Molecular-dynamical calculations of the polarized neutron scattering cross section for Heisenberg ferromagnets

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Molecular-dynamical calculations have been performed on the classical Heisenberg model. The method of calculation is the same as that used by Watson, Blume, and Vineyard. The polarized neutron scattering cross section was found from these calculations. Highly damped "forbidden" magnon scattering of neutrons (i.e., magnons excited by neutrons initially polarized along the magnetization and wave vector) was found, but only in the immediate vicinity of T_c. Below T_c the "forbidden" magnons are overdamped.

Mook, Lynn, and Nicklow[1] have observed in inelastic neutron scattering experiments that underdamped magnonlike excitations can exist well above T_c in nickel and iron over most of the Brillouin zone. It is generally believed that the existence of magnons above T_c is due to the presence of magnetization fluctuations which vary slowly in space and time.[2] According to this explanation, magnons should appear above T_c in all magnetic materials, not just in metals. Yet recent (as well as previous) experiments on EuO (Ref. 3) (a system well described by a short-range Heisenberg model) show well-defined magnons only at the zone boundary, in contrast to nickel and iron, although these magnons are found to persist out to $2T_c$. Recent polarized neutron scattering experiments on a nickel alloy have shown the existence of "forbidden" magnons (i.e., magnons excited by neutrons polarized along the magnetization. Such magnons, which are absolutely forbidden at zero temperature, can exist only because of short-range order). These "forbidden" magnons were found to exist down to at least 90% of T_c.[4] Since magnons above T_c are apparently not as readily observable in EuO as in nickel and iron, it would be interesting to also study the "forbidden" magnons in a short-range Heisenberg model system.

In order to predict what might be expected in such an experiment, the present author has performed molecular dynamical calculations on the classical near-neighbor Heisenberg model to calculate the polarized neutron scattering cross section. The calculation follows the treatment due to Watson, Blume, and Vineyard.[5] A Monte Carlo calculation is performed in order to determine a typical spin configuration for a given temperature as a scattering configuration for the molecular dynamics calculation. The equation of motion that was solved is

$$\frac{d\vec{S}_j}{dt} = -\left(\sum_l J_{jl}\vec{S}_l + \vec{H} \right) \times \vec{S}_j \ , \qquad (1)$$

where $\vec{H}$ is an external magnetic field, $\vec{S}_j$ is a classical spin vector on site j and the exchange interaction J_{jl} was restricted to nearest neighbors. The neutron scattering cross section for neutrons totally polarized along (opposite) the magnetization is proportional to the Fourier transform of the correlation function

$$C_{\vec{k}}^{\pm}(t) = \langle S_{\vec{k}}^{\mp}(t) S_{\vec{k}}^{\pm}(0) \rangle \ , \qquad (2)$$

where

$$S_{\vec{k}}^{\pm} = \frac{1}{(N)^{1/2}} \sum_j e^{i\vec{k}\cdot\vec{R}_j} S_j^{\pm} \ , \qquad (3)$$

where N is the number of lattice sites in the system (the indicated sums were performed over all sites in the system), and

$$S_j^{\pm} = S_j^x \pm i S_j^y \ . \qquad (4)$$

We may approximate the thermal average in Eq. (2) by a long-time average, and hence write Eq. (2) as

$$C_{\vec{k}}^{\pm}(t) = \frac{1}{\tau} \int_0^{\tau} dt_0 S_{\vec{k}}^{\mp}(t+t_0) S_{\vec{k}}^{\pm}(t_0) \ , \qquad (5)$$

where τ is the time over which the molecular dynamics calculation was run. The time Fourier transform of Eq. (5) then becomes

$$C_{\vec{k}}^{\pm}(\omega) = \int_0^{\tau} e^{-i\omega t} C_{\vec{k}}^{\pm}(t) dt = \frac{1}{\tau} |S_{\vec{k}}^{\mp}(\omega)|^2 \ , \qquad (6)$$

where

$$S_{\vec{k}}^{\pm}(\omega) = \int_0^{\tau} dt\, e^{i\omega t} S_{\vec{k}}^{\pm}(t) \ . \qquad (7)$$

It is then clear that the permitted ("forbidden") magnons which appear in $C_{\vec{k}}^{+}(\omega)$ $[C_{\vec{k}}^{-}(\omega)]$ are magnons in which the spin precesses counterclockwise (clockwise) in the applied magnetic field.

Most of the calculations were performed on a cube with 1000 spins on a simple cubic lattice. Staggered boundary conditions, as described in Ref. 5, were used. The calculations essentially follow the procedure of Ref. 5. Equation (1) was integrated out to

$t = 2.5\ \mathrm{J}^{-1}$, which was found to be sufficient to resolve the magnons. (Longer runs were made to test this.) The Fourier transforms of the correlation functions $[C_{\vec{k}}^{\pm}(\omega)]$ were found by standard Fourier-transform methods[6] for several values of the wave vector k. The noise in the spectrum, which results from integrating only over a finite time interval, was eliminated by taking an average over $C_{\vec{k}}^{\pm}(\omega)$ with a Gaussian of variance 0.4 J.[7] The way in which this procedure eliminates noise is described in Ref. 7. All reported results for $C_{\vec{k}}^{\pm}(\omega)$ have been averaged in this way. Such runs were made for 20 starting configurations for each temperature, and these results were then averaged. Each of the 20 runs was started with a Monte Carlo calculation with one million interactions, i.e., 1000 trial steps per atom. (The acceptance ratio in these calculations was about 50%.) Since we are studying the neutron scattering cross section as a function of whether the neutrons are polarized along or against the sample magnetization, it was necessary to apply a magnetic field of strength 0.1 J to guarantee that the sample magnetization remained aligned with the z axis, the assumed neutron polarization direction. Although this is a fairly large value for most real systems, such a large field was found to be necessary in order to guarantee that the sample magnetization remained aligned with the z axis in these calculations. The gap introduced in the spin-wave spectrum is 0.1 J, which is small compared to the spin-wave bandwidth and hence will not be relevant for our results. Spectra for positive and negative $\vec{k}$ were found to differ by a small amount, but the shapes of the spectra for $\vec{k}$ and $\vec{k}$ were qualitatively the same. Results of the calculations are shown in Fig. 1 for four temperatures. The results shown in Figs. 1(b)–1(e) are shown only for $\omega > 0.5$ J because the results for $\omega < 0.5$ J are believed to be spurious. The reasons for this conclusion are as follows: Runs were made for zero exchange (i.e., free spins) which show peaks in the spectral functions for $\omega \leq 0.5$ J and which shift closer to $\omega = 0$ when we reduce the width of the Gaussian function used to average over $C_k^{\pm}(\omega)$. When the width of the Gaussian is reduced for runs made with nonzero exchange and magnetic field, however, only those peaks in the spectral functions for $\omega \leq 0.5$ J shift towards $\omega = 0$. Clearly the peaks introduced by the field can only occur for $\omega \leq 0.1$ J and hence they clearly have been shifted by the Gaussian average. Hence, modes with $\omega < 0.5$ J are most likely overdamped (or they really occur at $\omega \leq 0.1$ J), and therefore the data for such low frequencies are meaningless. The origin of these spurious peaks can be understood if we consider the following averaging procedure that was used:

$$\overline{C}_{\vec{k}}^{\pm}(\omega) = \int_0^{\Delta} d\omega' \exp\left[-\left(\frac{\omega - \omega'}{\Gamma}\right)^2\right] C_{\vec{k}}^{\pm}(\omega') \ ,$$

where $C_{\vec{k}}^{\pm}(\omega)$ and $\overline{C}_{\vec{k}}^{\pm}(\omega)$ are the unaveraged and averaged spectral functions, respectively.[7] If $C_{\vec{k}}^{\pm}(\omega')$ were a constant, it is easily shown that $\overline{C}_{\vec{k}}^{\pm}(\omega)$ would have a peak at $\omega = \Delta/2$. If $C_{\vec{k}}^{\pm}(\omega')$ had a peak at the origin, however, it is clear that $G(\omega)$ should be larger if ω were closer to $\omega = 0$ so that the peaks in $C_{\vec{k}}^{\pm}(\omega')$ and the Gaussian coincide. The two competing tendencies should lead to a peak between $\omega = \Delta/2$ and 0. If the Gaussian is narrower, the latter tendency predominates, whereas if it is wider, the former predominates. Thus, as Γ decreases, the peak in $\overline{C}_{\vec{k}}^{\pm}(\omega)$ shifts towards $\omega = 0$. If the peak in $C_{\vec{k}}^{\pm}(\omega')$ occurred at a value of $\omega = \omega_0$ not equal to zero, the peak in $\overline{C}_{\vec{k}}^{\pm}(\omega)$ would shift towards ω_0. This is what was found.

We see several things in these spectra. First of all, we see a tendency for the spin waves to soften rapidly as T_c (about 1.5 J)[5] is approached. In fact at T_c, the permitted magnon at the Brillouin-zone boundary has an energy half as large as its energy near $T = 0$. At $T = 1.75$ J, the same magnon energy has dropped to about one-fourth of its $T = 0$ value. The magnons are also found to be broadened in the vicinity of T_c by an amount which is noticeably larger than the broadening introduced in the spectrum by the fact that the time interval over which Eq. (1) was integrated was finite. [This can be seen by comparing with Fig. 1(a).] At $T = 1.25$ J all of the "forbidden" magnons appear to be overdamped although a noticeable "forbidden" magnon intensity is still present (to be contrasted with the run made at $T = 0.02$ J). In Fig. 1(e), results for an 8000-spin system for $T = 1.75$ J are reported in order to check the effects of finite size on the previous runs. Although the results differ in detail from those shown in Fig. 1(d), the qualitative conclusions are not changed. Namely, the magnons have all shifted to very low frequencies at this temperature and the "forbidden" magnons exist at this temperature, but they occur at extremely low frequencies. If they are not overdamped, they are almost overdamped. The differences in the details of the spectra are probably due to the small size of the samples and to thermal fluctuations.

Figure 2 shows one slice of the cubic sample, giving a rough indication of the reversed spin cluster structure for one sample at $T = 1.75$ J. We see that despite the fact that well-defined magnons were found at this temperature, the clusters of spins with negative S^z are not all that well defined. Of course, this figure is only useful in understanding the magnons which occur in the transverse correlation functions studied in this article.

To summarize, molecular-dynamical calculations of the polarized neutron scattering cross section for the classical Heisenberg model show highly damped "forbidden" and permitted magnons in the vicinity of the Curie temperature. The results for permitted mag-

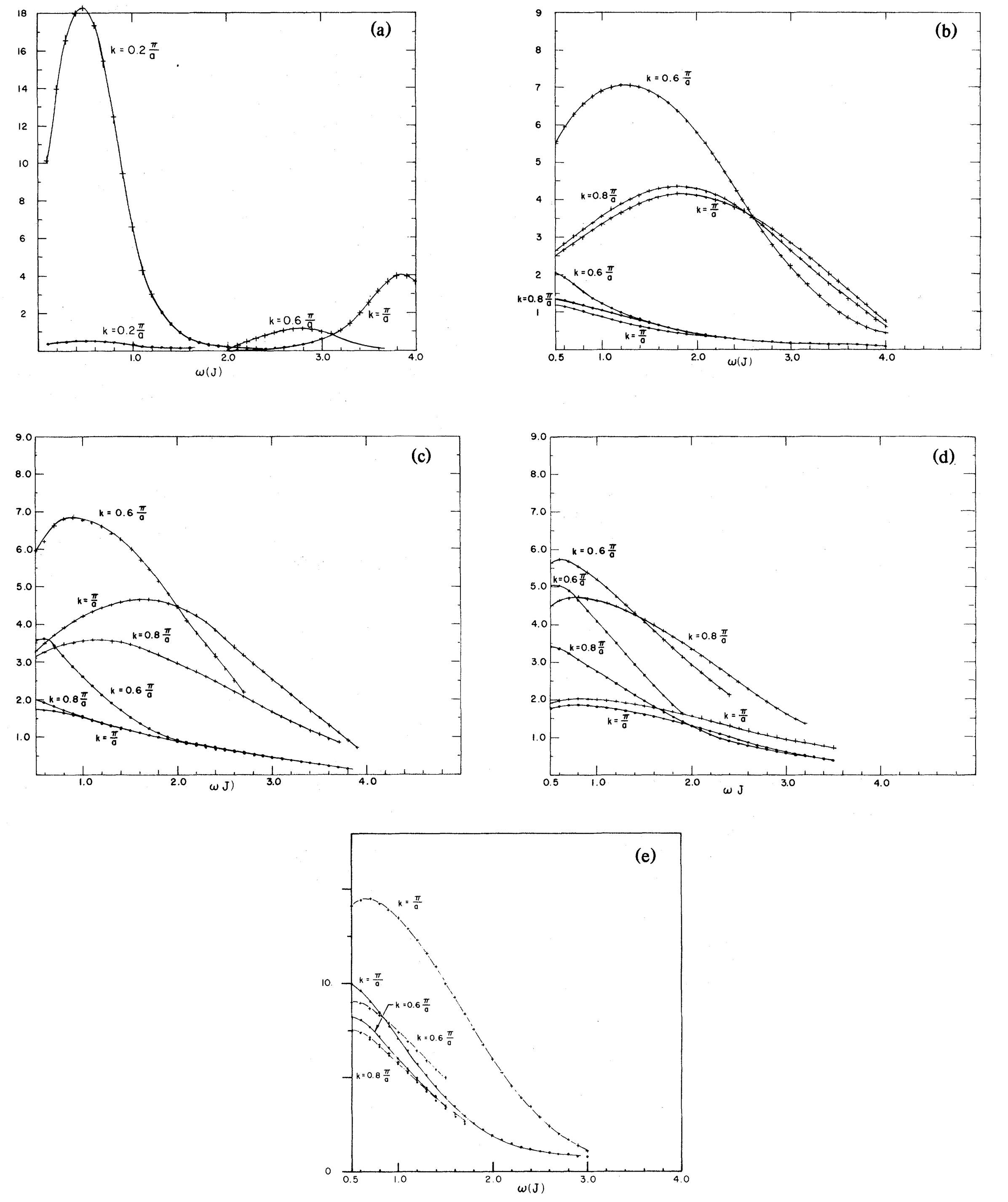

FIG. 1. "Forbidden" and permitted magnon intensities are plotted as a function of magnon energy for several temperatures. Here a cross (+) signifies permitted [i.e., $C^{\pm}_{\vec{k}}(\omega)$] and a dot (·) signifies "forbidden" magnons [i.e., $C^{\mp}_{\vec{k}}(\omega)$]. The results for $C^{\pm}_{\vec{k}}(\omega)$ shown here have been averaged over ω with a Gaussian of width 0.4 J. Thus, much of the width of the magnon peaks is due to this averaging and the finite range of integration. (a) $T = 0.02$ J ($m = 1.0$), (b) $T = 1.25$ J ($m = 0.5$), (c) $T = 1.5$ J ($m = 0.2$), (d) $T = 1.75$ J ($m = 0.1$), where m is the magnetization per lattice site ($m = 1.0$ is saturation). For a 1000-spin sample, (e) $T = 1.75$ J ($m = 0.043$), for an 8000-spin system. For this case only four runs rather than 20 were averaged. The intensity is in arbitrary units.

o	o	o	o	o	o	o	o	o	o	o
o	o	o	o	o	o	o	×	o	o	o
o	×	o	o	o	o	×	×	o	o	o
o	o	×	×	o	o	o	o	×	o	o
o	o	o	o	×	×	o	o	×	×	o
o	o	o	o	×	×	×	o	×	×	×
o	o	×	×	×	×	×	×	×	×	×
o	o	o	o	×	×	o	o	×	×	o
o	o	o	×	o	×	o	o	o	×	×
o	o	o	o	o	o	o	o	o	×	o
o	o	o	o	×	o	o	o	o	o	×

FIG. 2. Atomic positions of reversed spins, for $T = 1.75$ J. Here an × signifies an atom with a negative and a circle a positive Z component of spin.

nons in the vicinity of T_c compare favorably with approximate calculations on the simple cubic Heisenberg model due to Hubbard.[8] At lower temperatures, the "forbidden magnons" rapidly become overdamped. Although exact comparison is not possible because the Eu ions in EuO lie on a face-centered-cubic lattice which has the next-nearest-neighbor interaction, these calculations should give an indication of the results expected in a polarized neutron scattering experiment on a short-range Heisenberg model system such as EuO. Actually, the effect of having more nearest neighbors (as occur in a face-centered-cubic lattice) and of having further neighbor ferromagnetic exchange interactions will be to make the system better described by mean-field theory. That is, the fluctuations will be suppressed, which will make it less likely to have "forbidden" magnons in a system such as EuO. In any case, the present simulations should not be any less representative of EuO than Hubbard's results. Our conclusion is that short-range interaction Heisenberg systems are not expected to show prominent "forbidden" magnon scattering.

ACKNOWLEDGMENT

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