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# Australian Journal of Physics

Volume 50, 1997  
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## Ab Initio Theory of Magnetic Correlations in Itinerant Magnets\*

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### Abstract

One of the challenging problems in modern magnetism is the description of the paramagnetic state because at finite temperatures the effect of spin fluctuation plays a fundamental role in altering the electronic structure. In magnetic metals, the magnetic properties are linked to the underlying spin-polarised electronic structure in a highly complex manner. In recent years, *first-principles* electronic structure theories based on the spin density functional (SDF) theory have been able to provide a quantitative account of many ground-state magnetic properties. In this article, the effect of spin fluctuation on the spin-polarised electronic structure and thus the magnetic properties will be discussed and its incorporation into ab initio calculations explained. If the magnetic and compositional ordering energies of magnetic metallic alloys are close, the compositional and magnetic correlations can profoundly influence each other. A theoretical formalism for treating these correlations within the framework of a *first-principles* electronic structure theory will be elaborated and the application to a number of magnetic alloys discussed and compared with experiments.

### 1. Introduction

One of the fundamental problems in modern magnetism concerns the nature of finite-temperature magnetic correlations in transition metals and their alloys in which the magnetic correlations are set up by itinerant electrons. The theoretical challenge is to take into account the full many-electron interactions, incorporating the magnetic and translational degrees of freedom of the electrons responsible for the magnetism, via the underlying electronic structure. In the case of alloys, the problem is further complicated by the fact that magnetic and compositional correlations are intricately interrelated via their ‘electronic glue’. The problem of dealing with the many-electron interactions in the ground state has been largely solved with the advent of the density functional theory (DFT) of Hohenberg, Kohn and Sham (Hohenberg and Kohn 1964; Kohn and Sham 1965) and, in particular, its spin-dependent extension (von Barth and Hedin 1972; Rajagopal and Callaway 1973). Together with fast supercomputers and efficient electronic structure methods, it has become possible to provide a reliable theoretical description of the ground state and equilibrium properties of many itinerant magnetic metals and alloys. Indeed, the local spin density

\* Refereed paper based on a contribution to the Sixth Gordon Godfrey Workshop on Recent Developments in Magnetism and Magnetic Materials, held at the University of New South Wales, Sydney, in October 1996.

approximation (LSDA) (von Barth and Hedin 1972; Gunnarsson and Lundqvist 1976; Ceperley and Alder 1980; Vosko *et al.* 1980) remains the only effective means for incorporating the many-electron effects into ab initio calculations of the electronic and magnetic structures of these materials. In the sections below, we will first give the theoretical background to the problem of itinerant magnetism and some of the approaches to solving it. Next, we present in detail one of the *first-principles* theory of itinerant magnets based on the LSDA, capable of predicting the magnetic correlations in elemental magnets as well as the compositional correlations in metallic magnetic alloys. Finally, the application of this theory to calculating the magnetic and compositional correlations of a range of magnetic alloys will be discussed and the results compared with experiment.

## 2. Theory

The basic idea behind the density functional theory is that the ground-state energy of a many-body system is a unique functional of the particle density  $n(\mathbf{r})$  and is a minimum when evaluated for the true ground-state density. The theory further shows that the relevant one-particle effective potential can, in principle, contain all the effects of electron correlations, although in practice some approximations must be made. Formally, the local spin density approximation to the exchange-correlation functional  $E_{xc}$  can be expressed as

$$E_{xc}[n, m] \approx \int d\mathbf{r} \, n(\mathbf{r}) \epsilon_{xc}\{n(\mathbf{r}), m(\mathbf{r})\} \quad (1)$$

in terms of the charge and magnetisation densities, given by

$$n(\mathbf{r}) = \sum_i^{\text{occ}} \text{Tr}(\psi_i^*(\mathbf{r}) \psi_i(\mathbf{r})), \quad m(\mathbf{r}) = \sum_i^{\text{occ}} \text{Tr}(\psi_i^*(\mathbf{r}) \tilde{\psi}_i(\mathbf{r})),$$

where  $\tilde{\sigma}$  are the usual Pauli matrices, and  $\psi_i$  are the wave functions obeying the Schrödinger–Pauli equation  $(-\nabla^2 + \tilde{V}^{\text{eff}}[n, m]) \psi_i(\mathbf{r}) = \epsilon_i \psi_i(\mathbf{r})$ , and the effective potential operator is given by

$$\begin{aligned} \tilde{V}^{\text{eff}}[n, m] = & \left( V^{\text{ext}}(\mathbf{r}) + e^2 \int d\mathbf{r}' \frac{n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + \frac{\delta E_{xc}[n, m]}{\delta n(\mathbf{r})} \right) \tilde{1} \\ & - \left( B^{\text{ext}}(\mathbf{r}) - \frac{\delta E_{xc}[n, m]}{\delta m(\mathbf{r})} \right) \tilde{\sigma}. \end{aligned} \quad (2)$$

These one-electron Kohn–Sham equations can then be solved using electronic structure techniques to explain a variety of properties of many magnetic materials.

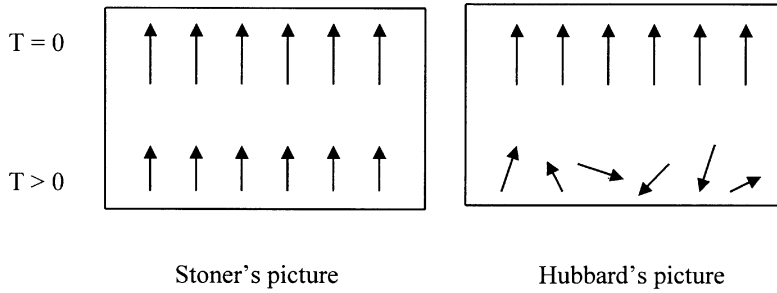
The first successful attempt at understanding itinerant magnetism is Stoner’s theory of ferromagnetism. Briefly, in the Stoner picture the magnetic excitations are the spin-flip excitations of the electrons across the Fermi surface. Mathematically and in the random phase approximation (RPA), the Stoner theory of magnetic susceptibility is given by

$$\chi(q, \omega) = \frac{\chi_0(q, \omega)}{1 - I\chi_0(q, \omega)}, \quad (3)$$

where  $\mathbf{q}$  is the wave-vector,  $\omega$  the frequency, and the Stoner parameter  $I = U/N_{\text{at}}$  is the ratio of the Coulomb interaction energy  $U$  and the total number of atoms  $N_{\text{at}}$ . Here  $\chi(q, \omega)$  is also known as the enhanced Stoner susceptibility whereas  $\chi_0$  is the ‘bare’ or unenhanced susceptibility given by

$$\chi_0(q, \omega) = \sum_k \frac{n_{k\uparrow} - n_{(k+q)\downarrow}}{\hbar\omega - [E_{k\uparrow} - E_{(k+q)\downarrow}]} \quad (4)$$

The Stoner condition for ferromagnetism is satisfied if  $In(E_F) \geq 1$ . Strictly speaking, the Stoner picture of ferromagnetism is valid only for a system in the ground state at  $T = 0$  because it assumes a translational invariance for the orientations of the local moments, i.e. a ‘global’ exchange-splitting of the electronic structure is assumed. A direct application of Stoner’s theory to calculate the thermodynamic properties of transition metals such as the Curie temperatures of Ni and Fe has resulted in values an order of magnitude higher than the experimental ones. The root of the problem is that, at finite temperatures, the translational invariance is destroyed by thermal excitations and consequently the effects of spin fluctuations must be taken into account, both in orientation and magnitude (see Fig. 1). In other words, we now have to think in terms of ‘local’ instead of ‘global’ exchange-splitting. This brings us to the central issue of this paper, namely how to deal with the spin fluctuations in the paramagnetic phase.



**Fig. 1.** The Stoner and Hubbard representations of ferromagnetic excitations.

Over the past decade or so, the problem of the nature of the paramagnetic state of itinerant magnetic metals has been solved to some extent, at least in principle (Moriya 1973). A picture of itinerant electrons moving in the fields set up by spin fluctuations whose orientational degrees of freedom slowly vary has proved useful. The basic assumption is that, on a time scale  $t$  long compared to an electronic ‘hopping’ time ( $\hbar/W \approx 10^{-15}$  s, where  $W$  is a relevant bandwidth), but short when compared with an appropriate spin fluctuation time ( $\hbar/\omega_{\text{SF}} \approx 10^{-12}$  s, where  $\omega_{\text{SF}}$  represents a typical spin fluctuation frequency), the spins of the electrons are sufficiently correlated to leave the magnetisation on a site non-zero. The magnitude of average magnetisation on a site  $k$  is called the local moment,  $\mu_k = \mu_k(\{\hat{e}_i\})$ , where the set of unit vectors,  $\{\hat{e}_i\}$ , picks out the orientational configurations. While this basic scenario is generally accepted to be able to provide an adequate description of many metal magnets at high temperatures, the severity of the neglect of the dynamic effects of the spin fluctuations is

not known and there is also the longstanding controversial question of which configurational orientations of the moments are most important (Murata and Doniach 1972; Moriya and Kawabata 1973; Lonzarich and Taillefer 1985).

The various approaches can be roughly divided into two. Firstly, there is the picture of the ‘fluctuating local band’ (FLB) theory (Korenman *et al.* 1977) of a large amount of short range magnetic order even in the paramagnetic state. This consists of a large amount of spatial region in which the local moments are nearly aligned, i.e. where the orientations vary slowly. In this region the Stoner theory can be applied and perturbations to it made. In the FLB model, it is difficult to carry out first-principles calculations in which both the magnetic and electronic structures are mutually consistent and consequently to examine the full implications of the model and to improve it systematically.

The second type of approach is the ‘disordered local moments’ (DLM) theory (Hubbard 1979; Hasegawa 1979; Edwards 1984) in which the local moments are supposed to fluctuate fairly independently in the high temperature paramagnetic state, but as the temperature is lowered correlations among the local moments begin to develop and the homogeneously disordered local moments (i.e. paramagnetic) system become unstable towards a magnetically ordered phase. The greatest advantage of this approach is that it allows *first-principles* calculations to be implemented based on a generalisation of the LSD theory such that the magnetic and electronic structures can be treated in a mutually consistent manner (Staunton *et al.* 1986; Ling *et al.* 1994).

In magnetic metal alloys compositional and magnetic correlations are linked in a highly complex manner via their underlying electronic structure. Therefore, the magnetic and compositional correlations must be treated on an equal footing from the start, in order to understand the roles they play in the magnetic and compositional structures of these alloys. Here, for the sake of simplicity, we restrict ourselves to binary alloys of the form  $A_cB_{1-c}$  and introduce a site-occupancy label  $\xi_k$ , such that  $\xi_k = 1(0)$  if the site  $k$  is occupied by an A(B) atom. Here the set of unit vectors  $\{\hat{e}_i\}$  is related to the local magnetisation  $M(\mathbf{r}_i, \{\xi_i\})$  by

$$\hat{e}_i = \frac{\int_{V_i} d\mathbf{r}_i M(\mathbf{r}_i, \{\xi_i\})}{\left| \int_{V_i} d\mathbf{r}_i M(\mathbf{r}_i, \{\xi_i\}) \right|}, \quad (5)$$

where  $V_i$  is the volume of the  $i$ th unit cell. The alloy can now be described in terms of a generalised grand potential  $\Omega(\{\xi_k\}, \{\hat{e}\})$ . By generalising the finite-temperature SDF theory (Mermin 1965; Gyorffy *et al.* 1985) one can show formally that  $\Omega(\{\xi_k\}, \{\hat{e}_k\})$  is obtained as a result of a functional minimisation of the grand potential functional

$$\begin{aligned} \Omega[\rho; m] = & \int d\mathbf{r} V^{\text{ext}}(\mathbf{r}, \{\xi_k\}) \rho(\mathbf{r}) + \frac{1}{2} \int \int d\mathbf{r} d\mathbf{r}' \frac{\rho(\mathbf{r}) \rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} \\ & + T_s[\rho, m] - TS_s[\rho, m] + \Omega_{\text{xc}}[\rho, m], \end{aligned} \quad (6)$$

with  $T_s$  and  $S_s$  being respectively the kinetic energy and entropy of a system of non-interacting electrons with densities  $n(\mathbf{r})$  and  $m(\mathbf{r})$  at a temperature  $T$ . The functional minimisation is then carried out using Lagrange multipliers as in

the conventional SDF theory, subject to the constraint that the magnetisation on the  $k$ th site satisfies

$$\int_{V_k} dr_k (m(r_k, \{\hat{e}_k\}) \times \hat{e}_k) = 0.$$

The solution to the Kohn–Sham equations can be obtained in terms of an effective one-electron Green function accessible from the self-consistent-field, Korringa–Kohn–Rostoker, multiple-scattering theory in combination with the coherent potential approximation (SCF–KKR–CPA) (Winter and Stocks 1983; Gyorffy and Stocks 1983). From here we can calculate the charge and magnetisation densities appropriate to each site:

$$n_{i,\alpha}(r_i, \hat{e}_i) = -\frac{1}{\pi} \text{Im} \int d\epsilon f(\epsilon - \nu_e) \text{Tr} \langle G(r_i, r_i; \epsilon) \rangle_{\hat{e}_i, \alpha}, \quad (7)$$

$$m_{i,\alpha}(r_i, \hat{e}_i) = -\frac{1}{\pi} \text{Im} \int d\epsilon f(\epsilon - \nu_e) \hat{e}_i \text{Tr} \langle \sigma \cdot \hat{e}_i G(r_i, r_i; \epsilon) \rangle_{\hat{e}_i, \alpha} = \mu_{i,\alpha}(\hat{e}_i) \hat{e}_i. \quad (8)$$

At temperatures well in excess of any magnetic ordering temperature  $T_C$ , each alloy species is equally likely to occupy a given lattice site  $k$  and the local moment at this site is also equally likely to point in any given direction in space so that the overall magnetisation is zero. Nonetheless, local moments  $\mu_A$  and  $\mu_B$ , and therefore ‘local’ exchange-splitting can exist in this paramagnetic state. As the temperature is lowered, magnetic correlations will set in. By considering the response of the paramagnetic system to a small perturbing magnetic field  $\{\hbar_i\}$ , the magnetic correlation and the static paramagnetic susceptibility  $\chi(q)$  of transition metals and their alloys can be obtained via the response and fluctuation dissipation theorem. After much algebra, it can be shown that in wave-vector space

$$\chi(q) = c\chi_A^\mu(q) + (1-c)\chi_B^\mu(q) + c\chi_A^m(q) + (1-c)\chi_B^m(q). \quad (9)$$

The first two terms describe the magnitude of the local moments’ response to the application of an external field, whereas the third and fourth terms describe how they tend to align with the field. The quantity  $\chi(q)$  can be directly compared with polarised neutron scattering measurements. A maximum of  $\chi(q)$  at  $q = 0$  indicates ferromagnetic ordering, whereas  $\chi(q)$  peaking at finite  $q$  means antiferromagnetic ordering. As an example, we consider the case of an elemental metal, i.e.  $c = 1$ . The components of  $\chi(q)$  in equation (9) can be written explicitly as

$$\chi^m(q, T) = \frac{\frac{1}{3}\beta\bar{\mu}[\bar{\mu} + (J^{\mu\mu}(q) - \Lambda^m)\chi^\mu(q) + \Sigma(q)]}{1 - \frac{1}{3}\beta(J^{\mu\mu}(q) - \bar{\mu}\Lambda^m)}, \quad (10)$$

$$\chi^\mu(q, T) = \frac{[\gamma^{\mu\mu}(q) - \bar{\mu}\Lambda^\mu]\chi^m(q)/\bar{\mu} + \chi^0(q)}{1 - [\gamma^{\mu\mu}(q) - \Lambda^\mu]}, \quad (11)$$

where the quantities  $J^{m\mu}(q)$ ,  $J^{mm}(q)$ ,  $\gamma^{\mu m}(q)$ ,  $\gamma^{\mu\mu}(q)$ ,  $\chi^0(q)$  and  $\Sigma(q)$  are all accessible from the SCF-KKR-CPA calculations performed for the completely disordered, paramagnetic state. The quantities  $\Lambda^m$  and  $\Lambda^\mu$  are the Onsager cavity field corrections which ensure that while the response theory has been derived within a mean-field approach, the important sum rules associated with the diagonal part of the fluctuation-dissipation theorem are satisfied.

For a rigid local moment system, Fe being a fair example,  $\chi(q) \approx \chi^m(q)$  and  $J^{m\mu}(q)$ ,  $\gamma^{\mu m}(q)$ ,  $\gamma^{\mu\mu}(q)$  and  $\chi^0(q)$  are all small. Equation (10) then assumes the form of a classical Heisenberg model within the spherical approximation. On the other hand, in a system such as Ni, where no local moment is set up on the average in the paramagnetic phase,  $\bar{\mu} = 0$  and  $J^{mm}(q)$ ,  $J^{m\mu}(q)$  and  $\gamma^{\mu m}(q)$  also vanish. Thus equation (11) becomes  $\chi(q) = \chi^\mu(q) = \chi^0(q)/[1 - \gamma^{\mu\mu}(q) - \Lambda^\mu]$ . Note that  $\chi(q)$  is now a product of a Stoner exchange-correlation term and a Pauli susceptibility  $\chi^0(q)$ . The susceptibility evidently describes an enhanced Stoner paramagnet where the exchange-correlation effects have been renormalised by the spin fluctuations.

As mentioned earlier, in a metallic alloy the magnetic and compositional correlations must be considered together and treated on an equal basis. While the main focus of this paper is on magnetism in itinerant magnets, it will not be complete without discussing the compositional response function which describes the nature of atomic short range ordering and the role it plays in our understanding of magnetic alloys. In essence, the tendency of a homogeneously disordered alloy to order as the temperature is lowered can be found using concentration-wave theory (Gyorffy and Stocks 1983) and the response and fluctuation-dissipation theorem. The resultant expression has the familiar Ornstein-Zernicke form:

$$\alpha(q) = \frac{\beta c(1-c)}{1 - \beta c(1-c)S^{(2)}(q)}, \quad (12)$$

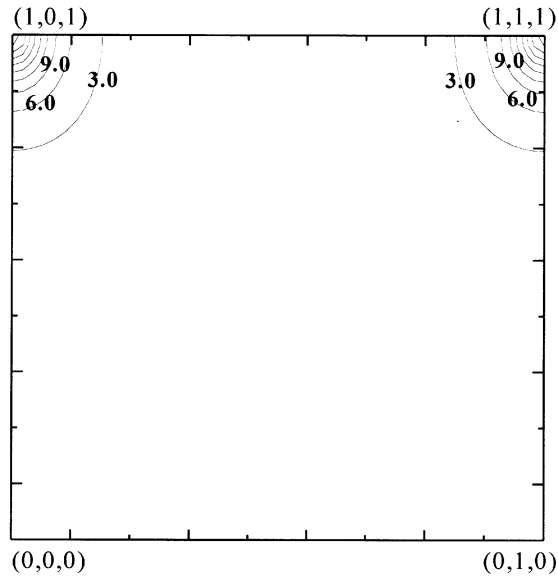
where the ‘interchange’ energy  $S^{(2)}(q)$  is calculated from an exchange-split electronic structure which is either ‘locally’ exchange-split if the system is paramagnetic and ‘globally’ exchange-split if ferromagnetic. Here  $S^{(2)}(q)$  captures the electronic effects such as the filling of the electronic states, the Coulomb interaction and screening, and charge transfer. Also,  $\alpha(q)$  indicates a phase segregation if it peaks at  $q = 0$  and a chemical ordering if its maximum is at finite  $q$ . By considering the behaviours of both  $\chi(q, T)$  and  $\alpha(q, T)$  as a function of  $q$  and  $T$ , we can begin to piece together a consistent picture of the magnetic and compositional structures of a magnetic metal alloy.

### 3. Comparison with Experiment

The disordered local moments theory of itinerant magnets has been successfully applied to several magnetic metallic alloys including  $\text{Cu}_{85}\text{Mn}_{15}$ ,  $\text{Au}_{75}\text{Fe}_{25}$ ,  $\text{Fe}_{87}\text{V}_{13}$  and  $\text{Fe}_{80}\text{Al}_{20}$  (Ling *et al.* 1994, 1995*a*, 1995*b*; Staunton *et al.* 1990, 1997). For the first time, the unusual antiferromagnetic ordering at  $\langle 1, \frac{1}{2}, 0 \rangle$  in  $\text{Cu}_{85}\text{Mn}_{15}$ , whose origin had long challenged experimentalists and theorists alike, was obtained from an ab initio calculation and its origin was explained in terms of a competing

electronic mechanism within the ‘exchange-split’ electronic structure (Ling *et al.* 1994). Some highly unusual compositional correlations (or atomic short-range ordering) in the magnetic alloys  $\text{Au}_{75}\text{Fe}_{25}$ ,  $\text{Fe}_{87}\text{V}_{13}$  and  $\text{Fe}_{80}\text{Al}_{20}$  have also been found from our *first-principles* calculations with very good general agreement with experiment (Ling *et al.* 1995; Staunton *et al.* 1996) and their origins explained. In all three cases, magnetism has been identified to be the driving force for the unusual atomic short-range ordering.

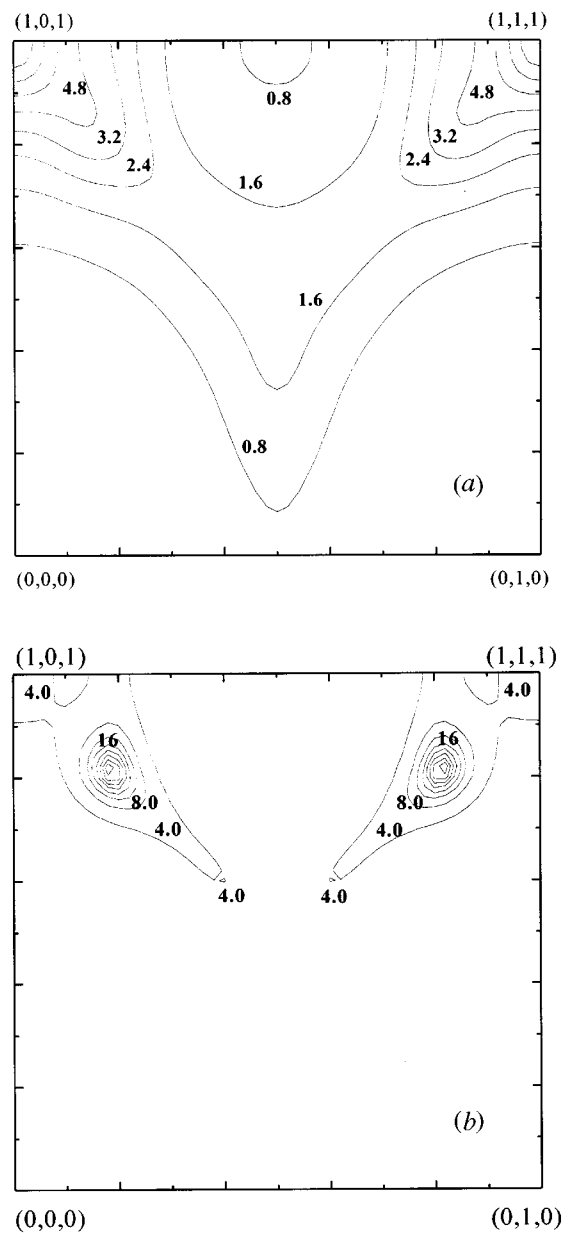
As an example, let us consider the case of  $\text{Fe}_{80}\text{Al}_{20}$  in some detail. *In situ*, unpolarised neutron scattering measurements were carried out by Schweika *et al.* (1990) on a single crystal of  $\text{Fe}_{80}\text{Al}_{20}$  over a range of temperatures of 823 and 1073 K which includes temperatures both below and above the alloy’s Curie temperature of 935 K. In the paramagnetic alloy, they found clear atomic short-range ordering (ASRO) characterised by peaks at  $\mathbf{q} = (1, 0, 0)$  and equivalent positions in wave-vector space. As the temperature is lowered into the ferromagnetic region, the intensity around  $(1, 0, 0)$  equivalent  $q$ -points remains but the intensity is now skewed towards the  $[1, 1, 1]$  direction with a weak subsidiary peak at  $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ . A later paper by Schweika (1990) revealed a second phase transition into an ordered phase characterised by  $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$  at around 650 K. This is consistent with a B32 transient ordered phase observed by Gao and Fultz (1993) in  $\text{Fe}_{75}\text{Al}_{25}$ . The results of Schweika *et al.* were confirmed by Pierron-Bohnes *et al.* (1990, 1991) in the temperature range of 772 and 1573 K in their diffuse x-ray and neutron scattering measurements.



**Fig. 2.** The ASRO  $\alpha^{\text{DLM}}(q, T)$  for the  $(1, \bar{1}, 0)$  plane for paramagnetic (DLM)  $\text{Fe}_{80}\text{Al}_{20}$  at 3000 K in Laue units.

The author and coworkers have calculated the ASRO in both ferromagnetic and DLM (paramagnetic) states of  $\text{Fe}_{80}\text{Al}_{20}$  (Staunton *et al.* 1997). A DLM calculation of  $\chi(q, T)$  indicated strong ferromagnetic correlations in the paramagnetic  $\text{Fe}_{80}\text{Al}_{20}$  and a  $T_C$  of 1130 K, in fair agreement with the experimental value of 937 K.





**Fig. 3.** The ASRO  $\alpha^{\text{FM}}(q, T)$  for the  $(1, \bar{1}, 0)$  plane for ferromagnetic (FM)  $\text{Fe}_{80}\text{Al}_{20}$  at (a) 1000 K and (b) 500 K in Laue units.

This confirms that this treatment of the correlations captures the correct order of magnitude for the energies of the spin fluctuations. The general features of our calculated ASRO of this alloy are best illustrated in Figs 2 and 3. Fig. 2 shows the paramagnetic ASRO,  $\alpha^{\text{DLM}}(q)$ , of  $\text{Fe}_{80}\text{Al}_{20}$  in the high temperature region of 3000 K. Fig. 3 shows the ASRO of  $\text{Fe}_{80}\text{Al}_{20}$  in the ferromagnetic state, i.e.  $\alpha^{\text{DLM}}(q)$ , at (a) 1000 K and (b) 500 K respectively. These results clearly

demonstrate that we have reproduced the general behaviour of the atomic short range order with temperature. Perhaps the most pleasing aspect of this work is that in our calculation the intensity peak at  $\mathbf{q} = (1, 0, 0)$  equivalent points remains at temperatures immediately below the compositional ordering temperature of 1095 K, but with streaking of intensities towards the  $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$  equivalent positions. As the temperature is reduced further, the intensity peak gradually shifts away from the  $(1, 0, 0)$  positions and towards the  $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$  positions. As the temperature is lowered below 600 K, the intensity peak finally settles at  $\mathbf{q} = (0.7, 0.7, 0.7)$ , in reasonably good agreement with the results of Schweika. We have explained this transition in ordering tendency with temperature in terms of the variation in the filling of bonding and antibonding states of the hybridised electronic structure of the alloy as the states around the Fermi level are repopulated with the change of temperature (Staunton *et al.* 1997). Our prediction of an intensity peak at  $\mathbf{q} = (0.7, 0.7, 0.7)$  is believed to be a result of the assumption of a perfect lattice while displacement effects in these alloys are known to be significant. We are in the process of incorporating displacement effects into our theoretical formalism.

#### 4. Conclusion

In conclusion, magnetism in metals is fundamentally linked to their electronic structure and a parameter-free approach to the electronic structure now forms an integral part of our understanding of some of the fundamental problems in magnetism. The author has attempted to demonstrate the need for a *first-principles* description of the electronic structure of magnetic metals and their alloys for a fundamental understanding of the magnetic interactions in these materials and how the magnetic and compositional correlations in alloys are closely interrelated via the underlying electronic structure. Despite over two decades of concerted efforts, our ability to carry out first-principles calculations of magnetic interactions is still rather limited and is usually restricted to relatively simple systems such as 3d transition metals. The correct treatment of spin fluctuations and excitations at finite temperatures still constitutes two of the most difficult and fundamental problems in modern magnetism and much work remains to be done.

#### Acknowledgments

This work is partly supported by the Australian Research Council. The author wishes to thank the organisers of the Gordon Godfrey Workshop for their invitation and hospitality.

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