Magnetic Order in Kondo-Lattice Systems due to Electron-Electron Interactions

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Abstract. The hyperfine interaction between the electron spin and the nuclear spins is one of the main sources of decoherence for spin qubits when the nuclear spins are disordered. An ordering of the latter largely suppresses this source of decoherence. Here we show that such an ordering can occur through a thermodynamic phase transition in two-dimensional (2D) Kondo-lattice type systems. We specifically focus on nuclear spins embedded in a 2D electron gas. The nuclear spins interact with each other through the RKKY interaction, which is carried by the electron gas. We show that a nuclear magnetic order at finite temperature relies on the anomalous behavior of the 2D static electron spin susceptibility due to electron-electron interactions. This provides a connection between low-dimensional magnetism and non-analyticities in interacting 2D electron systems. We discuss the conditions for nuclear magnetism, and show that the associated Curie temperature increases with the electron-electron interactions and may reach up into the millikelvin regime. The further reduction of dimensionality to one dimension is shortly discussed.

Keywords: nuclear magnetism; magnetic susceptibility; RKKY interaction; Kondo-lattice; two-dimensional electron gas

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INTRODUCTION

A major source of decoherence of electron spin qubits is the hyperfine coupling of the electron spin with the surrounding disordered nuclear spins [1]. If we want to control and eventually eliminate this source of decoherence, it is essential to fully understand the behavior of both the electron spin and the ensemble of nuclear spins. In this text we discuss such a fundamental aspect. We address the question whether the nuclear spins can achieve order through a (ferro)magnetic phase transition. If they do the decoherence source of the hyperfine interaction is massively reduced [2]. We focus specifically on GaAs-based semiconductor heterostructures confining two-dimensional electron gases (2DEGs). Such systems serve as the parent system for the single electron quantum dots defining the spin qubits [3]. Yet we emphasize that the described physics remains valid for general Kondo-lattice systems with interacting electrons. It turns out that the anomalous properties of the 2DEG, resulting from electron interactions, are crucial for the nuclear magnetic order. The exposure to follow is an overview of our recent work published in [4,5,6].

Even though we shall be concerned mainly with fully translationally invariant 2DEG, let us introduce the problem by considering a single electron spin, confined to a quantum dot. The important interaction discussed here is the hyperfine coupling between the electron spin on the dot, $S = (S^x, S^y, S^z)$, and the surrounding lattice of nuclear spins $I_i = (I^x_i, I^y_i, I^z_i)$ ($i$ is the index for the lattice site at position $r_i$). The interaction Hamiltonian can be written in the form

$$H_{hyp}^{dot} = \sum_i A_i S \cdot I_i = \sum_i A_i [S^x I_i^x + S^y I_i^y + S^z I_i^z],$$

with $S^\pm = S^x \pm i S^y$ and $I_i^\pm = I_i^x \pm i I_i^y$, and where $A_i \approx A |\psi(r_i)|^2$ with $A$ a proportionality constant and $\psi(r_i)$ the wavefunction of the confined electron on the quantum dot. The number of nuclear spins is large, typically of order $10^5$, and so the nuclear spins generally act as a disordered bath on the electron spin. While $I_i^\pm$ has a role similar to an external magnetic field, the last two “flip-flop” terms in Eq. (1) flip the electron spin and lead to the decoherence.

This decoherence source can be largely suppressed when the nuclear spins order, ferromagnetically or differently, which effectively suppresses the flip-flop terms in the Hamiltonian [2]. There are two ways of polarizing the system, which we shall call the extrinsic (or dynamic) and the intrinsic (or thermodynamic) polarization of the nuclear spin system. The extrinsic polarization consists in an active manipulation of the nuclear spins by the experimentalist. Several methods to do this have been proposed and partially experimentally realized: The development of quantum control techniques that effectively lessen or even suppress the nuclear spin coupling to the electron spin [7,8]; the narrowing of the nuclear spin distribution [2,9,10]; or the dynamical polarization of the nuclear spins [11,12,13]. Yet in order to extend the spin decay time by one order of magnitude through
polarization of the nuclear spins, a polarization of above 99% is required \[2\], quite far from the best result reached to date in quantum dots, which is about 60% \[13\].

It is possible, however, that a full polarization is achieved intrinsically as well, i.e. through a thermodynamic phase transition to, for instance, a ferromagnetic state. This is our main topic here. In what follows we give a qualitative, physical account to this possibility by introducing step by step the model, the necessary conditions, and the results. For details we refer to \[4\, 5\]. We first indicate how to obtain from a microscopic model an effective Hamiltonian for the nuclear spins only. We show that the conditions of the Mermin-Wagner theorem are not met so that long range order is not forbidden in the 2D system. With a simple mean field theory we can then see under which conditions ferromagnetic order is possible and allows us to identify the first important temperature scale \(T_{MF}\). A refinement leads to the second important temperature scale \(T^*\), which depends on the electron-electron interactions in the 2DEG. The calculation of the susceptibility for interacting electrons allows to estimate \(T^*\) and on the stability of the magnetic order. It turns out that nonanalytic corrections to the electron spin susceptibility are crucial. We discuss the possible forms of these corrections, allowing us to conclude with some numerical estimates for a possible nuclear order.

**MODEL AND EFFECTIVE MODEL**

In order to discuss thermodynamics we have to shift our point of view from electrons in quantum dots to a fully translationally invariant 2DEG as it can be obtained, for instance, in a GaAs heterostructure. The Hamiltonian of such a system can be written as

\[
H = H_{el} + H_{hyp},
\]

where \(H_{el}\) describes the Hamiltonian of the interacting electron gas, and

\[
H_{hyp} = \sum_i A S_i \cdot I_i
\]

is the hyperfine interaction between the electron and nuclear spins. We have chosen here a tight binding, Kondo-lattice formulation of the problem, where \(S_i = (S_i^x, S_i^y, S_i^z)\) is the operator of an electron spin in a Wannier state centered at lattice site \(i\) and the \(I_i = (I_i^x, I_i^y, I_i^z)\) are the nuclear spin operators as before. For GaAs we have \(I = 3/2\). Notice that \(A\) now is position-independent due to the translational symmetry.

In Eq. 2 we have not included the direct dipolar interaction between the nuclear spins. This interaction has the smallest energy scale in the system, \(E_{dd} \approx 100\, nK\) \[14\]. It is much weaker than the effective nuclear spin interaction discussed below and, in particular, \(E_{dd}\) is much smaller than typical experimental temperatures. This allows us to entirely neglect the direct dipolar interaction.

For GaAs, \(A \approx 90\, \mu eV\) \[14\], which compares to typical Fermi energies of \(E_F \approx 10\, meV\) \[13\]. The small ratio \(A/E_F \sim 10^{-2}\) implies a separation of time scales. Electron relaxation times are much shorter than typical time scales of the nuclear spins. This allows us to decouple the systems and focus on the magnetic properties of the nuclear spin system alone, where the effective spin-spin interactions are carried through the response of the equilibrium electron gas to local magnetic excitations, i.e. the electron spin susceptibility.

Technically, the first step is to reduce the still quasi-2D (due to the finite thickness of the 2DEG) problem to a true 2D problem. Since the electrons are confined in a single mode in the direction orthogonal to the 2D plane, the nuclear spins along a column in this direction are all almost identically coupled to the electrons and so effectively locked in a ferromagnetic alignment and behave like a single (effectively large) nuclear spin. The problem becomes, therefore, truly 2D.

The effective Hamiltonian describing this situation can then be obtained, for instance, through a Schrieffer-Wolff transformation followed by the integration over the electron degrees of freedom \[4\, 5\]. It is given by

\[
H_{eff} = - \sum_{ij} J^{qz}(r_i - r_j) I_i^z I_j^z = - \frac{1}{N} \sum_{qz} J^{qz} S_i^z S_j^z,
\]

where \(N\) is the number of sites in the system, \(\alpha = x, y, z\) and the lattice indices \(i, j\) run over the two-dimensional lattice with site positions \(r_i, r_j\). Furthermore

\[
J^{qz}(r_i - r_j) = \frac{A^2}{8 \hbar a} \chi^{qz}(r_i - r_j)
\]

is the effective Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction \[16\], with \(n_s = a^{-2}\) the nuclear spin density and

\[
\chi^{qz}(r_i - r_j) = - \frac{i}{\hbar} \int_0^\infty dt \langle [S^{qz}(t), S^{qz}(0)] e^{-i\eta t} \rangle,
\]

is the static electron spin susceptibility \((\eta > 0\) is infinitesimal). The Fourier transforms are defined as \(I^{qz} = \sum e^{i\eta \cdot q} I^{qz}\) and \(J^{qz} = F \int d\mathbf{r} e^{-i\eta \cdot q} J^{qz}(\mathbf{r})\), and \(N\) is the number of sites in the system. We shall henceforth consider only isotropic electron systems, allowing us to drop the \(\alpha\) index in \(\chi\) and \(J\).

Much of the magnetic properties of the nuclear spins depends, therefore, directly on the shape of \(J_q\), i.e. on the electron susceptibility \(\chi(q)\). Below we will first investigate which features of \(\chi(q)\) are required such that nuclear ferromagnetism is stable. Then we shall see that electron interactions can indeed lead to such a behavior.

Yet before starting we have to comment on the Mermin-Wagner theorem \[17\], which states that long
range order in Heisenberg-like models in 2D is impossible, provided that the interactions are sufficiently short ranged. The RKKY interaction, however, is long ranged. But it is also oscillatory, and it has been conjectured recently [18] that the Mermin-Wagner theorem extends to RKKY interactions that are carried by noninteracting electrons. Below we find a direct confirmation of this conjecture, indicating that electron interactions in addition to the long range character of $J_q$ play the crucial role.

MEAN FIELD THEORY

As a first (naive) approach, we can look at the problem of nuclear magnetism on the mean field level, similar to the approach used by Fröhlich and Nabarro (FN) for bulk metals more than 60 years ago [19]. We skip here the explicit calculation as it is a standard Weiss mean field calculation. Instead, let us clearly state the main assumption behind FN’s approach: If we look at the Hamiltonian \( H \) we see that the energy can classically be minimized if the \( I_i^a \) align in a single spatial Fourier mode \( q \) corresponding to the maximum of \( J_q \). If this maximum is reached at \( q = 0 \), the ground state is ferromagnetic. FN implicitly assumed that the physics is entirely determined by this maximum energy scale \( J_0 \) and investigated the Hamiltonian \( H_{FN} = -\frac{k_B}{4} \sum_{ij} \mathbf{I}_i \cdot \mathbf{I}_j \), for which the mean field theory is exact.

We have to retain two important points from this theory: On the one hand, the ground state depends on the maximum of the \( J_q \), and a ferromagnet is only possible if it is reached at \( q = 0 \). If \( q \neq 0 \) implies a different magnetic order, e.g. a helimagnet. On the other hand, this theory depends on a single energy scale, \( \max_q J_q \) (e.g. \( J_0 \)), which we associate with a temperature \( T_{MF} \) through

\[
k_B T_{MF} = \max_q J_q, \tag{7}\]

where \( k_B \) is the Boltzmann constant. Since this is the only energy scale in the system, all thermodynamic quantities must directly depend on it. For instance, the Curie temperature is given by [19]

\[
T_c^{MF} = \frac{I(I+1)}{3} T_{MF}. \tag{8}
\]

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The mean field theory is, however, inconsistent with the RKKY interactions for noninteracting electrons. In the noninteracting case the electron spin susceptibility is exactly known and is given by the Lindhard function [20], which is constant for \( 0 < |q| < 2k_F \) [see Fig. 1(a)], where \( k_F \) is the Fermi momentum. There is therefore no well defined \( q \) at which \( J_q \) is maximum. “Guessing” a ground state about which we can proceed with a mean field theory as above is no longer possible.

In fact, it is straightforward to show that such a ground state cannot be stable. For this purpose we look at fluctuations about an ordered ground state. In Heisenberg type systems the lowest lying excitations are magnons, collective long ranged spin wave excitations. The calculation of the magnon dispersion about a ferromagnetic order is a standard textbook exercise (see e.g. [21]) and leads to

\[
\hbar \omega_q = 2I(J_0 - J_q). \tag{9}\]

We shall henceforth assume that \( J_q \) is independent of the direction of \( q \) and write \( J_q, \chi(q) \), as well as \( \omega_q \). The fact that \( J_q \) is constant for \( 0 < q < 2k_F \) means that there is a continuum of magnon excitations at zero energy \( \omega_q = 0 \) [see Fig. 2(c)], and every magnon slightly decreases the global magnetization. The assumed ground state magnetization cannot be stable, the nuclear spin system is disordered. This is a direct illustration of the extension of the Mermin-Wagner theorem conjectured in [18].

From Eq. (9) we see, however, that a ferromagnetic ground state becomes stable if \( \omega_q \) [i.e. \( \chi(q) \)] increases monotonically with \( q \) [Fig. 1(b) and Fig. 2(d),(e)]. As we shall see below, nonanalytic corrections to \( \chi(q) \) by electron-electron interactions can indeed lead to a linear increase in \( q \). The main effect is illustrated in Fig. 1(b). The electron-electron interactions modify the RKKY interaction \( J_q \) in that they introduce a new energy scale \( T^* \), which characterizes the shape of \( J_q \). Stronger electron-electron interactions lead to a larger slope of the linear increase of \( \chi(q) \) at small \( q \). Using this slope and \( 2k_F \) as the only inverse length scale available for the electron gas, the new energy scale must be set by the quantity

\[
k_B T^* = (2k_F) \frac{d \omega_q}{dq} \bigg|_{q=0}. \tag{10}\]
The physics is, therefore, no longer dominated by the single scale $T_{MF}$. Thermodynamic quantities such as the magnetization per site $m(T)$ or the critical temperature $T_c$ must be a function of these available scales: $m(T) = m(T_{MF}, T^*, T)$, $T_c = T_c(T_{MF}, T^*)$, etc.

We can again use the magnon description to shed more light on this dependence. The average magnetization per site can be written as

$$m = I - \frac{1}{N} \sum_q \frac{1}{\omega_q - \hbar \omega_0/k_B T - 1} = I - \frac{a}{2\pi} \int \frac{dq}{\omega_q - \hbar \omega_0/k_B T - 1},$$

with $a$ the nuclear lattice constant. The summation/integration runs over the first Brillouin zone of the nuclear system. We see that this integral converges if $\omega_0 \approx q$ for $q \to 0$, and so the linear corrections to the susceptibility are essential for the existence of a finite critical temperature. The magnon integral is dominated by this linear behavior up to $T \approx T^*$. For these temperatures, we can explicitly calculate the magnon integral and obtain

$$m = I \left(1 - T^2/T_0^2\right),$$

with

$$T_0 = \frac{I}{2k_F} \sqrt{\frac{3}{\pi}} \frac{\lambda_F}{a} T^*,$$

where $\lambda_F = 2\pi/k_F$ is the Fermi wavelength.

We stress that the calculation leading to Eq. (12) is valid only in the limit $T \lesssim T^*$, where the spin waves form a dilute gas. At higher temperatures the number of spin waves increases and their wavelengths become shorter, leading to a breakdown of the spin wave theory. The temperature $T_0$ in Eq. (12) has to be interpreted accordingly: We see that $T_0$ sets a characteristic temperature scale for the magnetization $m(T)$. Further corrections to Eq. (12), and mainly the dependence on $T_{MF}$, are of exponential form, $\sim e^{-T_{MF}/T}$, and are uniquely determined by the spin wave modes beyond the validity of the theory. In addition, from Fig. 1 we see that $T^*$ generally (i.e. for not too weak interactions) is comparable to $T_{MF}$, which is proportional to the maximum of $\chi(q)$. This means that up to temperatures $T \approx T_{MF}$, the magnetization is essentially independent of the $T_{MF}$ scale.

If we conjecture on this basis that $T_{MF}$ indeed is insignificant for the thermodynamics, then $T_0$ must set the scale for the Curie Temperature $T_c$,

$$T_c \sim T_0 \sim \frac{\lambda_F}{a} T^*.$$

In contrast to the mean field result [8], this estimate is consistent with $T_c \to 0$ for noninteracting electrons. The prefactor $\lambda_F/a \approx 10^2$ (in GaAs) is a consequence of coupling the electron system (with length scale $\lambda_F$) to the nuclear spin system (with length scale $a$), and leads to a strong increase of the characteristic temperature $T_0$ compared with $T^*$. A further discussion of the renormalization of $T_0$ can be found in [9].

In order to give a numerical estimate of $T_0$ we need to investigate the interactions between electrons in the 2DEG.

**INTERACTION CORRECTIONS TO THE SUSCEPTIBILITY**

Interactions and correlations between particles become increasingly important in low dimensions due to a reduction of the available phase space for particle scattering. It is not too surprising, therefore, that electron-electron interactions in the 2DEG lead to deviations from the thermodynamics of standard Fermi liquids. Such deviations have attracted some attention over the last years theoretically [22–24, 25, 26, 27, 28, 29, 30, 31, 32] and experimentally [33]. Of specific importance for our case are Refs. [24, 27, 28], where explicitly self-energy corrections to the susceptibility were calculated for a screened Coulomb interaction $U(x-y) = U \delta(x-y)$ in 2D. Nonanalytic behavior appears at second order perturbation in $U$ and leads to

$$\delta \chi(q) = \chi(q) - \chi(0) = -q |\chi(0)| |\Gamma_s|^2/3\pi k_F^2, \quad q \ll 2k_F,$$

where $\Gamma_s = -U m/4\pi$ is the bare $2k_F$ backscattering vertex and $m$ the effective mass. We emphasize the nonanalytic behavior on the modulus $q = |q|$, which cannot be

![FIGURE 2. Possible shapes for the magnon dispersion $\omega_q$](image)
derived within a standard Fermi liquid theory. This linear \(|q|\) dependence is indeed the necessary dependence for a nuclear ferromagnet as discussed above. The problem here is, however, the sign of this correction: If we feed this \(\chi(q)\) back into the spin wave spectrum \([9]\), we see that \(\omega_q\) becomes negative for \(q \neq 0\), meaning that the assumption of ferromagnetic order is incorrect and that such an order is in fact unstable. In such a situation a different, helical order can be possible, and this interesting possibility is discussed in \([5]\). Here we focus instead on a further renormalization of the nonanalytic correction, which can reestablish the ferromagnetic order.

We restate the comment made at the beginning of this section that correlation effects are important in low dimensions. A result from perturbation theory, such as Eq. (13), undergoes further renormalization by higher order processes and can so change considerably its shape. This indeed can happen if we push further the diagrammatic calculation of \([26, 27, 28]\) and include the full summation of selected classes of diagrams such as, for instance, the Cooper channel renormalization of the two-particle scattering vertex \([34]\). The Cooper channel renormalization for \(\chi(T)\) has been considered recently in \([29, 30, 31, 32]\). The effect of this renormalization on \(\chi(q)\) has been estimated in \([5]\) and very recently explicitly calculated in \([35]\). The obtained corrections to \(\chi(q)\) are nonuniversal and depend on detailed cutoff scales of the renormalization such as Fermi energy, temperature, level spacing, etc. The stability of the ferromagnetic (or helical) phase seems, therefore, to depend on a quantity which is difficult to control.

Yet we must note that this is the result of a perturbative renormalization, the summation over selected classes of diagrams, as well as the result of a screened short ranged electron-electron interaction. Alternative approaches lead to more predictable results: If we consider long ranged Coulomb interactions within a local field factor approximation (which is a semi-empirical generalization of the RPA approximation \([20]\)), the spin wave spectrum is always positive and a ferromagnet is stable. Such effective theories, however, have eventually the same difficulty of control as the summation over classes of diagrams. Due to this, it seems so far that the final determination of the shape of the susceptibility probably has to rely on experiments and numerics. The latter two approaches should then probe not only the slope \(\partial \chi / \partial q \) at \(q \approx 0\) but also \(\chi(q \sim 2k_F)\), which is important to discriminate between the different scenarios shown in Fig. 2 by the curves (a) and (b) on the one hand, and by (e) and (f) on the other hand. We stress that numerics should directly target \(\chi(q)\) and not, as for instance done in \([34, 37]\), the local field factor. The relation between the latter and \(\chi(q)\) is actually singular at \(q = 2k_F\), which amplifies the noise in the Monte Carlo data of \([34, 37]\) and makes a conclusion on \(\chi(q)\) unreliable.

Assuming, however, that we have found a window in which a nuclear ferromagnet is stable, we can estimate \(T_0\) from the different calculation schemes. A detailed discussion is given in \([5]\). We find that remarkably all schemes provide comparable values. As anticipated much depends on the strength of the electron interactions, which can be quantified by the commonly used dimensionless parameter \(r_s\) (see e.g. \([20]\)) expressing roughly the ratio of Coulomb over kinetic energies of the electrons. In the 2DEG \(r_s\) scales with the electron density \(n_e\) and \(k_F\) as \(r_s \propto 1/\sqrt{n_e} \propto k_F^{-1}\). Values up to \(r_s \sim 8\) can be reached experimentally nowadays. With increasing \(r_s\), the scale \(T_0\) is enhanced through two main effects: First, \(k_F^{-1}\) increases linearly with \(r_s\). Second, \(|\chi(0)|\), which is essentially the Pauli susceptibility at small \(r_s\), increases linearly with \(r_s\) \([20]\). We moreover note that larger \(r_s\) drive the system closer to the ferromagnetic Stoner instability, which would occur at \(r_s \sim 20\) \([38]\). At this instability \(\chi(0)\) would diverge. The proximity to the instability provides an additional prefactor enhancing \(|\chi(0)|\). Let us note that the increase of \(|\chi(0)|\) enhances both, \(T^*\) and \(T_{MF}\). For \(r_s \sim 5\), we then obtain \(T_0 \sim 0.3 - 0.4\) mK, and for \(r_s \sim 8\) the larger \(T_0 \sim 0.7 - 1\) mK, where the spread is due to the different analytic approaches \([5]\).

**ONE-DIMENSIONAL SYSTEMS**

With a further reduction of dimensionality electron-electron correlations become even more important. One-dimensional conductors of interacting electrons, such as quantum wires or carbon nanotubes, form a Luttinger liquid rather than a Fermi liquid. Accordingly the shape of the RKKY interaction \(J_q\) between the nuclear spins in these systems changes drastically. \(J_q\) is dominated by backscattering processes at \(q = 2k_F\), and the nuclear spins order in a helical phase with this wave vector.

In contrast to the 2D case, the feedback of the ordered nuclear field on the electrons is now crucial. It leads to a spontaneous restructuring of the electron wave functions, i.e., to an order in the electron system as well. The feedback stabilizes this order, and the critical temperature \(T_c\) can increase by several orders of magnitude compared with the case where the feedback has been neglected.

We have performed a detailed study of this effect in \([39]\) for the example of single-wall carbon nanotubes made from the \(^{13}\text{C}\) isotope (which has a nuclear spin \(I = 1/2\)). Such nanotubes have become available very recently \([40, 41, 42]\). The hyperfine interaction is very weak in such systems. Due to the feedback, however, we determine a \(T_c\) in the millikelvin range. The ordered phase leads furthermore to a universal reduction of the conductance and should be detectable by standard transport measurements.
CONCLUSIONS

We have discussed here the conditions necessary for ferromagnetism of nuclear spins embedded in a 2DEG. Such a system is naturally in the RKKY regime and the interaction between the nuclear spins is carried by the electron spin susceptibility. Electron correlations are crucial, and the stability of the magnetic (ferro or different) phase depends on the nonanalytic behavior, linear in momentum $|q|$, of the electron spin susceptibility. The nonanalytic behavior cannot be found from standard Fermi liquid theory. It is a consequence of a strong renormalization of the electron-electron interaction which, to the best of our knowledge, strongly depends on a nonuniversal cutoff scale specific for the sample under investigation. The stronger the interaction, however, the higher also the critical temperature for the nuclear (ferro)magnet. Our estimates show that a transition temperature reaching up to the millikelvin range may be achievable. A similar temperature range is estimated for one-dimensional nanotubes, where the ordered phase should be detectable by conductance measurements.

To conclude, we stress that such physics is not restricted to nuclear spins in metals. We expect a similar behavior for any Kondo-lattice system in low dimensions with interacting conduction electrons.

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