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An *ab initio* effective Hamiltonian for magnetism including longitudinal spin fluctuations

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We discuss the use of the magnetic force theorem using different reference states upon which the perturbative approach is based. Using a fixed spin disordered local moment state one finds good Curie (or Neél) temperatures, and good energetics for planar spin spirals in the $3d$ magnets Fe, fcc Co, Ni, Mn, and Cr, though worse agreement for small θ spin spirals. On the other hand, the ferromagnetic reference state provides excellent energetics for small θ spin spirals in Fe, fcc Co, and Ni, and by extension magnon energies under the assumption of adiabacity. However, planar spin spiral energetics and transition temperatures 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 within force theorem based approaches, 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 this approach and directly calculated *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 are still only practically possible for system sizes of the order of hundreds of atoms, and so in order to explore physics on a greater length scale than this, the substitution of the quantum problem by a 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.[1] 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

$$E = -\sum_{ij} J_{ij}^{(2)} \mathbf{s}_i \cdot \mathbf{s}_j, \tag{1}$$

where $J_{ij}^{(2)}$ are the effective exchange interactions and $\{\mathbf{s}_i\}$ unit vectors. In this case the emergence of such local moment variables is not as obvious as in the alloy situation, where the site occupation is very naturally described by the Ising variable ξ_i which takes on values of 1 or 0 depending on whether the site is occupied by an A or B atom, respectively.

In fact the fundamental justification for the usefulness of such variable comes from a separation of time scales.[2] 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. The validity of this approach is less secure since it involves the neglect of both quantum statistics and longitudinal fluctuations of the magnetization.

There are several ways of constructing the mapping between the quantum system and the Heisenberg Hamiltonian, which can be divided into what are known as structure inverse methods 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. These are most often spin spirals, and there has been some debate as to whether small θ or planar spin spiral structures should be used.[3,4] The alternative perturbative approach comes from considering the rotation of a pair of spins embedded 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.[5] 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 include multisite interactions) is not known beforehand, and 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 configuration dependent. In perturbative methods the situation is reversed. Within such an approach the convergence of the interaction set is relatively trivial, and by probing different reference states one can (as will be shown) elucidate the configuration dependence if any. However, if there are strong hybridization effects specific to a particular noncollinear state then these cannot be captured by interactions calculated from some different reference state. Hence, in general perturbative methods will be less accurate than the underlying electronic structure method used.

The two original formulations of force theorem based methods used the ferromagnetic[6,7] and disordered local moment reference states,[8] and we will refer to these two approaches as the ferromagnetic magnetic force theorem (FM-MFT) and disordered local moment magnetic force theorem (DLM-MFT) throughout this work. The FM-MFT has become widely used in the intervening years but not the DLM-MFT. This may perhaps be explained as a result of the initial calculation of the Curie temperature of Fe which turned out to be 2700 K,[8] substantially above the experimental value of 1045 K. However, it was shown recently that this large value was most likely due to a numerical error in the original work, a modern calculation giving a value of around 1080 K.[5] A more serious problem with the DLM-MFT is found by considering it as a procedure for extracting interactions from the high temperature paramagnetic phase. In this case one is confronted by the fact that the DLM state is manifestly a poor description of this phase as it neglects both magnetic short range order and spin fluctuation entropy. A dramatic example of this failure is that the equilibrium DLM moment for Ni is *zero*, in contradiction to experiment where a local moment may be observed above the Curie temperature.

In contrast, force theorem approaches based on extracting interactions from the ground state structure, such as the FM-MFT, have no such problem as modern electronic structure calculations are known to provide a rather good description of ground state magnetism. The FM-MFT is known to suffer from some failures, however. A long standing problem has been the Curie temperature of Ni which has been found to be much smaller than the experimental value of 624–631 K in all works using this method, typically around 300 K. Further, in a surprising result[5] it was recently shown that using an accurate underlying method for the electronic structure of the FM reference state resulted in a Curie temperature of 550 K *for bcc* Fe in the local spin density approximation, roughly half the experimental value.

Furthermore the fact that the DLM state is only useful as a model of the paramagnetic phase in systems with well defined local moments such as bcc Fe would seem to prevent the widespread application of the DLM-MFT. The problem results from allowing the DLM state to attain its equilibrium moment, which is only strictly necessary if one regards the DLM state as a model of the high temperature paramagnetic phase. In the context of force theorem based methods the DLM can be considered as simply another reference state for the interactions to be calculated from, and *not* as a model of the paramagnetic phase. In this case one may construct a *constrained* DLM state in which (for example) the local moments are *fixed to the same size as the ground state moment*.

This paper addresses the question of the role of the reference state in the mapping of the quantum problem. In Secs. II and III we describe briefly the formalism used and give details of our computations. The usefulness of any MFT approach is easily determined by a comparison of the MFT predictions of magnetic energetics with direct quantum mechanical calculation. Hence, for a fixed spin FM or DLM reference state one should compare the MFT results with direct calculations on structures of identical moment size. Section IV presents this comparison for the energetics of planar and small θ spin spirals. We then present, in Sec. V, the results of Curie and Neél temperatures calculated with both methods. In Sec. VI 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.[6] For the FM-MFT (for simplicity we here consider systems with one basis atom only) one arrives at the following equation for the effective bilinear exchange term in the Heisenberg model

$$J_{ij}^{(2)} = \frac{1}{4\pi}\mathrm{Im}\int^{E_F} dz\, \mathrm{Tr}_L(\Delta g_{ij}^{\uparrow}\Delta g_{ji}^{\downarrow}) \tag{2}$$

with $\Delta = P^{\uparrow} - P^{\downarrow}$ where P^{σ} are the potential functions and g_{ij} elements of the auxiliary Green's function. Considering now the DLM state one can model this as an $A_{0.5}B_{0.5}$ *alloy* with the A and B components the $+m$ and $-m$ spins, respectively.[2] There will now be three possible effective exchange interactions $[J_{ij}^{(2)}]_{AA}$, $[J_{ij}^{(2)}]_{BB}$, and $[J_{ij}^{(2)}]_{AB}$. However, the symmetry of the DLM state ensures that $[J_{ij}^{(2)}]_{AA} = [J_{ij}^{(2)}]_{BB} = -[J_{ij}^{(2)}]_{AB}$. With the aid of the vertex cancellation theorem[9] one can write the interactions simply by substitution of the coherent Green's function for the FM Green's function

$$[J_{ij}^{(2)}]_{xy} = \frac{1}{4\pi}\mathrm{Im}\int^{E_F} dz\, \mathrm{Tr}_L[\Delta_x(\tilde{g}_{ij}^{\uparrow})_{xy}\Delta_y(\tilde{g}_{ji}^{\downarrow})_{xy}], \tag{3}$$

where x and y label component type. This equation may be simplified by noting that

$$t_x^{\sigma} = \frac{(\tilde{P} - P_x^{\sigma})}{1 + (\tilde{P} - P_x^{\sigma})\tilde{g}_0}, \tag{4}$$

$$(g_{ij}^{\uparrow})_{x^{\sigma}y^{\sigma}} = \frac{1}{1+\tilde{g}_0(\tilde{P}-P_x^{\sigma})}\tilde{g}_{ij}\frac{1}{1+(\tilde{P}-P_y^{\sigma})\tilde{g}_0}. \tag{5}$$

Using these and the symmetry properties of the DLM state one finally arrives at

$$[J_{ij}^{(2)}]_{xx} = \frac{1}{4\pi}\mathrm{Im}\int^{E_F} dz\, \mathrm{Tr}_L(\Delta t\tilde{g}_{ij}\Delta t\tilde{g}_{ji}), \tag{6}$$

where $\Delta t = t^{\uparrow} - t^{\downarrow}$.

Equation (6) is reminiscent of the corresponding expression in alloy theory for the calculation of the pairwise interactions $V_{ij}^{(2)}$ needed in the Ising model, and in that context the force theorem approach is known as the generalized perturbation method.[1] In fact it is easy to show that $V_{ij}^{(2)} = -1/8[J_{ij}^{(2)}]_{xx}$.

The close analogy allows one to write down the exchange integral required for the calculation of any embedded cluster of spins in the DLM-MFT

$$J_{ij}^{(n)} = \frac{1}{4\pi}\mathrm{Im}\int^{E_F} dz\, \mathrm{Tr}_L(\Delta t_i\tilde{g}_{ij}\Delta t_j\ldots\Delta t_k\tilde{g}_{ki}). \tag{7}$$

Note that due to the symmetry of the exchange interaction[10] only even clusters have nonzero energy. The full Heisenberg expansion is then given by

$$\begin{aligned}E = &-\sum_{ij} J_{ij}^{(2)}\mathbf{s}_i\cdot\mathbf{s}_j - \sum_{ij} J_{ij}^{(2-2)}(\mathbf{s}_i\cdot\mathbf{s}_j)^2 - \ldots - \sum_{ij} J_{ij}^{(n-n)}(\mathbf{s}_i\cdot\mathbf{s}_j)^n \\ &- \frac{1}{4!}\sum_{ijkl} J_{ijkl}^{(4)}[(\mathbf{s}_i\cdot\mathbf{s}_j)(\mathbf{s}_k\cdot\mathbf{s}_l) + (\mathbf{s}_j\cdot\mathbf{s}_k)(\mathbf{s}_l\cdot\mathbf{s}_i) + (\mathbf{s}_l\cdot\mathbf{s}_i) \\ &\times(\mathbf{s}_j\cdot\mathbf{s}_k)] - \ldots,\end{aligned} \tag{8}$$

where $J_{ij}^{(2)}$ is the bilinear exchange term, $J_{ij}^{(2-2)}$ the biquadratic exchange term (double scattering), and $J_{ijkl}^{(4)}$ a four site exchange interaction. Higher order pair exchange terms are indicated by $J_{ij}^{(n-n)}$.

III. COMPUTATIONAL DETAILS

The electronic structure calculations were performed in the Korringa-Kohn-Rostocker (KKR) scheme in the atomic sphere approximation (ASA) (see Ref. 5, and references therein). The basis used contained s, p, d, and f orbitals. Multipole moment corrections for the charge density up to $l_{max}=6$ were included. The exchange correlation functional used was the local spin density approximation[11] (LSDA) for Ni, fcc Co, Fe, and Mn, whereas for Cr we used the generalized gradient approximation[12] (GGA) since in the ASA approximation the equilibrium moment with LSDA turns out to be rather low at 0.30 μ_B. The integration of the Green's function was taken in the complex plane with 16 energy points on a semicircular 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 antiferromagnetic solution.

In order to calculate the Fourier transform of our effective exchange interactions, one needs quite a high cutoff in the number of shells used, particularly for interactions derived from the FM-MFT. We use up to 200 shells for this procedure, which in turn requires a quite fine **k** mesh for the calculation of g_{ij}. For the fcc, bcc, and sc structures we found that 3345, 3080, and 1540 **k** points, respectively, in the irreducible Brillouin zones was sufficient.

IV. SPIN SPIRALS

The energy of a spin spiral in the Heisenberg model is given very simply by a Fourier transform of Eq. (8) as

$$\begin{aligned}E(\mathbf{q}) = &-J^{(2)}(\mathbf{q}) - \sin^2\theta[J^{(2)}(\mathbf{q}) - J^{(2)}(\mathbf{0})] - J^{(2-2)}(\mathbf{q}) \\ &- 2\sin^2\theta[\mathbf{J}^{(2-2)}(\mathbf{q}) - J^{(2-2)}(\mathbf{0})] - \sin^4\theta[1/2J^{(2-2)}(2\mathbf{q}) \\ &- 2J^{(2-2)}(\mathbf{q}) + 3/2J^{(2-2)}(\mathbf{0})] - \ldots.\end{aligned} \tag{9}$$

Note that if only bilinear terms are included in the Heisenberg expansion then the θ and **q** dependencies decouple and the difference between a small θ spin spiral and a planar spin spiral becomes simply a matter of scale.

One would intuitively expect that, by virtue of its perturbative nature, the magnetic force theorem would describe well at least the magnetic energetics for configurations close to the reference state. Hence, one is not guaranteed to find good total magnetization energies, which are relative to the nonmagnetic state, but rather good magnetization energy *differences* between the reference state and some arbitrary spin configuration close to it. In the case of the FM-MFT one would expect that states close to the FM state would be well described. Since the DLM state is a profoundly noncollinear one, it might be expected that the DLM-MFT would better treat noncollinear states than the FM-MFT.

Planar spin spirals thus form a useful tool for probing the regions where the FM-MFT and DLM-MFT work well, since as a function of **q** such structures smoothly contain both a ferromagnetic limit as well as noncollinear limits near the Brillouin zone boundary. In Figs. 1 and 2 we present the results of direct calculation of the magnetization energies of planar spin spirals in Ni, fcc Co, Fe, Mn, and Cr where the moment of the spiral is constrained to be that of the ground state moment, and with **q** vectors in the standard reciprocal space paths as indicated. Also shown is the evaluation of Eq. (9) using only bilinear interactions determined via the FM-MFT and DLM-MFT where the reference states were again constrained, if necessary, to have the same moment as the ground state structure. For a unified comparison we show all energies here measured from the nonmagnetic state, hence, all energies are magnetization energies. This is achieved simply by the addition of E_{dlm} to Eq. (9), a point which we shall exploit further in Sec. VI.

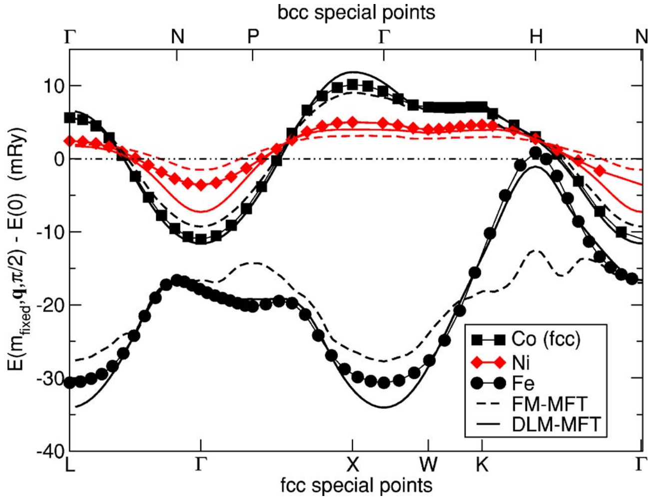

FIG. 1. (Color online) Planar spin spiral energetics for Fe, fcc Co, and Ni directly calculated from KKR-ASA method (filled symbols) and evaluated using magnetic force theorem with ferromagnetic (FM-MFT, dashed lines) and disordered local moment reference states (DLM-MFT, continuous lines).

These results show that the DLM-MFT provides a better description of the energetics of the planar spin spirals with **q** away from the Γ point. On the other hand, it is seen that near the Γ point the *form* of the FM-MFT energy is much closer to the directly calculated results, and remembering that for a "fair" comparison one should measure the energy relative to the FM state in this case, it is clear that the FM-MFT performs better in this limit. For example, spin stiffness constants would be better treated in the FM-MFT, as will be shown in the next section.

A further point of interest is the dramatic difference between the MFT results of the ferromagnets Fe, fcc Co, and Ni. The FM-MFT interactions in the case of Fe are completely unable to reproduce the energetics of the planar spin spirals, whereas for fcc Co both the FM-MFT and DLM-MFT interactions provide a good description for *both* the spirals near the Γ point and those near the Brillouin zone boundary. For Ni the differences are pronounced only at the Γ point.

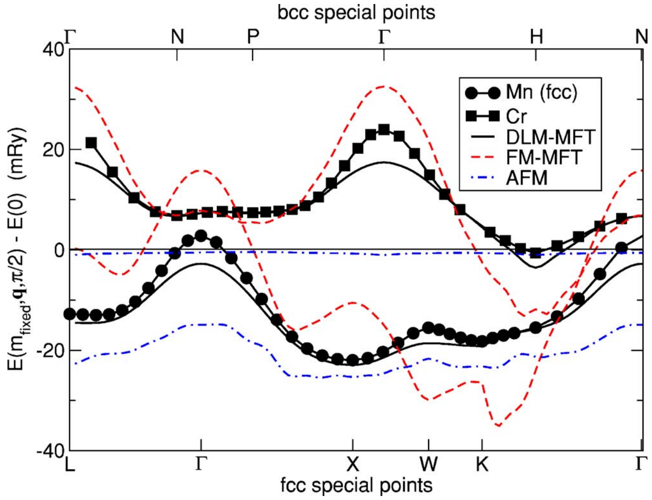

FIG. 2. (Color online) Planar spin spiral energetics for Cr and Mn directly calculated from KKR-ASA method (filled symbols) and evaluated using magnetic force theorem with ferromagnetic (FM-MFT, dashed lines), antiferromagnetic (AFM-MFT, dot-dashed lines), and disordered local moment (DLM-MFT, continuous lines) reference states.

TABLE I. j_0 (in mRy) for $3d$ magnets calculated from the bilinear interactions derived using the magnetic force theorem with either FM or DLM reference states. Also shown is the value calculated directly using the KKR-ASA method.

	DLM-MFT	FM-MFT	KKR-ASA
Ni	3.71	9.50	5.60
Co (fcc)	15.51	15.50	15.09
Fe	15.58	9.26	12.13

One possible explanation for this behaviour is the neglect of higher order terms in Eq. (9). To check this for the DLM-MFT case we have converged the Heisenberg expansion using Eq. (7) and have found that there is no significant contribution from such terms. However, it may still be the case that higher order terms are necessary for convergence of the Heisenberg expansion when using the FM-MFT. A possible reason for this may be found in the asymptotic behavior of the interactions in the FM-MFT and DLM-MFT. In the former case for weak ferromagnets (in the Stoner sense) such as Fe with a large density of states at the Fermi level in both the minority and majority channels the asymptotic behaviour is governed by a long range RKKY decay.[13] This may also lead to important multisite interactions. On the other hand, for strong ferromagnets such as Ni and fcc Co, the asymptotic behavior of the interactions will show an exponential decay, and this shorter range of interaction would likely lead to a reduced importance of multisite interactions. It should be noted that the DLM-MFT interactions will *always* show an exponential decay due to the disorder of the reference state.

In Fig. 2 it can be seen that for the antiferromagnets Cr and Mn the FM-MFT and, more surprisingly, results using an antiferromagnetic reference state are in quite poor agreement with direct calculation. However, for both materials the DLM-MFT is seen to provide a good description of the energetics.

It is interesting to observe that in all cases the FM-MFT and DLM-MFT provide a bound on the magnetization energy at the Γ point: The FM-MFT consistently underestimates the stability of the FM state whilst the DLM-MFT consistently overestimates it.

The quantity $j_0 = \Sigma_j J_{0j}^{(2)}$ is equal to the difference in energy between the DLM and FM states and thus should be a difficult quantity to predict accurately using perturbative approaches based on *either* the DLM or FM reference states. This is indeed seen in Table I, where for Ni and Fe the values from the DLM-MFT and FM-MFT differ quite considerably from each other and also from the directly calculated result.

We now turn to the calculation of small θ 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

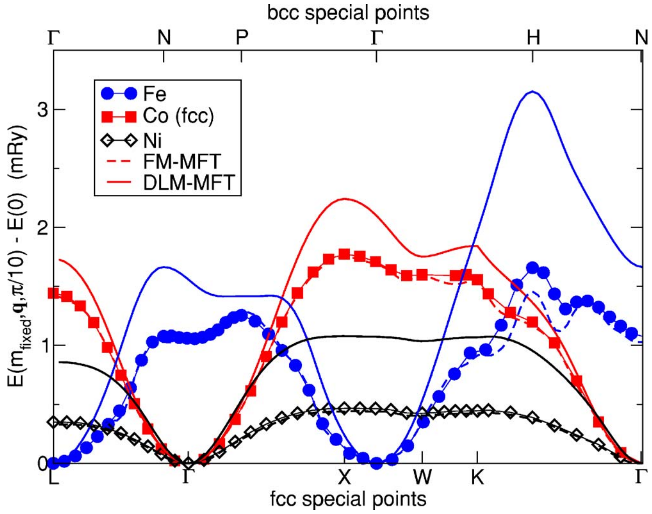

FIG. 3. (Color online) Small θ spin spiral energetics for Fe, fcc Co, and Ni directly calculated from KKR-ASA method (filled symbols) and evaluated using the magnetic force theorem with ferromagnetic (FM-MFT, dashed lines) and disordered local moment (DLM-MFT, continuous lines) reference states.

this case the energy is measured relative to the FM energy. In fact, for Ni and fcc 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. Near the *H* point in Fe are clearly seen effects of the long ranged RKKY nature of the magnetic interactions in Fe, known as Kohn anomalies. These of course are completely absent in the DLM-MFT spectrum due to the exponential decay of the interactions in that case. From Fig. 3 it is further seen that the DLM-MFT overestimates small θ 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 fcc Co the difference between the FM-MFT and the DLM-MFT results is rather small.

From a linearization of the Bloch equation of motion the energetics of small θ 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.*[14] who find the correction to the FM-MFT provided recently by Bruno[15] 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.

It is interesting at this point to return to the question of why there is such a profound difference in the description of spin spiral energetics provided by the FM and DLM magnetic force theorem interactions for the case of Fe and Ni, but not in the case of fcc Co. Since each reference state provides a good description of, respectively, the small θ and planar spin spiral limits, another explanation for this behaviour sugests itself.

One notes that in the FM state majority and minority bands are orthogonal and may cross each other, whereas in noncollinear states these bands are no longer orthogonal and will, to a greater or lesser degree, repel each other creating hybridization gaps and a change in the electronic structure. Such hybridization may be expected to be important in the case of weak ferromagnets, where there is a large density of states (DOS) in both the spin up and spin down channels at the Fermi level E_F, and not so important in the case of strong ferromagnets, where the spin up channel will a low DOS at E_F. Clearly, this effect can only be accounted for in the perturbative approach by its *inclusion in the reference state*. One can then view the DLM and FM as representing strong hybridisation and no hybridisation limits for the reference state.

So in the case of weak ferromagnets highly noncollinear states such planer spin spirals will be far more accurately treated in interactions taken from the DLM reference state than those from the FM reference state, whereas for nearly collinear magnetic configurations such as small θ spin spirals the situation will be the opposite. On the other hand, for strong ferromagnets with weak hybridization effects upon changes in the spin configuration, both reference states should provide a good description. Thus one can naturally understand relative importance of the reference state in the case of Fe and fcc Co.

For the case of Ni another explanation must be found, but one readily suggests itself. In this case a difference with Fe and fcc Co is that Ni is a rather itinerant ferromagnet with a large magnetization density in the interstitial region. The electronic structure would be expected to be much more sensitive to changes in the spin configuration that in the case of very good local moment magnets. In the next section a physical manifestation of these two effects will be discussed.

V. CURIE TEMPERATURE AND SPIN STIFFNESS CALCULATIONS

At second order magnetic transitions, where the long range order parameter goes to zero at the critical temperature T_C, the spin configurations of relevance will be much closer to the DLM state than any ground state magnetic structure. The appropriate reference state for extracting interactions is then the DLM state. In the light of the above discussion, it is thus interesting to investigate the different Curie temperatures one obtains for Fe, fcc Co, and Ni using the FM and DLM reference states. The interactions derived may be used in Monte Carlo calculations to determine the magnetic transition temperature. Results of this procedure are shown in Table II.

The arguments given above explain the result for Fe found in Ref. 5, 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 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 fcc Co being close in both approaches but there existing a large difference for Ni and Fe. The effect of the reference state amounts to 500 K in each of these cases. One notes that a large body of work exists where Curie temperatures have been calculated via the FM-MFT for half metallic systems, such as dilute magnetic

TABLE II. Transition temperatures (in K) for $3d$ magnets calculated using the Monte Carlo method and experimental transition temperatures.

	DLM-MFT	FM-MFT	Exp.
Ni	820	320	624–631
Co (fcc)	1350	1120	1388–1393
Fe	1190	550	1044–1045
Mn	450	-	
Cr	421		321

semiconductors. However, these materials are expected to be a case where neither hybridization or itinerancy effects will be important, and so the use of the FM-MFT or DLM-MFT should make little difference.

It is extremely interesting that the Curie temperature for Ni comes out to be 820 K which is *higher* than the experiment. 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 necessary 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 cannot simply be allowed to be a free parameter, as works well for Fe.[5] A way in which progress can be made is described in the next section.

Near the Γ 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

$$D = \frac{2}{3m}\sum_j J_{0j}\mathbf{R}_{0j}^2, \tag{10}$$

where m is the magnetic moment of the ferromagnetic state. Results for the spin stiffness of the ferromagnetic $3d$ metals are shown in Table III. The FM-MFT provides values in reasonable agreement with experiment, as expected, however the DLM-MFT also does for fcc Co and Fe. For fcc Co this may be expected from the arguments above, however, for Fe it is more surprising since there are 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 evaluated from the derivative of the magnon spectra at the Γ point directly. For the DLM-MFT interactions Eq. (10) may be used due to their much quicker decay in real space.

TABLE III. Spin stiffness constants (in mRy Å^2) for Fe, fcc Co, and Ni directly calculated from KKR-ASA method and evaluated using magnetic force theorem with FM and DLM reference states.

	DLM-MFT	FM-MFT	Exp.
Ni	1796	541	555[a], 420[b]
Co (fcc)	520	480	580[a], 510[b]
Fe	313	322	280[c], 330[a]

[a] See Refs. 16 and 17.

[b] See Ref. 18.

[c] See Ref. 19.

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.[4,20] The crucial step is the addition of an on-site term to the energy expression, which for our purposes can be written as

$$E(\{\mathbf{s}_i\},\{m_i\}) = \sum_i J_i^{(1)}(\{m_i\}) - \sum_{ij} J_{ij}^{(2)}(\{\mathbf{s}_i\},\{m_i\})\mathbf{s}_i \cdot \mathbf{s}_j. \tag{11}$$

The inclusion of this on-site term then brings the energetics of the coupling between the local intrasite exchange and the nonlocal intersite exchange into the Hamiltonian. In Ref. 4 the coefficients of Eq. (11) were found by deploying the structure inverse method using as the basis a set of planar spin spirals with different constrained moments. However, what we wish to point out here is that the coefficients of this equation may be found using the perturbative scheme discussed so far in this paper.

The key observation is that if the energy of the DLM state of fixed spin moment m is calculated from Eq. (11), the vanishing of the spin product results in the expression

$$E_{dlm}(m^2) = \sum_i J^{(1)}(m^2), \tag{12}$$

here the dependence is on m^2 and not m due to the $m=-m$ symmetry of the DLM state. Thus if the DLM state is used as a reference state then the m dependence of $J^{(1)}$ is given by the m dependence of the DLM state itself. Further, an inspection of the equation for $J_{ij}^{(2)}$ in the DLM state shows that it is very easily generalised to the case where one embeds two spins of moments m_i and m_j in a DLM state composed of random local moments of size $\bar{m}$. This may be achieved by the use of the CPA impurity formula. Equation (6) may now be written schematically as

$$J_{ij}^{(2)}(\bar{m},m_i,m_j) = \frac{1}{4\pi}\mathrm{Im}\int^{E_F} dz\, \mathrm{Tr}_L\{\Delta t_i(\bar{m},m_i)\tilde{g}_{ij}(\bar{m}) \times \Delta t_j(\bar{m},m_j)\tilde{g}_{ji}(\bar{m})\}. \tag{13}$$

One has now obtained expressions for both the intrasite and intersite parts of Eq. (11) which can now be written as

TABLE IV. Magnetic moments (in μ_B) of ground state structures of $3d$ magnets calculated directly from KKR-ASA and from magnetic force theorem.

	Model Hamiltonian	KKR-ASA
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

$$E = \sum_i J^{(1)}_{DLM}(m_i^2) - \sum_{ij} J^{(2)}_{ij}(\bar{m},m_i,m_j)\mathbf{s}_i \cdot \mathbf{s}_j. \tag{14}$$

Thus including the magnetization energy of the DLM reference state as well as the exchange integrals calculated from that state, allows for the inclusion of longitudinal fluctuations in approaches based on the magnetic force theorem.

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. Equation (14) should then reproduce the *ab initio* equilibrium moments and relaxation energies. For practical calculations one must parameterise the quantities $J^{(1)}_{DLM}$ and $J^{(2)}_{ij}$. In the case of homogeneous structures such as spin spirals all spins have the same moment size and so this is easily done by fitting polynomials to values calculated for several different m. We find that the best fitting is achieved with polynomials between order 6 and 10. Since the theory is based on the DLM as the reference state it is interesting to see initially how well it works in the FM limit, which should be the worst case. Using Eq. (14) one may calculate the equilibrium moments of the ground state structures, which are either ferromagnetic or antiferromagnetic. These results are presented in Table IV. As expected the fcc 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 from the discussion in previous sections of this paper, are Fe and Cr.

In Fig. 4 are shown the equilibrium moments of the planar spin spirals. As can be seen the agreement between *ab initio* results and Eq. (14) 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). Again the agreement is seen to be very reasonable, though slightly worse for Fe, in particular on the linear path between the N and Γ special points. However, this is to be expected in the light of the discussion in Sec. IV. Since Fe is a weak ferromagnet, hybridization effects will be important in the noncollinear state and may be expected to differ according to the particular noncollinear state considered, i.e., they will change with the $\mathbf{q}$ vector of the planar spin spiral. Perturbative approaches based on a particular reference state will neglect these $\mathbf{q}$ dependent hybridization effects completely, leading to the greater errors for Fe in Fig. 5.

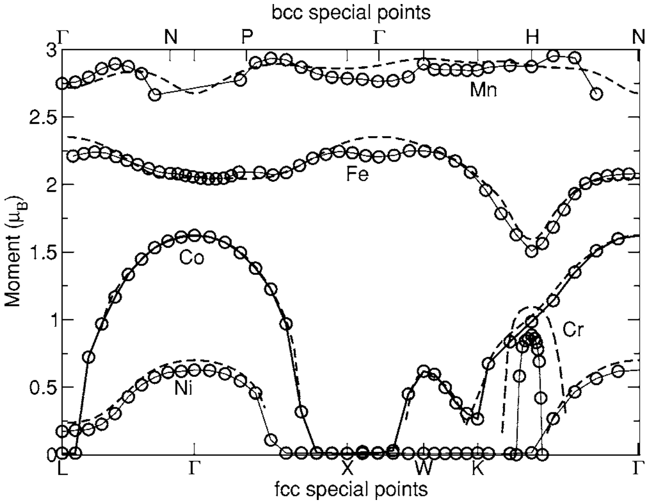

FIG. 4. Relaxation of the magnetic moment of planar spin spirals in Ni, fcc Co, Fe, Mn, and Cr. Shown are both the results of direct calculation via KKR-ASA method (open circles) and evaluation from the m-dependent magnetic force theorem approach.

It must be stressed that the results obtained are based only on the DLM reference state, but nevertheless this allows one to capture the energetics of the *ferromagnetic state*, as well as planar spin spirals. The method thus seems to include within it an accurate enough treatment of the m and orientational dependence of the magnetization energy to calculate the equlibrium moments of a variety of structures. Using this energy expression within the classical partition function one would then expect to be able to calculate a Curie temperature with the moment responsive to the orientational disorder. Further work along these lines is now in progress, in particular the relation of our work to spin fluctuation theory,[20] and the role of short range order in the paramagnetic state.

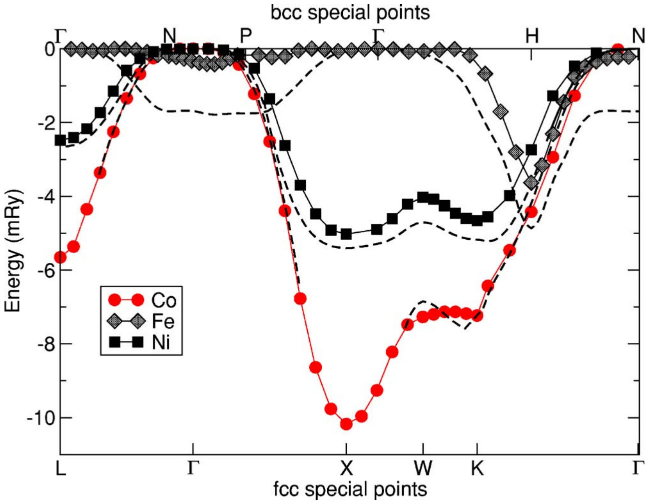

FIG. 5. (Color online) Relaxation energy of planar spin spirals in Ni, fcc Co, and Fe. Shown are both the results of direct calculation via KKR-ASA method (open and filled symbols) and evaluation from the m-dependent magnetic force theorem approach (dashed lines).

VII. CONCLUSIONS

We have presented a study of the effect of the choice of 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 for planar spin spiral calculations and Curie temperatures, but that for strong local moment ferromagnets both approaches may be used interchangeably. We find that the FM-MFT provides an excellent description of the energetics of small θ spin spirals, and hence, magnon spectra. We have further proposed a method whereby *longitudinal* and well as transverse fluctuations may be incorporated into methods based on the magnetic force theorem. We have demonstrated the accuracy of this latter method by comparison with *ab initio* results.

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[1] F. Ducastelle, *Order and Phase Stability in Alloys* (North-Holland, Amsterdam, 1991).

[2] B. L. Gyorffy, A. J. Pindor, J. Staunton, G. M. Stocks, and H. Winter, J. Phys. F: Met. Phys. **15**, 1337 (1985).

[3] S. V. Halilov, H. Eschrig, A. Y. Perlov, and P. M. Oppeneer, Phys. Rev. B **58**, 293 (1998).

[4] N. M. Rosengaard and B. Johansson, Phys. Rev. B **55**, 14975 (1997).

[5] A. V. Ruban, S. Shallcross, S. I. Simak, and H. L. Skriver, Phys. Rev. B **70**, 125115 (2004).

[6] A. I. Lichtenstein, M. I. Katsnelson, and V. A. Gubanov, J. Phys. F: Met. Phys. **14**, L125 (1984).

[7] A. I. Lichtenstein, M. I. Katsnelson, V. P. Antropov, and V. A. Gubanov, J. Magn. Magn. Mater. **67**, 65 (1987).

[8] T. Oguchi, K. Terakura, and H. Hamada, J. Phys. F: Met. Phys. **13**, 145 (1983).

[9] P. Bruno, J. Kudrnovský, V. Drchal, and I. Turek, Phys. Rev. Lett. **76**, 4254 (1996).

[10] J. A. Blanco and V. M. P. Pidal, Eur. J. Phys. **16**, 195 (1995).

[11] J. P. Perdew and Y. Wang, Phys. Rev. B **45**, 13244 (1992).

[12] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996).

[13] M. Pajda, J. Kudrnovský, I. Turek, V. Drchal, and P. Bruno, Phys. Rev. B **64**, 174402 (2001).

[14] M. I. Katsnelson and A. I. Lichtenstein, J. Phys.: Condens. Matter **16**, 7439 (2004).

[15] P. Bruno, Phys. Rev. Lett. **90**, 087205 (2003).

[16] R. Pauthenet, J. Appl. Phys. **53**, 2029 (1982).

[17] R. Pauthenet, J. Appl. Phys. **53**, 8187 (1982).

[18] G. Shirane, V. J. Minkiewicz, and R. Nathans, J. Appl. Phys. **39**, 383 (1968).

[19] H. A. Mook, J. W. Lynn, and M. R. Nicklow, Phys. Rev. Lett. **30**, 556 (1973).

[20] M. Uhl and J. Kübler, Phys. Rev. Lett. **77**, 334 (1995).