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ENERGY AND ENTROPY IN ITINERANT MAGNETS

J. H. Samson

Department of Physics, University of Technology, Loughborough, Leics LE11 3TU, G.B.

Abstract. - This paper describes a method for calculating short-range magnetic order in itinerant magnets. An energy and an entropy are assigned to configurations of the magnetization. The energy comes from the electronic structure and the entropy from the number of configurations satisfying constraints on the short-range order.

1. Introduction

A major question in itinerant magnetism concerns the degree of magnetic short-range order (SRO) above \( T_c \) in the magnetic transition metals. To what extent does the itinerancy of the d electrons lead to correlations of longer range than in magnetic insulators, as some neutron scattering data suggest? Most workers have assumed either giant SRO, of sufficiently long range to support a local bandstructure, or local moments, disordered on an atomic scale, that can be treated by methods such as the coherent potential approximation. What is needed is a means of calculating the SRO.

The proposed method consists of two stages. Firstly, we derive an effective classical spin Hamiltonian for the magnetic excitations \[1\]. This is calculated by electronic structure techniques. Secondly, we assign an "entropy" to each of these configurations. By minimizing the resulting "free energy", we calculate both the order parameter and a measure of the SRO.

If the range of the effective exchange interactions in the Hamiltonian increases with decreasing temperature (due to quantum fluctuations) the SRO will similarly be large. In this paper we concentrate on the second stage, the variational calculation of the SRO for a given set of calculated energies.

In mean field theory (MFT) one takes the energy \( E \) to be a function of an order parameter \( X \), and calculates the free energy \( F(X) = E(X) - TS(X) \) by associating an entropy \( S(X) \) to each value of \( X \). This is given by the logarithm of the number of configurations satisfying a constraint given by the order parameter. The free energy and order parameter are then found by minimizing \( F(X) \). The approximation is exact if the energy is a function of the order parameter only. It is unsatisfactory for the present Purpose, where we are interested in the SRO. We therefore take an energy that depends on both the long-range order parameter and a short-range order parameter, such as the nearest neighbour correlation function. By minimizing the free energy with respect to both of these parameters one can determine the SRO. Similar approaches have been used elsewhere with a different approximation for the entropy \[2\], and for certain other assumptions about the form of the Hamiltonian \[3\].

2. Method

The itinerant magnets are modelled by an \( N \)-band Hubbard model. One can derive an effective Hamiltonian of the form \( H(\{\Delta_i\}) \) by the functional integral method. Here \( \{\Delta_i\} \) is the exchange field, a three-component vector on each site \( i \); the statistics of this field determine the statistics of the magnetization. Quantum effects can be included through a \( 1/N \) expansion \[1\]. The Hamiltonian has been evaluated for various sets of configurations by electronic structure techniques \[4, 5\]. Suitable sets of configurations include frozen spin waves (SW) of varying amplitude and wavelength, in which all atoms are equivalent, and random configurations taken from an ensemble with a given correlation function. The SW are given by

\[
\Delta_i = (\Delta \sin \theta \cos (Q \cdot r_i), \Delta \sin (Q \cdot r_i), \Delta \cos \theta),
\]

which describes a spin wave of wavevector \( Q \), with a conical arrangement of exchange splittings of magnitudes \( \Delta \) at an angle \( \theta \) to the \( z \) axis. Thus the amplitude of the exchange field, order parameter \( (\Delta \cos \theta) \) and SRO are specified. For any arbitrary configuration \( \{\Delta_i\} \) we shall assign a SW with the same values of those parameters, as defined below.

We fit the Hamiltonian to a trial Hamiltonian that is a function of a small number of macroscopic parameters \( X_i \),

\[
H(\Delta) \approx H_0(X),
\]

for example by computing the energy for a suitable set of SW. Here \( \Delta \) represents the exchange field \( \{\Delta_i\} \) and \( X \) the parameters \( \{X_i\} \). The parameters spanned by the SW, expressed in terms of the parameters of equation (1), are

\[
X_i = \frac{1}{N} \sum \Delta_{i\alpha} = \Delta \cos \theta
\]
where $c$ is a nearest-neighbour correlation function, $N$ is the number of atoms, $z$ is the coordination number and the sum in (5) is over nearest-neighbour pairs. More generally, these parameters could represent arbitrary constraints on the exchange field, $f_n(\{\Delta_i\}) = X_n$, such as a sum of the form $\sum J_{ij} \Delta_i \Delta_j$, with $J_{ij}$ a set of exchange integrals for a Heisenberg fit to the Hamiltonian. Typically these might extend as far as third or fourth nearest neighbours [4]. It should be stressed that the Hamiltonian $H_0(X)$ can be any function of the parameters and need not be well approximated by a Heisenberg model.

The partition function is then

$$Z = \int d\Delta e^{-\beta H(\Delta)}$$

$$= \int dX \int d\Delta e^{-\beta H(\Delta)} \delta(X - f(\Delta))$$

$$= \int dX e^{-\beta[H_0(X) - TS(X)]}. \quad (6)$$

Here the entropy $S(X)$ is derived from the area of phase space subject to these constraints,

$$S(X) = k \ln \int d\Delta \delta(X - f(\Delta)). \quad (7)$$

This integral can be evaluated exactly, provided that the constraints are all linear or quadratic in the fields, as are the constraints (3-5). For those constraints the entropy per atom is

$$S(\Delta, \theta, c)/N = 3k \ln (2\pi e/3)/2 +$$

$$+ 3k \ln \Delta + 3k \ln \sin \theta +$$

$$- 3k (\ln G(s) + K(s))/2 \quad (8)$$

where $G$ is the lattice Green function and $K = \int G ds$, with the value of $s$ given implicitly in terms of $c$ by $c = s - 1/G(s)$. This is equal to the entropy in the spherical model [6] for the same value of the nearest neighbour correlation. The present approximation, if applied to the Heisenberg model, reduces to the spherical model; however, the method can be used with an arbitrary Hamiltonian or an arbitrary dependence of the energy on the parameters. The correlation function is determined if the nearest neighbour correlation function is known (with the parametrization of Eq. (5)), and is identical to that in the spherical model. The temperature dependence of the correlation function, however, depends on the form of the trial Hamiltonian and differs from that of the spherical model. At $T_c$, the correlation function is the same in both models. We then find the free energy $F$ by minimizing $F(X) = H_0(X) - TS(X)$ with respect to the parameters. This gives the order parameter and the SRO, as well as other thermodynamic quantities.

3. Results and discussion

A preliminary calculation on iron gives $T_c = 800$ K, compared with an experimental value of 1043 K, and a nearest neighbour correlation function at $T_c$ of 0.28 ($= \cos 74^\circ$). The energies are taken from calculations [5] of the energy of "spin spirals" (SW with $\theta = \pi/2$) that use a full spd tight-binding Hamiltonian without quantum fluctuations; the entropies are taken from a trial Hamiltonian with nearest neighbour interactions. This result should not be seen as proof of the absence of significant SRO. In a nearest neighbour model, the nearest neighbour correlation will be small at $T_c$ and significant SRO, such as a long tail in the correlation function, cannot exist in the paramagnetic state. Longer-range interactions are needed to obtain SRO. It is speculated that at low temperatures quantum fluctuations act to increase the range of the effective exchange interactions. This is currently being examined in a simple one-dimensional model.