J. Phys. C: Solid State Phys., **14** (1981) L665–L670. Printed in Great Britain

LETTER TO THE EDITOR

The paramagnetic state of BCC iron

B Sriram Shastry†, D M Edwards and A P Young
Department of Mathematics, Imperial College, London SW7 2BZ, UK

Received 6 March 1981

Abstract. Calculations are reported of the static and dynamic properties of an effective Heisenberg model of Fe with exchange interactions extending to fifth-nearest neighbours. Exchange parameters chosen to fit the spin wave dispersion curve at room temperature lead to little short-range order above T_c and to a neutron scattering function $S(\boldsymbol{q}, \omega)$ which is compatible with Lynn's 'constant ω' plots. The dispersion curve obtained by plotting the positions of peaks in 'constant ω' plots is in excellent agreement with the observed one but it does not correspond to propagating spin waves above T_c. Our results do not, however, reproduce the observed rapid drop of intensity at low frequency in 'constant q' scans.

There has recently been considerable controversy over the nature of the paramagnetic state, above the Curie temperature T_c, of the ferromagnetic transition metals Fe and Ni. Mook *et al* (1973) and Lynn (1975) have proposed, on the basis of their inelastic neutron scattering measurements, that propagating spin-wave modes with wavevectors $q \geqslant 0.2$ Å^{-1} exist at temperatures up to 1.4 T_c in Fe and even higher in Ni. To explain the existence of such modes Prange and Korenman (1979 and references therein) have postulated that a large amount of temperature-independent short-range order exists far above T_c in these metals. Edwards (1980) has pointed out that this is incompatible with specific heat data on Fe. In fact to a first approximation the thermodynamic properties of Fe correspond to the mean-field treatment, with no short-range order above T_c, of a spin 1 Heisenberg model. This may be understood on the basis of the work by Evenson *et al* (1970) and Cyrot (1970), who showed how a system of interacting local moments can emerge from an itinerant-electron picture. The connection between recent developments of this viewpoint by Roth (1978), Hasegawa (1979) and Hubbard (1979) and a mean-field treatment of a Heisenberg model, augmented by smaller terms of an itinerant nature, has been stressed by Edwards (1980). Recent calculations by You *et al* (1980) indicate that in Fe the exchange interaction extends to fourth- or fifth-nearest neighbours, and that the more distant neighbours interact antiferromagnetically. You *et al* (1980) suggest that compensation of this sort, with competing ferromagnetic and antiferromagnetic interactions, may lead to the giant short-range order postulated by Prange and Korenman (1979).

It is surprising that there appear to be no published calculations of the dynamics of the paramagnetic Heisenberg model with interactions beyond nearest neighbours. In this Letter we report such calculations for the first time, together with calculations of the

† Present address: Department of Physics, University of Utah, Salt Lake City, UT 84112, USA.

equilibrium (static) properties. In particular, we consider a BCC lattice with interactions out to fifth-nearest neighbours as is appropriate for Fe. We show the following:

(i) Strongly competing ferromagnetic and antiferromagnetic interactions can lead to giant short-range order but the paramagnetic susceptibility, like the specific heat, is very different from that observed in Fe.

(ii) Exchange parameters chosen to fit the low-temperature spin-wave dispersion curve of Fe lead to satisfactory static properties with little short-range order and to a neutron scattering function $S(\boldsymbol{q}, \omega)$ compatible with Lynn's (1975) 'constant ω' plots. In fact we use parameters appropriate to $Fe_{88}Si_{12}$ on which alloy Lynn's detailed measurements were made. We could not use precisely the parameters of You *et al* (1980) since they lead to negative spin-wave energies for small q at $T = 0$. We cannot, however, explain the rapid drop of intensity at small ω observed in 'constant q' plots (Lynn 1975, 1981).

To evaluate the paramagnetic static properties we use the spherical model with $S = 1$, which is a good approximation to the Heisenberg model except near T_c. Thus writing the Heisenberg model in Fourier form

$$H = -\tfrac{1}{2} \sum_{q} J(\boldsymbol{q}) \boldsymbol{S}(\boldsymbol{q}) \cdot \boldsymbol{S}(-\boldsymbol{q}) \tag{1}$$

we have

$$C(\boldsymbol{q}) \equiv \langle S^z(\boldsymbol{q})\, S^z(-\boldsymbol{q}) \rangle = k_B T [J(0) - J(\boldsymbol{q}) + \chi^{-1}] \tag{2}$$

with the inverse susceptibility χ^{-1} determined by the sum rule

$$\tfrac{1}{3}S(S+1) = N^{-1} \sum_{q} C(\boldsymbol{q}) \tag{3}$$

where N is the number of atoms. The correlation between a spin at the origin and its nth nearest neighbour at $\boldsymbol{R}_n$ is given by

$$\langle S_n^z S_o^z \rangle = N^{-1} \sum_{q} \exp(-i\boldsymbol{q} \cdot \boldsymbol{R}_n) C(\boldsymbol{q}). \tag{4}$$

In figure 1 results are given for the reduced inverse susceptibility $\chi^{-1} S(S+1)/3k_B T_c$ as a function of T/T_c and for the reduced correlation function

$$\Gamma(n) = \langle S_n^z S_0^z \rangle / [\tfrac{1}{3}S(S+1)]$$

at $T = 1.28\, T_c$ for nth-nearest neighbours. The exchange parameters are specified in the caption by α, β and γ which are respectively J_2/J_1, J_3/J_1 and J_5/J_1 where J_n is the exchange parameter for nth nearest neighbours. We set $J_4 = 0$ for convenience since the difficult geometry of the fourth-nearest neighbours leads to algebraic complexity in our subsequent evaluation of the dynamic properties. Case II corresponds to a least-squares fit of Lynn's (1975) room temperature spin wave dispersion curve and case III is a highly compensated situation leading to $\Gamma(1) = 0.82 = \cos 35°$. The giant short-range order postulated by Prange and Korenman (1979) corresponds to $\Gamma(1) = \cos 36°$ (Edwards 1980). The observed susceptibility χ of BCC Fe follows a Curie–Weiss law quite closely with a Curie constant only about 30% larger than the mean field value for the $S = 1$ Heisenberg model. The huge susceptibility of case III is thus in sharp disagreement with experiment. Although we have only presented results for one set of parameters which lead to $\Gamma(1) = \cos 35°$, any Heisenberg model with both strong ferromagnetic correlations out to a few atomic distances and a ferromagnetic ground state has a greatly enhanced susceptibility above T_c. This is

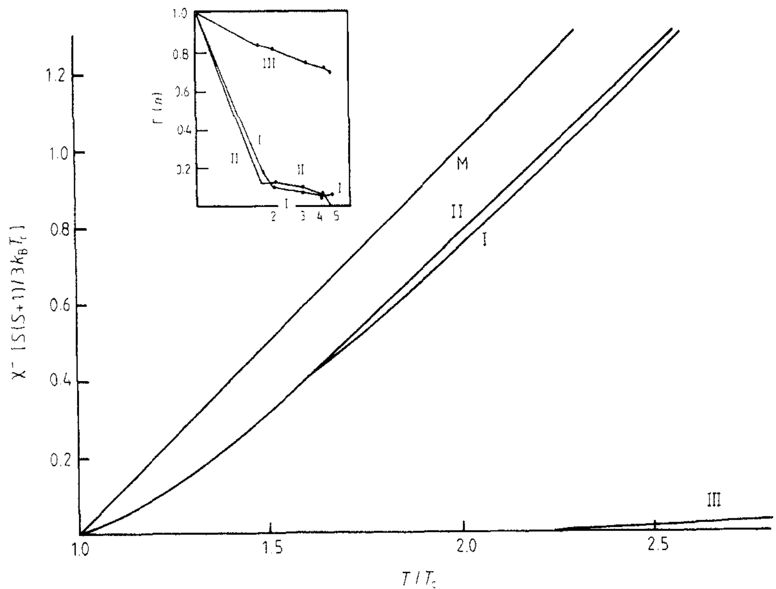

Figure 1. The calculated reduced inverse susceptibility $\chi^{-1}S(S+1)/3k_BT_c$ as a function of T/T_c for: (I) the nearest-neighbour case $\alpha = \beta = \gamma = 0$; (II) $\alpha = 1.412$, $\beta = 1.625$, $\gamma = -1.152$; (III) $\alpha = 1.412$, $\beta = -0.512$, $\gamma = -0.060$. Here α, β and γ are J_2/J_1, J_3/J_1 and J_5/J_1, where J_n is the exchange interaction between nth neighbours. The parameters for curve II were obtained from a fit to Lynn's room temperature spin wave data and curve III represents a strongly compensated case with the amount of short-range order postulated by Prange and Korenman. The inset shows the reduced correlation functions $\Gamma(n)$ for $n = 1, 2, \ldots 5$ in cases I, II and III with horizontal scale linear in distance.

because we found it necessary to make the coefficient of q^2 in $J(0) - J(\boldsymbol{q})$ very small to satisfy these conditions. As a result, fluctuations at many wavevectors become large so that T_c is considerably reduced from its mean field value T_c^{mf}. For $T \gg T_c^{mf}$ the reduced inverse susceptibility curve always lies parallel to the mean field curve M, so that χ is strongly enhanced for $T_c < T \ll T_c^{mf}$. The susceptibility in case II is very close to that for the nearest-neighbour case I and agrees qualitatively with the observed χ. We conclude that the Prange–Korenman picture is unlikely to apply to Fe and therefore does not provide a viable explanation of Lynn's neutron data (1975).

To calculate the neutron scattering functions $S(\boldsymbol{q}, \omega)$ we use the three-pole approximation of Lovesey and Meserve (1973) for the relaxation shape function $F(\boldsymbol{q}, \omega)$. The relation between S and F is

$$S(\boldsymbol{q}, \omega) = \hbar\omega[1 - \exp(-\hbar\omega/k_BT)]^{-1}\chi(\boldsymbol{q})F(\boldsymbol{q}, \omega) \tag{5}$$

where $\chi(\boldsymbol{q})$ is the static wavevector-dependent susceptibility. In the three-pole approximation

$$\mathrm{F}(\boldsymbol{q}, \omega) = \frac{1}{\pi}\frac{\tau\delta_1\delta_2}{[\omega\tau(\omega^2 - \delta_1 - \delta_2)]^2 + (\omega^2 - \delta_1)^2} \tag{6}$$

where

$$\delta_1 = \langle\omega^2\rangle, \qquad \delta_1\delta_2 = \langle(\omega^2 - \langle\omega^2\rangle)^2\rangle$$

$$\tau = (\pi\delta_2/2)^{-1/2} \tag{7}$$

and, for a given $\boldsymbol{q}$, $\langle\omega^2\rangle$ and $\langle\omega^4\rangle$ are the second and fourth moments of F, which may be expressed in terms of static correlation functions. Following Lovesey and Meserve, we approximate the four-spin correlation functions which appear in $\langle\omega^4\rangle$ by products of two-spin correlation functions given by equation (4). For the spherical model this procedure is exact and $\chi(\boldsymbol{q})$ is given by $(k_B T)^{-1}C(\boldsymbol{q})$ with $C(\boldsymbol{q})$ determined by equation (2). We have calculated $F(\boldsymbol{q}, \omega)$ as a function of ω, at various temperatures in the range $1.28\ T_c < T < \infty$, for values of $\boldsymbol{q}$ along various symmetry axes in both SC and BCC lattices. Many different sets of exchange parameters extending to fourth (SC) or fifth (BCC) nearest neighbours were considered. In general the shape function has either one maximum at $\omega = 0$ or two maxima, one at a non-zero frequency. The latter situation implies a peak in $S(\boldsymbol{q}, \omega)$ at a non-zero frequency and hence a (damped) propagating mode. In no case did we find a propagating mode for $q/q_{max} < 0.4$, where q_{max} corresponds to the zone boundary in the direction considered. The minimum value of q at which a propagating mode exists tends to decrease with increasing range of interaction. We have not considered the case of strong compensation because in this regime, which is anyway not applicable to Fe, factorisation of the four-spin correlation functions may be inaccurate. Of greatest interest are the results for case II considered above, with exchange parameters α, β, γ for the BCC lattice chosen to fit Lynn's room temperature spin wave dispersion curve for $Fe_{88}Si_{12}$. In fact in the (110) direction, the spin wave energies depend on J_1 and J_2 only through the sum $J_1 + J_2$. We chose $\alpha = J_2/J_1 = 1.412$ in accordance with results of You *et al* (1980) and the values of β and γ then follow by curve fitting. The absolute energy scale is fixed by choosing the nearest-neighbour exchange parameter $J_1 = 9.89$ meV so that the

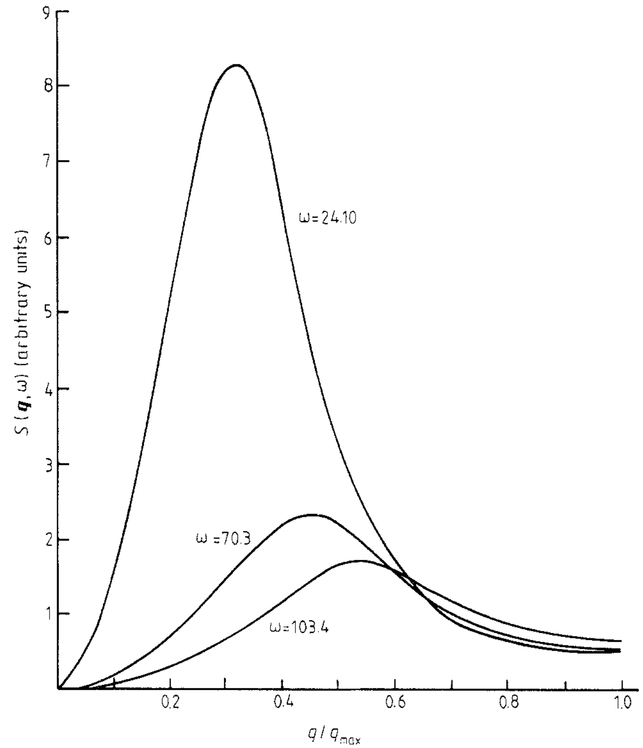

Figure 2. The calculated scattering function $S(\boldsymbol{q}, \omega)$ as a function of q in the (110) direction for parameters appropriate to $Fe_{88}Si_{12}$, case II of figure 1, at $T = 1.28\ T_c$. The curves correspond to three different values of ω specified in meV.

observed room temperature curve lies 15% lower than the calculated dispersion curve at $T = 0$, this renormalisation being of the magnitude expected (e.g. see Lynn 1975). The root mean square error in the fit to the dispersion curve is less than 3 meV. The calculated value of T_c is then 1290 K compared with the observed 970 K. To compare with Lynn's data we take $T = 1.28\ T_c$ and $\boldsymbol{q}$ in the (110) direction. We find that $F(q, \omega)$ shows peaks in the 'constant q' plots as a function of ω for $q \gtrsim 0.6\ q_{max}$, but for $q < 0.6\ q_{max}$ is monotonic. However, in 'constant ω' plots, which is how the experimental data is recorded, the calculated $S(\boldsymbol{q}, \omega)$ always shows a pronounced peak as a function of q, as shown for three values of ω in figure 2. It is the position of such peaks in the experimental data which Lynn plots as a dispersion curve and in figure 3 we compare this with our similarly plotted theoretical curve for $T = 1.28\ T_c$. The experimental curve is apparently independent of temperature between T_c and $1.4\ T_c$ and we find that, even for the largest value of ω plotted, the theoretical value of q increases by less than 2% on raising the temperature from $1.28\ T_c$ to $1.4\ T_c$. The observed region corresponds to $q/q_{max} < 0.5$ and the dispersion curve does not correspond to propagating modes in our theory. Lynn fits his 'constant ω' plots to a gaussian 'spin wave peak' plus sloping background. If the background lines are taken to slope downwards, more steeply asymmetric curves of the type shown in figure 2 are obtained. Korenman and Prange's theory, assuming giant short-range order, predicts a gaussian peak and the existence of propagating modes within regions of local order. Liu (1976) has previously applied a semi-empirical theory of the Heisenberg model above T_c to calculate 'constant ω' plots similar in shape to ours. He compares with experimental data on Ni (Mook *et al* 1973) and Fe (Lynn 1975) and concludes that there is no convincing indication of propagating modes. Our theory, leading to the same conclusion, is more firmly based and calculates the renormalisation of the 'dispersion curve' accurately (see figure 3). We conclude that a Heisenberg model, with exchange parameters leading to reasonable static properties, does not lead to spin wave peaks in 'constant q' plots for $q/q_{max} < 0.6$. The conclusion is particularly convincing since we have shown, by comparing with numerical integration of the equations of motion for a one-dimensional model with long-range interactions, that the method of moments tends to favour propagating modes, at least when there is little short-range order, so factorisation of the four-spin correlation function is a good approximation. Thus although we understand the form of Lynn's 'constant ω' scans and the renormalisation of the dispersion curve, we are unable to explain the existence of peaks in his 'constant q' plots (Lynn 1975, 1981). Notice, however, that the peak arises solely from a 'hole' in the spectrum at low frequency, the high frequency part being broad, as would be expected if there is little short-range order. We have no explanation of this rapid drop in intensity at low frequency but argue that it is unlikely to be vast short-range magnetic order for reasons given above.

We have assumed that the Heisenberg model can be roughly applied to Fe even though the local moments which exist above T_c do not correspond to localised electrons. A similar theory of Ni would be dubious since local moments are unlikely to exist above T_c in this case (Pettifor 1980). In applying the Heisenberg model it was essential to use fairly long-range interactions for two reasons. First, as is well known (Collins *et al* 1969), the low-temperature spin wave dispersion curve demands it. Secondly, the shift in the dispersion curve from low temperature to $T > T_c$ is found to be considerably larger for the nearest-neighbour model than that shown in figure 3 for interactions of longer range. Clearly our model cannot explain the observed sudden loss of intensity in the room-temperature spin waves which occurs at about

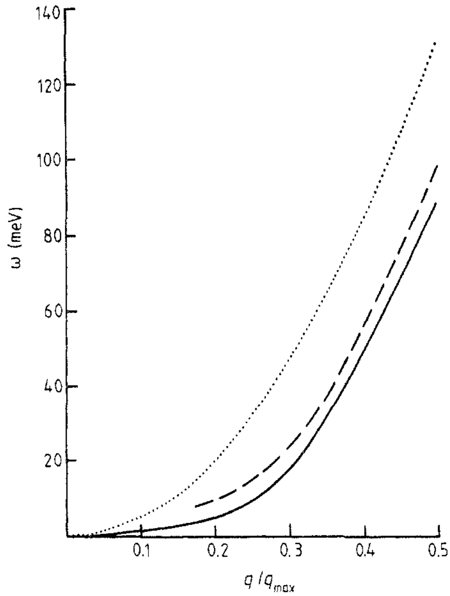

Figure 3. Experimental (broken curve) and theoretical (full curve) dispersion curves for $Fe_{88}Si_{12}$ at $T = 1.28\ T_c$ in the (110) direction. These were obtained by plotting the locus of peaks in constant ω scans. The theory used parameters corresponding to case II in figure 1. The dotted curve is the experimental spin wave dispersion curve at room temperature, which was used to determine these exchange parameters.

100 meV in both Fe and Ni where the modes encounter the Stoner continuum. Above T_c the experimental evidence for a similar sudden effect, which might support the giant short-range order concept, seems slender. A more gradual loss of intensity under the peaks in 'constant ω' plots is compatible with our results and this is unrelated to a 'Stoner cut-off' above T_c.

We are grateful to V Heine, J A Hertz, A J Holden, R D Lowde and C Windsor for useful conversations concerning this work. B S Shastry received a Commonwealth Bursary from the Royal Society and wishes to thank members of the Mathematics Department at Imperial College for their warm hospitality.

References

Collins M F, Minkiewicz V J, Nathans R, Passell L and Shirane G 1969 *Phys. Rev.* **179** 417
Cyrot M 1970 *Phys. Rev. Lett.* **25** 871
Edwards D M 1980 *J. Magn. Magn. Mater.* **15–18** 262
Evenson W E, Schrieffer J R and Wang S Q 1970 *J. Appl. Phys.* **41** 1199
Hasegawa H 1979 *J. Phys. Soc. Japan* **46** 1504
Hubbard J 1979 *Phys. Rev.* B**19** 2626, **20** 4584
Liu S H 1976 *Phys. Rev.* B**13** 2979
Lovesey S W and Meserve R A 1973 *J. Phys. C: Solid State Phys.* **6** 79
Lynn J W 1975 *Phys. Rev.* B**11** 2624
—— 1981 *Physics of Transition Metals 1981* (Inst. Phys. Conf. Ser. 55) p 669
Mook H A, Lynn J W and Nicklow R M 1973 *Phys. Rev. Lett.* **30** 556
Pettifor D G 1980 *J. Magn. Magn. Mater.* **15–18** 847
Prange R E and Korenman V 1979 *Phys. Rev.* B**19** 4691
Roth L M 1978 *Transition Metals 1977* (Inst. Phys. Conf. Ser. 39) p 473
You M V, Heine V, Holden A J and Lin-Chung P J 1980 *Phys. Rev. Lett.* **44** 1282