An \textit{ab initio} effective Hamiltonian for magnetism including longitudinal spin fluctuations

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We discuss the use of the magnetic force theorem (MFT) using different reference states upon which the perturbative approach is based. Using a disordered local moment (DLM) state one finds good Curie (or Neél) temperatures, and good energetics for planar spin spirals in the 3d magnets Fe, Co (fcc), Mn, Cr. On the other hand the ferromagnetic reference state provides excellent energetics for small $\theta$ spin spirals in Fe, Co and Ni, and by extension magnon energies under the assumption of adiabacity. However, planar spin spiral energetics and transition temperatures for Ni, Fe, Mn, and Cr show worse agreement. The reasons for this, and for the case of fcc Co where both approaches work very well are discussed. We further provide an extension of the mapping of the quantum problem to include longitudinal fluctuations, and discuss the role they will play in magnetic phase transitions. This construction is tested using planar spin spirals where $\mathbf{q}$ is fixed but the moment is allowed to relax. It is demonstrated that results from the magnetic force theorem approach and directly calculated \textit{ab initio} values agree very well.

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I. INTRODUCTION

Constructions based upon the mapping of the ground state energetics of a quantum system onto an appropriate classical model have been found to be of great use in solid state physics. Direct and accurate quantum mechanical calculations of a system are still only practically possible for system sizes of the order of hundreds of atoms, and so in order to explore systems of much larger size the substitution of the quantum problem by an classical one is necessary. The most prominent example of such a procedure is probably the Ising model in alloy physics, and a great deal of progress in understanding the ground state and phase behavior of binary alloys has been made in this way. In this case the classical variable is just the site occupation. In the case of magnetic systems the appropriate classical model is the Heisenberg model, which in its simplest form reads

\begin{equation}
E = -\sum_{ij} J_{ij}^{(2)} s_i \cdot s_j
\end{equation}

where $J_{ij}^{(2)}$ are the exchange parameters and $s_i$ a $d$ dimensional classical vector. In the case of magnetism the emergence of such local moment variables is not as obvious as that of site occupation in the alloy situation.

In fact the fundamental justification for the usefulness of such variable comes from a separation of time scales. If one considers a magnetic material at some low temperature the moment on a particular site will fluctuate rapidly on a time scale given by the electron hopping. However, averaged over longer time scales the expectation value of the site moment is a more stable quantity. In particular, the magnon frequency is often orders of magnitude slower. This adiabatic condition allows the deployment of a classical Heisenberg model to study the low temperature magnetic dynamics. Given an accurate procedure for mapping between the quantum and classical regimes the study of the dynamics of complex materials is facilitated. A further use, in general less justified, is the study of phase transitions in magnetic systems. Here one extends the concept of the local moment to high temperatures, and assumes that the statistical mechanics of the high temperature state is given simply by the classical partition function of the ground state energy.

There are several ways of performing such a map, which can be divided into so called structure inverse method and perturbative methods. In structure inverse methods one extracts the parameters for the Heisenberg model from the total energies of a set of magnetic structures. The structures used in this set are most often spin spirals, and there has been some debate as to whether small $\theta$ spin spiral or planar spin spiral structures should be used. The alternative perturbative approach comes from considering the rotation of a pair of spins in some reference state by opposite and vanishing angles. This leads directly to the parameters of a classical Heisenberg model. Note that there is no restriction on the form of reference state in this method.
The relative merits of such approaches have been well cataloged in the case of the analogous alloy situation. The structure inverse type methods have the advantage that they are in principle limited in accuracy only by the underlying method used to determine the total energies. On the other hand the range and importance of specific interactions (which may be more than simply pair interactions) is not known beforehand, and further it is difficult to systematically converge the method with respect to the range and type of interactions. A particular problem may arise if the interactions are further configuration dependent. In perturbative methods the situation is reversed. With such an approach the convergence of the interaction set is relatively trivial, and by probing different reference states one can (as will be discussed) elucidate the configuration dependence if any. On the other hand such methods make approximations beyond those of the underlying total energy method.

The two original formulations of force theorem based methods were based on the ferromagnetic and disordered local moment reference states, we will refer to these two approaches as ferromagnetic magnetic force theorem (FM-MFT) and disorder local moment magnetic force theorem (DLM-MFT). The former has become widely used in the intervening years, but not the latter version. This lack of use is most likely a combination of the result of the initial calculation of the Curie temperature of Fe, which turned out to be 2700 K, and the fact that the equilibrium DLM moment in many systems is substantially different from that of the ground state magnetic structure.

With regard to the first point it was noted recently that the large value found for the Curie temperature was simply due to numerical error in the original work, the correct result being around 1080 K which compares well with the experimental Curie temperature of 1040 K.

The second point is more critical, however when one regards the DLM state as merely a reference medium, and not as a state with which one attempts to describe the paramagnetic state, then this problem may be circumvented by the use of a fixed spin moment procedure.

On the other hand, the FM-MFT is also known to suffer from some failures however. Most recently it was shown that using an accurate underlying method for the electronic structure of the FM reference state resulted in a Curie temperature of 550 K in the Local Density Approximation, roughly half the experimental value. A long standing problem has been the Curie temperature of Ni which has been found to be much smaller than the experimental value of 640 K in all works using this method.

This paper addresses the question of the role of the effective medium in the mapping of the quantum problem, and in particular whether certain properties are best calculated with one or the other approach. In Section 2 we describe briefly the formalism used and give details of our computations. Section 3 presents results for the energetics of planar and small \( \theta \) spin spirals, and compares the results of direct calculation with those from the FM-MFT and DLM-MFT methods. We then present the results of Curie and Néel temperatures calculated with both methods. In section 5 we introduce an approach for the inclusion of longitudinal fluctuations in conjunction with perturbative methods, after which we conclude.

II. FERROMAGNETIC AND DISORDERED LOCAL MOMENT EXCHANGE INTEGRALS

The magnetic force theorem makes use of Lloyd’s formula in Green’s function formalism for the change in the integrated density of states upon the embedding of some cluster. For the FM-MFT one arrives at the following equation for the bi-linear exchange term in the Heisenberg model,

\[
J^{(2)}_{ij} = \frac{1}{4\pi} Im \int d\tau_{LR} \langle \sum_{\sigma} \tau_{ij} \rangle \langle \sum_{\sigma} \bar{g}_{ij} \rangle \langle \sum_{\sigma} \bar{g}_{j\sigma} \rangle \ (2)
\]

where \( P_{i} \) and \( P_{j} \) are the potential functions and \( g_{ij} \) elements of the auxiliary Green’s function. The DLM-MFT makes use of the Coherent Potential Approximation (CPA) for the construction of the disordered local moment reference state. Note that the DLM is not here deployed to model the paramagnetic state, which will in general have non-zero short range order parameters not treatable within CPA, but simply as a reference medium for the embedding of a spin cluster. The formula for the exchange parameters becomes now

\[
J^{(n)}_{ij} = \frac{1}{4\pi} Im \int d\tau_{LR} (\Delta t_{ij} \bar{g}_{ij} \Delta t_{j} \ldots \Delta t_{k} \bar{g}_{kl}) \ (3)
\]

where \( \bar{g}_{ij} \) are now elements of the auxiliary coherent Green’s function and \( \Delta t_{i} \) is the difference of spin up and spin down on site scattering \( t \) matrices at site \( i \) which are related to the potential function at site \( i \) and coherent potential function by

\[
t_{i} = [1 + (\bar{P} - P_{i})\bar{g}_{0}]^{-1}(\bar{P} - P_{i}) \ (4)
\]

Eq. (3) allows for the calculation of any embedded cluster of spins, however from the symmetry of the exchange interactions only even clusters have non zero energy. The full Heisenberg expansion is in fact given by

\[
E = \sum_{ij} J^{(2)}_{ij} \langle s_{i}, s_{j} \rangle - \sum_{ij} J^{(2-2)}_{ij} \langle s_{i}, s_{j} \rangle^{2} - \ldots - \sum_{ij} J^{(n-n)}_{ij} \langle s_{i}, s_{j} \rangle^{n}
\]

\[
- \frac{1}{4!} \sum_{ijkl} J^{(4)}_{ijkl} \langle s_{i}, s_{j} \rangle \langle s_{k}, s_{l} \rangle + \langle s_{j}, s_{k} \rangle \langle s_{l}, s_{i} \rangle + \langle s_{l}, s_{i} \rangle \langle s_{j}, s_{k} \rangle
\]

\[
+ (s_{i}, s_{j})(s_{k}, s_{l}) - \ldots \ (5)
\]
III. COMPUTATIONAL DETAILS

The electronic structure calculations were performed in the Korringa-Kohn-Rostocker (KKR) scheme in the atomic sphere approximation (ASA). The basis used contained s, p, d, and f orbitals. Multipole moment corrections for the charge density up to \(l_{\text{max}} = 6\) were included. The exchange correlation functional used was the Local Density Approximation in all cases with the Perdew, Burke, and Ernzerhof parameterization of the results of Ceperly and Alder. The integration of the Green’s function was taken in the complex plane with 16 energy points on a semi-circular contour. The experimental room temperature lattice parameters were used in all cases expect for that of fcc Mn where an expanded lattice parameter was used to ensure an anti-ferromagnetic solution.

IV. SPIN SPIRALS

The energy of a spin spiral in the Heisenberg model is given very simply by a Fourier transform as

\[
E(q) = -J(q) - \sin^2 \theta [J(q) - J(0)] - \ldots
\]

Note that if only bi-linear terms are included in the Heisenberg expansion then the \(\theta\) and \(q\) dependencies decouple and the difference between a small \(\theta\) spin spiral and a planer spin spiral becomes simply a matter of scale.

One would intuitively expect that, by virtue of its perturbative nature, the magnetic force theorem would provide a good description of the magnetisation energy changes for configurations close to the reference state. Hence in the case of the FM-MFT one must thus measure energies from the energy of the FM state, and in the case of the DLM-MFT from the DLM state. One can note that Eq. (6) (and hence Eq. (6)) measures energies relative to the DLM energy, which follows simply from the vanishing of the spin products in this case.

In Fig. 1 and Fig. 2 we present the results of direct calculation of the magnetization energies of planar spin spirals in Ni, Co, Fe, Mn, and Cr with \(q\) vectors in the standard reciprocal space paths as indicated. Also shown is the evaluation of Eq. (6) using only bi-linear interactions determined via the FM-MFT and DLM-MFT. For simplicity we show all energies here measured from the non-magnetic state, which is achieved simply by the addition of \(E_{\text{dln}}\) to Eq. (6). Expect near the \(\Gamma\) the perturbation from the FM state is significant for planar spin spirals and indeed one can see that the agreement is generally better between the DLM-MFT and the directly calculated results, particularly in the case of the anti-ferromagnets (AFM) Cr and Mn, and Fe near the H point (the special point generating the AFM structure in real space). In particular, the FM-MFT can hardly be said to work at all for Cr and Mn. Since the ground state is not FM this is not surprising, and one might expect that the use of the AFM state as the reference medium would produce better results. As can be seen in Fig. 2 this is not the case.

The reason for this failure may come from the neglect of higher order terms in Eq. (6). For the DLM-MFT we find that the higher order terms in the Heisenberg expansion are very small in comparison to the bi-linear terms and may be neglected. On the other hand for the FM-MFT, although we have not directly evaluated the higher order terms, the "renormalized" bi-linear interactions differ substantially from the bare interactions which indicates this may be true, in particular for the case of Mn and Fe. In a weakly ferromagnetic system (in the Stoner sense), such as Fe there will be both spin up and spin down Fermi surfaces and hence the scattering from spin up to spin down states in Eq. (6) will be greater than in the case of a strong ferromagnet such as fcc Co or Ni.

One can also see that, with the exception of Co, the DLM-MFT appears to systematically overestimate the stability of the FM state as compared to the direct calculation. This is particularly pronounced in the case of Ni. The energetics of these planar spin spirals give an indication of the regime where the DLM-MFT is applicable in the cases where it fails in the FM limit. It can be seen that for both Ni and Fe the deviation between the directly calculated magnetization energy and the DLM-MFT result is small except for region around the \(\Gamma\) point. Interestingly, one notes that the FM-MFT actually underestimates the magnetization energy of the FM state in all cases.

We now turn to the calculation of small \(\theta\) spin spirals. As discussed above it is expected that the FM-MFT should work very well in this case, and that is indeed seen in Fig. 3. In this case the energy is measured relative to the FM energy. In fact, for Ni and Co the method appears to give practically exact results, whereas for Fe it is slightly off at the H point. However, this difference might well vanish as \(\theta \rightarrow 0\) since the spin spirals shown here actually had \(\theta = \pi/10\), and furthermore the difference with the planar spin spiral is most pronounced at the H point. From a linearisation of the Bloch equation of motion the energetics of small \(\theta\) spin spirals may be simply related to magnon energies, and hence the FM-MFT appears to provide an excellent way to calculate magnon spectra.

This provides strong computational support for the recent work by Katsnelson et al. who find the correction to the FM-MFT provided recently by Bruno not to be needed for the calculation of magnon energies, although it might be for Curie temperatures. The question as to whether the FM-MFT should be able to provide correct Curie temperatures, and under what conditions, will be discussed in the next section.

From Fig. 3 it is further seen that the DLM-MFT overestimates small \(\theta\) spin spirals by a factor of 2 for Ni and for Fe near the H point. Again, as was seen in the case of planar spin spirals, for Co the difference between the FM-MFT and the DLM-MFT results is rather small.
It is interesting to consider the reason for the profound difference between the description of energetics of spin spirals provided by the FM and DLM magnetic force theorem interactions for the case of Fe and the antiferromagnets Mn and Cr, and on the other hand the good description provided by both approaches for the case of Co. It is well known that hcp and fcc Co are strong ferromagnets with few minority bands crossing the Fermi energy $E_F$. Since the effect of non-collinearity is to mix spin up and spin down states, what were previously orthogonal majority and minority spin bands are no longer so and thus which could cross in the FM state repel creating hybridisation gaps. This effect will of course be strongest in weak ferromagnets with a large spin up and spin down density of states (DOS) at $E_F$, and less important for strong ferromagnets. Thus the reason for the good description provided by both the FM and DLM reference states in the case Co is the hybridisation is not important and hence the difference between the FM and non-collinear DLM reference states is less important. In contradistinction the energetics of Fe, being a weak ferromagnet, should be profoundly affected by this difference. This also naturally explains why the DLM-MFT gives better agreement in the case of planar spin spirals in all cases.

An inspection of the small $\theta$ energetics for Ni shows however that the DLM-MFT does not work very well in this strong ferromagnet. The reason for this is most likely the itinerant nature of the magnetism in Ni.

V. CURIE TEMPERATURE AND SPIN STIFFNESS CALCULATIONS

Using the interactions derived via either the FM or DLM versions of the MFT one may perform Monte Carlo calculations to determine the magnetic transition temperature. Results of this procedure are shown in Table I.

The arguments given above explain the result for Fe found in Ref. 4, however they also allow for the following criteria for the FM-MFT to provide accurate Curie temperatures to be formulated: The FM-MFT should work
FIG. 3: Small $\theta$ spin spiral energetics for Fe, Co and Ni directly calculated from KKR-ASA method (filled symbols) and evaluated using magnetic force theorem with ferromagnetic (FM, dashed lines) and disordered local moment (DLM, continuous lines) reference states.

FIG. 4: Relaxation of the magnetic moment of planar spin spirals in Ni, Co, Fe, Mn, and Cr. Shown are both the results of direct calculation via KKR-ASA method (open circles) and evaluation from the m-dependent magnetic force theorem approach.

for strong local ferromagnets, where it will agree with DLM-MFT, and not otherwise. The results of Table II show that this is indeed the case, with the Curie temperatures for Co being close in both approaches but there existing a large difference for Ni and Fe. It is extremely interesting that the Curie temperature for Ni comes out to be 820 K which is higher than the experiment. The result from FM-MFT is 320 K which agrees with previous works.

The reason for the overestimate of the Curie temperature comes back to the imposition of a fixed spin moment constraint on the DLM state. This is neccessary for the use of the DLM as a reference medium for the force theorem argument, but is artificial for Ni near the Curie temperature. There the local moment will certainly be lower than the ground state moment and hence it is that which should be used, and this will have the effect of lowering the Curie temperature from that obtained with the ground state moment. Thus if the moment was allowed to take on its true value, the agreement between experiment and theory may be quite reasonable for Ni. Of course, the DLM state has zero equilibrium moment for Ni and so it can not simply be allowed to be a free parameter, as works well for Fe. A way in which progress can be made is described in the next section.

Near the $\Gamma$ point the magnon spectra behaves as $Dq^2$ and this fact is easily used to show that $D$ can be expressed in terms of the interactions as

$$D = \frac{2\mu_B}{3m} \sum_j J_{0j} R_{0j}^2$$

where $m$ is the magnetic moment of the ferromagnetic state. Results for the spin stiffness of the ferromagnetic 3d metals are shown in Table II. The FM-MFT provides values in reasonable agreement with experiment, as expected, however the DLM-MFT also does for Co and Fe. For Fe this may be expected from the arguments above, however for Fe it is more surprising since there appear pronounced differences between the interactions in the FM and DLM states. Due to the long ranged nature of the interactions derived from the FM-MFT, D was evalu-
FIG. 5: Relaxation energy of planar spin spirals in Ni, Co, and Fe. Shown are both the results of direct calculation via KKR-ASA method (open and filled symbols) and evaluation from the m-dependent magnetic force theorem approach (dashed lines).

\[ E = \sum_{i} J^{(1)}_{i} m_{i}^2 - \sum_{ij} J^{(2)}_{ij} m_{i} m_{j} s_{i} s_{j} \]  

where \( m \) is the magnetic moment. The question now is how to determine \( J^{(1)}_{i} \) in a way consistent with the MFT. An obvious way that suggests itself is a modified application of the structure inverse method whereby one demands that the DLM energy is well reproduced. Since under complete spin disorder all spin products vanish Eq. 8 reduces to \( E_{\text{dlm}} = J^{(1)}_{i} \), where for simplicity of expression we consider only simple lattices having one inequivalent lattice. The \( m \) dependence of both \( E_{\text{dlm}} \) and the exchange integrals \( J^{(2)}_{ij} \) may then be easily obtained analytically by fitting polynomials to a set of such values calculated for different \( m \). This forms a very natural link with the DLM-MFT where the DLM reference medium is used itself for the perturbation theory that results in the exchange parameters, however there is no reason why it cannot be used with the FM-MFT as well. In this section we shall consider only the former version though.

The effectiveness of this theory may be tested by again calculating planar spin spirals but with the magnetic moment allowed to take on the equilibrium value, the \( m \)-dependent DLM-MFT (referred to here on in as mDLM-MFT) should then reproduce the \textit{ab initio} equilibrium moments and relaxation energies. It is interesting to see initially how the theory works in the FM limit and these results are presented in Table III. As expected the Co moment is well reproduced, but more surprisingly there is a reasonable agreement in all other cases too. The worst cases, as would be expected, are Fe and Cr.

In Fig. 4 are shown the equilibrium moments of the same planar spin spirals calculated with fixed spin constraint in Section 3. As can be seen the agreement between \textit{ab initio} results and the mDLM-MFT approach is quite reasonable in all cases. In Fig. 5 is shown the relaxation energy of the spin spirals, defined simply as the difference of the energy between the spin spiral with its equilibrium moment and the spin spiral with the moment fixed to that of the equilibrium ground state structure (either FM or AFM).

VI. INCLUSION OF LONGITUDINAL FLUCTUATIONS

The essential reason for the overestimate of the Curie temperature was that the classical Heisenberg Hamiltonian allows for only transverse fluctuations. This means there is no way for the size of the Ni moments to respond to the energy cost of orientational disorder by reducing the exchange splitting. A number of authors have proposed ways to lift this constraint. In both of these methods the parameters of the classical model were obtained by the use of the structure inverse method. What both methods have in common is the addition of an on-site term to Eq. 1 which for our purposes can be written as

\[ E = \sum_{i} J^{(1)}_{i} m_{i}^2 - \sum_{ij} J^{(2)}_{ij} m_{i} m_{j} s_{i} s_{j} \]  

where \( m \) is the magnetic moment. The question now is how to determine \( J^{(1)}_{i} \) in a way consistent with the MFT. An obvious way that suggests itself is a modified application of the structure inverse method whereby one demands that the DLM energy is well reproduced. Since under complete spin disorder all spin products vanish Eq. 8 reduces to \( E_{\text{dlm}} = J^{(1)}_{i} \), where for simplicity of expression we consider only simple lattices having one inequivalent lattice. The \( m \) dependence of both \( E_{\text{dlm}} \) and the exchange integrals \( J^{(2)}_{ij} \) may then be easily obtained analytically by fitting polynomials to a set of such values calculated for different \( m \). This forms a very natural link with the DLM-MFT where the DLM reference medium is used itself for the perturbation theory that results in the exchange parameters, however there is no reason why it cannot be used with the FM-MFT as well. In this section we shall consider only the former version though.

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VII. CONCLUSIONS

We have presented a study of the effect of the choice of the effective medium in approaches based on the magnetic force theorem. We find that for weak or itinerant ferromagnets it is essential to use the DLM-FM reference state, but that for strong local moment ferromagnets both approaches may be used interchangeably. We have further proposed a method whereby longitudinal and well as transverse fluctuations may be incorporated into methods based on the magnetic force theorem.
TABLE I: Transition temperatures, spin stiffness, and $j_0$ for 3d magnets.

|      | $T_c$ | Spin stiffness | $j_0$ | KKR-ASA |
|------|-------|----------------|-------|---------|
| Ni   | 820   | 320            | 624-631 | 1796     | 541     | 555,420 | 3.71 | 9.50 | 5.60 |
| Co (fcc) | 1350 | 1120            | 1388-1393 | 520     | 480     | 580,510 | 15.51 | 15.50 | 15.09 |
| Fe   | 1190  | 550            | 1044-1045 | 313     | 322     | 280,330 | 15.58 | 9.26 | 12.13 |
| Mn   | 450   | -              | -      | -       | -       | -       | -     | -    | -    |
| Cr   | 421   | -              | 321    | -       | -       | -       | -     | -    | -    |

TABLE II: Magnetic moments calculated directly from KKR-ASA and from magnetic force theorem.

|      | $\mu_{DLM}$ | $\mu_{KKR}$ |
|------|-------------|-------------|
| Ni   | 0.69        | 0.62        |
| Co (fcc) | 1.62 | 1.62        |
| Fe   | 2.35        | 2.22        |
| Mn   | 2.86        | 2.79        |
| Cr   | 1.09        | 0.87        |

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