scattering at Kondo singlets leads to local-moment formation at $a$ priori uncorrelated sites and to spin and isospin couplings via virtual excitations of the Kondo singlets. This IIME is “inverse” to the conventional RKKY coupling. Its oscillatory distance dependence can be utilized to construct nanostructures with tailored magnetic properties, e.g., by placing magnetic atoms in certain geometries on a metallic layer, similar to RKKY-based artificial structures [13–15]. Alternatively, systems of ultracold fermionic atoms trapped in optical lattices [35, 36] may realize multi-impurity Kondo systems in the strong-coupling regime essential to the IIME. Likewise, the strong-coupling limit is reached for only moderate coupling strengths with impurities at larger distances. Future theoretical work may explore systems with correlated conduction electrons and preformed local moments, IIME in ferromagnetic ($J < 0$) multi-impurity Kondo models and in spin-only, e.g., Kondo necklace models. DMFT and DMRG studies of filling dependencies appear particularly exciting. Temperature dependencies are accessible to quantum Monte-Carlo techniques [27] on bipartite lattices at half-filling.

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