Spin Dynamics of Paramagnetic Iron

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The spin dynamics of the classical Heisenberg model on the bcc lattice is computed with a Monte Carlo molecular-dynamics approach in the paramagnetic phase at $T=1.025T_c$ and $1.275T_c$. The resulting scattering function $S(q,w)$ and its integral over restricted energy windows are computed and compared with the recent neutron-scattering experiments on iron. Reasonable overall agreement with much of the experimental data is found.

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Recent experiments by Wicksted *et al.*[1] and Brown *et al.*[2] on paramagnetic iron using spin-polarized neutron scattering have brough a new and welcome exactitude to the old debate on the magnetism of iron. These measurements yield the scattering function $S(q,w)$, and its convolution with resolution functions of a given energy width, *on an absolute scale.* These measurements are of considerable importance since they provide stringent tests of possible theories. The existing theories are broadly in two classes. Theories of one class assume that vast short-ranged order is present in the paramagnetic phase of iron[3] as suggested by the early work using unpolarized neutron scattering.[4] Another class of theories[5-7] view iron as a disordered local-moment situation wherein fluctuating Anderson-type local moments exist in the paramagnetic phase. These theories have been rather successful in explaining the thermodynamic data on iron. In addition, the spin dynamics of the Heisenberg model in an approximation scheme[6] due to Shastry, Edwards, and Young (SEY) is consistent with the raw data of the early neutron-scattering work.[4] The one apparent difficulty with the calculation was with regard to the propagating modes; the theory yielded none at the value of q suggested by the experiment. In view of the recent data, this particular feature would appear to be satisfactorily resolved.

Turning to the recent experiments, Brown *et al.* have measured the scattering function and its convolution with an instrumental resolution function with width [full width at half maximum (FWHM)] $\simeq 43$ meV at various temperatures. If the width were infinite, then the measurement would yield the static correlation function (CF) in q space, from which the r-space CF could be deduced. However, Edwards[8] pointed out that the measured pseudo-CF's lead to unreasonably small values of the rms local moment, and suggested that the energy window used was insufficient to extract the true static CF. The initial experiments at Brookhaven used an even smaller energy window ($\simeq 40$ meV), and more recent measurements use a larger window (100 meV).[9] The quantum of missing intensity and its distribution in energy are problems of great current interest.

The aim of this investigation is to calculate $S(q,w)$ of iron from a Heisenberg model, with the object of finding the expected ideal intensities and also of finding the expected pseudo-CF by convoluting the ideal spectrum with resolution functions of various widths. The results are expected to be sensitive to the extent and nature of inelasticity (i.e., the line shapes, and widths) in addition to the absolute values of the static CF's. I compared the spectrum of SEY with preliminary experimental data from Wicksted *et al.* and found that for small q, the theoretical lines were narrower than the data. Also, Brown *et al.* pointed out that the situation at larger q ($0.5Q_{zb}$) was reversed; the theoretical lines were considerably wider than the experimental ones (see Fig. 1).

This situation motivated the present study. In the present work we avoid analytical approximations, and seek to estimate $S(q,w)$ from a direct computation of the time evolution of spin arrays by the Monte Carlo molecular-dynamics technique.

The technique used in this work has been applied previously[10-12] to the Heisenberg model on sc and fcc lattices. The standard Monte Carlo (MC) method is used to generate typical arrays at a given temperature, and these are used as initial conditions for the natural dynamics. We consider the nearest-neighbor exchange model in this work. An analysis of the room-temperature spin-wave spectrum shows that this hypothesis is reasonably adequate up to 100 meV. We consider the spins S to be classical vectors with length $[s(s+1)]^{1/2}$ where $s=1$. Thus the semiclassical Heisenberg model is given by

$$\mathscr{H} = -Js(s+1)\sum_{\langle ij\rangle}\vec{\sigma}_i\cdot\vec{\sigma}_j; \quad |\vec{\sigma}_i|=1, \tag{1}$$

with $\vec{S}_i=\vec{\sigma}_i[s(s+1)]^{1/2}$. The dynamics of the

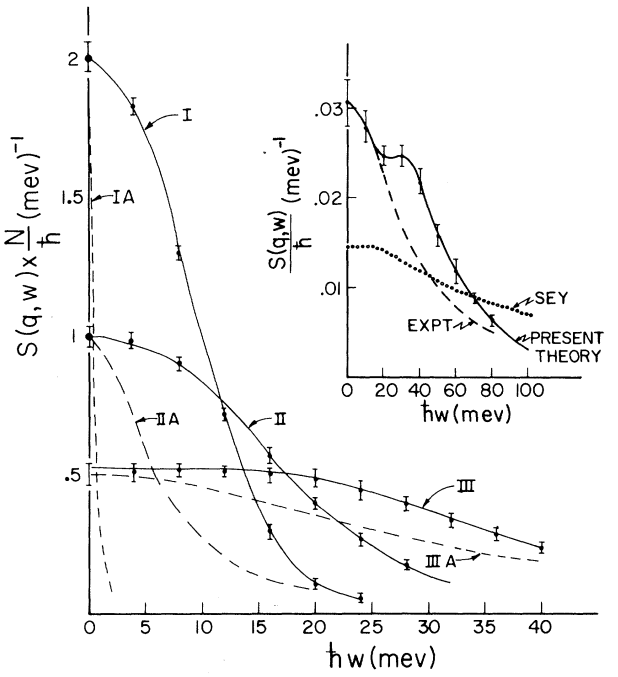

FIG. 1. $S(q,w)/h$ multiplied by a constant N at $1.025T_c$. Curves I, II and III are from present calculation for $q/q_{zb}=0.125$, 0.25, 0.375 with $N=1.28$, 3.93, and 8.4. Curves IA, etc., are from SEY at the same q's with $N=0.074$, 2.0, and 9.98. Inset is at $q=0.5q_{zb}$ with $T=1.275T_c$ and the dashed line is from data of Brown *et al.* normalized at $w=0$.

unit vectors is given by replacing the quantum-mechanical equation of motion of $\vec{S}_i$ by appropriate classical equations. Thus

$$t_0\frac{d}{dt}\vec{\sigma}_i=\vec{\sigma}_i\times\sum_{j\in \mathrm{nn}i}\vec{\sigma}_j, \tag{2}$$

where "nn$i$" denotes "nearest neighbors i." The natural time unit in the problem is $t_0=\hbar/J[s\times(s+1)]^{1/2}$. The equation of motion [Eq. (2)] leads to spin waves in the perfectly ordered state with dispersion $E_q=8J[s(s+1)]^{1/2}\Psi_q$, where

$$\Psi_q=1-\cos(q_xa/2)\cos(q_ya/2)\cos(q_za/2).$$

We determine J, the only parameter in the theory, by comparing this expression with the observed spin-wave data (extrapolated to $T=0$ by multiplying by 1.15). This yields $J=22.42$ meV. The high-temperature series estimate of T_c for the classical Heisenberg model[13] for the bcc lattice gives $T_c=1069.4$ K, in reasonable agreement with experimental values $\simeq 1000$ K.

The scattering function $S(q,w)$ is found from the auto-CF of $\vec{\sigma}$. With $\vec{\sigma}_q=N^{-1/2}\sum_j\vec{\sigma}_j\exp(-i\vec{q}\cdot\vec{r}_j)$, and $C_q(t)=\langle\vec{\sigma}_{-q}(t)\cdot\vec{\sigma}_q(0)\rangle$, we have

$$\hat{C}_q(w)=\int_{-\infty}^{+\infty}(dt/2\pi)C_q(t)\exp(-iwt), \tag{3}$$

$$S(q,w)=\frac{2s(s+1)\hat{C}_q(w)}{1+\exp(-\hbar w/k_BT)}. \tag{4}$$

The detailed-balance factor in Eq. (4) has been introduced following Windsor[10] and is in the spirit of the semiclassical approximation. It should be noted that there is no unique prescription for reconstructing the quantum correlations given the classical ones. The popular prescription used here has several advantages.[10] I have also checked that other treatments of the detailed-balance factor lead to comparable results. The object of special interest is the pseudostatic CF defined as

$$M_q^2(w_0)=4\int_{-\infty}^{+\infty}dw\,S(q,w)R(w;w_0). \tag{5}$$

Here $R(w;w_0)$ is an instrumental resolution function assumed to be Gaussian with $R=\exp(-w^2/\sigma^2)$, where $\sigma=\frac{1}{2}\omega_0(\ln 2)^{-1/2}$. Thus w_0 is the full width at half maximum (FWHM). In the limit $w_0=\infty$, the ideal CF's are recovered as $M_q^2(\infty)\to 8\chi_q$ with $\chi_q=C_q(t=0)$, an equilibrium quantity. The bulk susceptibility is related to χ_0 through $\chi_B=(N/3)g^2\mu_B^2s(s+1)\chi_0/k_BT$.

In the present calculation, the major effort involved is in the time evolution of typical arrays through Eq. (2). The time evolution must necessarily be truncated at some value of time, say $t_{\max}$. This introduces an intrinsic linewidth into the computed $S(q,w)$, of order $\hbar/t_{\max}$, and one would like to minimize it. I chose $t_{\max}=24t_0$ and constrained the maximum time lag to be $12t_0$. This calculation is, I believe, the longest for the array sizes considered, and was forced on me by the large value of J in comparison with typical experimental resolution widths. Each dynamical run requires some 6 h central-processing-unit time on a Cyber 170/730 computer.

In brief, the computation consists of a MC procedure by which I generated ten samples of arrays at each temperature. The array size was chosen to be of $2\times16^3(=8192)$ spins in order to minimize finite-size effects. The MC ages of the samples retained were 2000 ,..., 11 000 steps per spin. I estimated the equilibrium values of static CF's in real space out to the tenth-neighbor shell. The CF's of $\vec{\sigma}_q$ were also estimated. Stable averages of CF's resulted in the q space on averaging over a few thousand samples. This procedure was followed by the time evolution through Eq. (2). The initial-

value problem was integrated by the fourth-order Runge-Kutta method due to Gill.[14] This method has the advantage of requiring only $3N$ storage spaces for a fourth-order method where N is the number of coupled equations solved (24 576). The step size was chosen (0.01) after tests involving the behavior under time reversal. The variables monitored were $\vec{\sigma}_q(t)$ which were stored for $t=0$, 0.3, 0.6 ,..., 24. We study the popular $\langle 110\rangle$ direction in this work, which provides eight nonzero q vectors with periodic boundary conditions.

The procedure adopted was to compute the auto-CF $C_q(t)/C_q(0)$ for each sample. The estimate of the spectral function was obtained by multiplying the auto-CF by the average of the static CF's obtained from the MC procedure and further multiplying by the Tukey spectral smoothing function $0.5[1+\cos(2\pi t/t_{\max})]$ in order to reduce the distortions in the spectrum.[15] The resultant function was Fourier transformed. The intrinsic linewidth is 16.6 meV (FWHM). This procedure was carried out for each sample at a given temperature and the final estimate of the $S(q,w)$ was found by averaging over the ten samples. The rms fluctuations were computed and are indicated as error bars in Fig. 1.

The resulting $S(q,w)$ at the two temperatures considered are shown in Fig. 1. The SEY spectrum should be broadened by convoluting with the Tukey window function in order to compare with the present spectrum. In the inset in Fig. 1 the spectrum is compared with the data of Brown *et al.* It is seen that the energy scale over which S falls off is considerably smaller in the present calculation as compared to the SEY spectrum and closer to the experimental curve. A shoulder in the theoretical curve is also consistent within the error bars.

The computed static CF's in q space are expressible rather neatly in terms of the Ritchie-Fisher functional form[16] for the CF $\chi_q=\alpha(\beta+\gamma\psi_q)^{\eta/2}/(\beta+\psi_q)$, where $\eta=\frac{2}{45}$ and $\gamma=(5-\eta)/3$. The uniform static susceptibility is $\alpha\beta^{\eta/2-1}$ and the correlation length $\xi=a/(8\beta)^{1/2}$. The CF's in real space can be obtained by Fourier transforming χ_q and are in close correspondence with the direct estimates from the MC procedure. The parameters α and β are estimated as (0.7528, 0.0043) and (0.9647, 0.1305) at $T=1.025T_c$ and $T=1.275T_c$, respectively, leading to the uniform susceptibility and correlation length (156.5 ± 10, 15.55 Å) and (7.07 ± 0.6, 2.81 Å) at the two temperatures. The CF in real space is quite similar in nature to the spherical-model estimates of SEY and does not possess giant short-range order.

In Fig. 2 I compare the computed $M(w=100$ meV) with the experimental data of Wicksted *et al.*[11] at $T=1.02T_c$. The data are seen to be in good accord on an absolute scale over the entire zone. The recent results of Wicksted *et al.* are with pure single crystals of Fe and are in better accord with theory as compared to earlier data using Fe(4% Si). The computed $M(w_0=43$ meV) is compared with the data of Brown *et al.*[2] at $T=1.273T_c$ in the inset of Fig. 2. It is remarkable that in both cases the SEY results for M_q are quite close to the present calculation and also the experiments despite the deviations in Fig. 1.

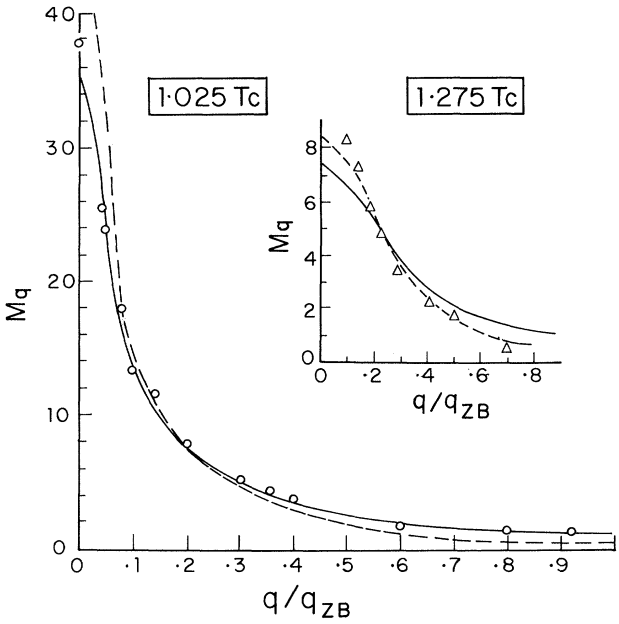

FIG. 2. M_q vs q at $T/T_c=1.025$ compared with the Brookhaven data at $1.02T_c$ (circles) using pure Fe (FWHM = 100 meV). The inset corresponds to $T=1.275T_c$ and the triangles are from the Grenoble data (FWHM = 43 meV). The dashed lines are from SEY. M_q is in units of μ_B.

In Fig. 3 I have plotted the intensity loss as a result of the frequency cutoff for various values of the energy window. The remarkable feature is the somewhat large loss for windows as large as 100 meV for q close to the zone boundary. This is a consequence of the small tails in $S(q,w)$ extending out to 150 meV or so. For the itinerant-electron model we should expect weak tails extending out to much higher energies, a few electronvolts. This implies that it is extremely difficult, if not impossible, to extract the true static CF from limited energy integrations as performed in the recent experiments. I propose that meaningful results can be extracted by performing limited energy integrations

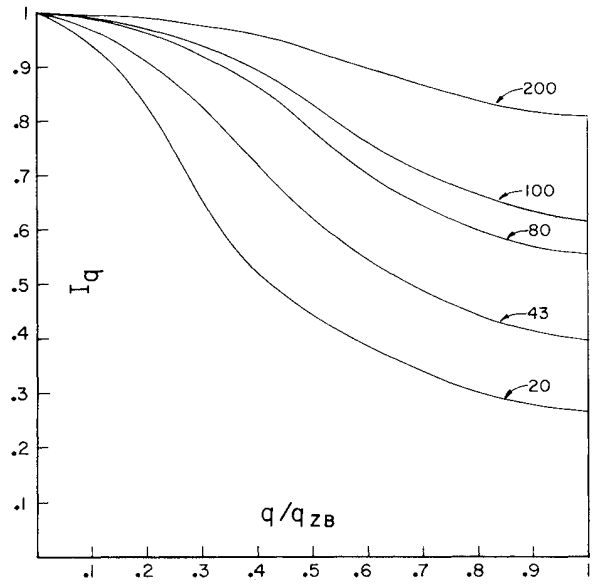

FIG. 3. Intensity loss $I_q = M_q(w_0)/M_q(\infty)$ at $1.275T_c$. The FWHM of windows are indicated along the curves.

with varying windows at *a fixed temperature* and comparing with theoretical predictions, such as are implicit in Fig. 3. In conclusion I feel that this investigation demonstrates that the Heisenberg model description of paramagnetic iron is in reasonable accord with a considerable amount of existing experimental data, both in equilibrium and away from it.

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