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Propagating Energy Modes in the Classical Heisenberg Chain: "Magnons" and "Second Magnons"

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The ferromagnetically coupled classical Heisenberg chain in an applied magnetic field has been studied by computer simulation. The results indicate the presence of a second collective mode, in addition to the damped spin-wave–like modes which have previously been observed in the absence of a magnetic field. For intermediate wavelengths, the mode manifests itself by well-defined oscillations in the energy-density correlation function, and by a second peak in the spectrum of the longitudinal spin-density correlation functions.

It is now well established that a classical one-dimensional Heisenberg magnet can support short-wavelength propagating spin-density modes, in spite of the lack of long-range order. The existence of such modes can be understood in terms of the strong short-range order present in one-dimensional magnets at low temperature ($k_B T < |J|$), the short-range order being characterized by an inverse correlation length κ which, for low temperatures, is proportional to the temperature. For wavelengths less than κ^{-1} the system appears ordered, and can therefore support collective spin-density oscillations, or "magnons"; however, the overall lack of long-range order leads to a damping of these excitations, and this damping increases as the temperature is raised. This qualitative picture is confirmed by a number of theoretical[1,2] and computer-simulation studies,[1,3,4] and is also in agreement with experimental results on $(CD_3)_4NMnCl_3$.[1,5]

In this Letter, we report computer simulation results which show that an applied field leads to striking new features in the response functions of the classical Heisenberg chain, for the case of ferromagnetic coupling.

Our computer-simulation calculations are based on the method described in detail in Steiner, Villain, and Windsor[1] and in Windsor and Wheaton.[6]

Besides including the effects of the applied field, we have improved the accuracy with which the equations of motion are integrated, which permits simulation over much longer periods of time.[3]

The computer simulation is performed for a system of N classical spins $\vec{S}_i$, $i=1,2,\ldots,N$, which we shall take to be unit vectors. The calculation is performed in a canonical ensemble, a Monte Carlo procedure being used to simulate a spin array in thermal equilibrium at the required temperature. For each spin array chosen, the motion of the spins is evaluated by integration of the equations of motion ($\hbar=1$),

$$\frac{d}{dt}\vec{S}_i = J\sum_{\delta=\pm1}\vec{S}_i\times\vec{S}_{i+\delta}+\vec{S}_i\times\vec{h}, \tag{1}$$

where $J>0$ for ferromagnetic coupling, the lattice spacing has been taken as unity, and factors of the gyromagnetic ratio and Bohr magneton have been absorbed into the field $\vec{h}$, which has been chosen to lie along the z direction. For later reference, we introduce the reduced temperature $T^*=k_B T/|J|$ and reduced field $h^*=h/|J|$. We have used a seventh-order approximation for integration of the equations of motion, the required derivatives being calculated by repeated differentiation of (1). We have calculated[7] both the spin-spin and energy-energy density correlation functions, defined by

$$C_\alpha{}^S(q,t)=\langle S_{-q}{}^\alpha(0)S_q{}^\alpha(t)\rangle-\langle S_q{}^\alpha\rangle^2,$$
$$\alpha=x,y,z,\quad C^E(q,t)=\langle E_{-q}(0)E_q(t)\rangle-\langle E_q\rangle^2, \tag{2}$$

where $S_q{}^\alpha$ and E_q are Fourier components of, respectively, the spin components and the local energy density. The angular brackets denote an ensemble average which, in the simulation calculation, corresponds to an average over N_c starting arrays, chosen independently by the Monte Carlo procedure.

In order that size effects should not be important, the number of spins N must be much greater than the correlation length κ^{-1}, and must also satisfy $N>vt_{max}$, where v is the maximum velocity of propagation of excitations.[8] Assuming the latter to be given approximately by $2|J|$ (the spin-wave value), we obtain the condition $N>2|J|t_{max}$. The calculations which we report below are for a ferromagnetically coupled chain at a reduced temperature $T^*=0.3$ and with an applied field $h^*=1.0$, and also $h^*=0.0$ for purposes of comparison. The equations of motion have been integrated up to times $t_{max}=20|J|^{-1}$ and, to satisfy the condition above, we have taken $N=80$.

In Fig. 1(a), results for the longitudinal spin-density correlation function $C_z{}^S(q,t)$ for finite field ($h^*=1.0$) and zero field are compared, for $q=0.2\pi$. For this wave vector, the field is seen to have a pronounced effect on the time dependence of the correlation function, but with increasing wave vector the effect is less marked. The effect of the field is even more pronounced in the behavior of the energy-density correlation function $C^E(q,t)$, which is plotted in Fig. 1(b), again for $h^*=0.0$ and 1.0 and for the same wave vector. In zero field, the decay of $C^E(q,t)$ is strongly damped for all wave vectors, and gives little indication of oscillatory behavior, while for $h^*=1.0$ the correlation function shows clear oscillatory behavior, characteristic of a damped propagating mode. The correlation function continues to show oscillations, but with increasing damping, as the wave vector is increased.

The behavior of this "second magnon" mode is more transparent in the frequency spectrum of the correlation functions and, because it is directly related to, for example, the neutron scattering cross section which one would observe in a "real" experiment, we have concentrated on

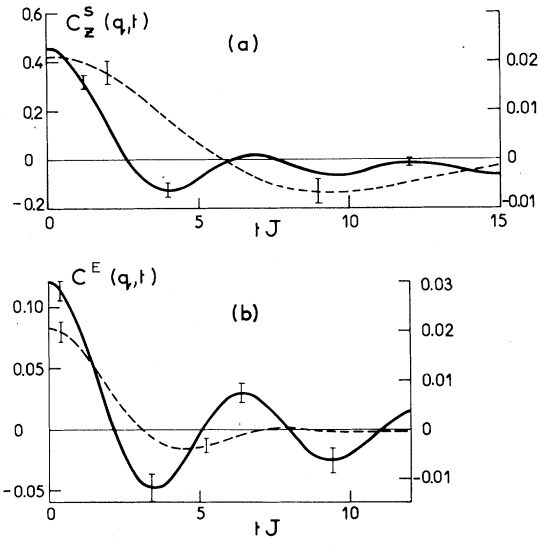

FIG. 1. Comparisons of the variation with time of (a) the spin-density and (b) the energy-density autocorrelation functions with an applied field $h^*=1.0$ (full lines) and in zero field (broken lines) for a wave vector $q=0.2\pi$. The vertical scales on the right are for $h^*=1.0$, those on the left for $h^*=0$. Averages and rms deviations (error bars) were obtained from five runs each averaged over 100 configurations, or 200 configurations [(a), $h^*=1.0$].

the spectrum of the spin-density fluctuations. Formally, the spectrum $F(q,\omega)$ is readily obtained as the Fourier transform of the normalized correlation function. However, spurious features in the Fourier transform can be obtained because of the cutoff, t_{max}, on $C(q,t)$, and because of thermal and statistical "noise." Consequently, following previous practice,[3,4,6] we characterize the frequency response by fitting the computer simulation data to some suitable function. We have, in fact, explored several possible fitting functions, to eliminate the possibility that any structure found in the spectrum is an artifact of the fitting procedure. The forms which we have investigated include a single-damped-harmonic-oscillator (DHO) form, a sum of two DHO's, and a form derived from a continued-fraction expansion of $\tilde{C}(q,s)$, the Laplace transform of $C(q,t)$. For the latter procedure, which is explained in detail in Ref. 4, we have investigated terminations of the continued fraction at different levels, and we shall refer to the different forms by the order of the polynomial in ω in the denominator of the resulting expression for $F(q,\omega)$.[9] It is clear that the amount of structure possible in the frequency spectrum, obtained by fits to the simulation data, is limited by the form of the fitting function, and this means that, for example, fits to a single-DHO form, or to the third-order continued-fraction form, cannot yield a structure with two peaks at nonzero positive frequencies. However, apart from such considerations, we find qualitative, and even semiquantitative, agreement between the spectra obtained using different fitting functions, and shall present only results obtained using the fourth-order continued-fraction form.

The pronounced modification of the frequency spectrum, caused by the applied field, is illustrated in Fig. 2, which compares the spectra obtained with and without the applied field, for two wave vectors. While for the larger wave vector the results are qualitatively similar (although the field shifts the peak position, narrows the peak somewhat, and also reduces the weight in the peak, and enhances the weight in the spectrum at low frequencies), those for $q = 0.2\pi$ indicate that the presence of an applied field induces a qualitatively different structure into the frequency response, and, in particular, two peaks at nonzero frequency are apparent. The way in which the structure changes with wave vector is perhaps best illustrated by a three-dimensional plot, Fig. 3.

The results which we have obtained indicate that the spin dynamics of a ferromagnetically coupled Heisenberg chain are dramatically modified by an applied magnetic field, and can be interpreted in terms of a second magnon mode, which is fairly well defined for small wave vectors q (but still such that $q > \kappa$ the inverse correlation length) but becomes increasingly damped as q increases. In this respect, it should be not-

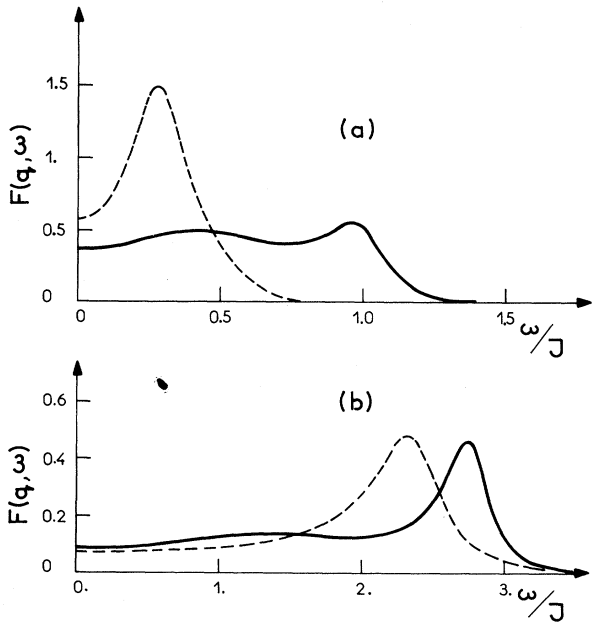

FIG. 2. The frequency spectra of the spin-density correlation function obtained for $h^* = 1.0$ (full line) are compared with those for $h^* = 0.0$ (broken line) for (a) 0.2π and (b) $q = 0.6\pi$.

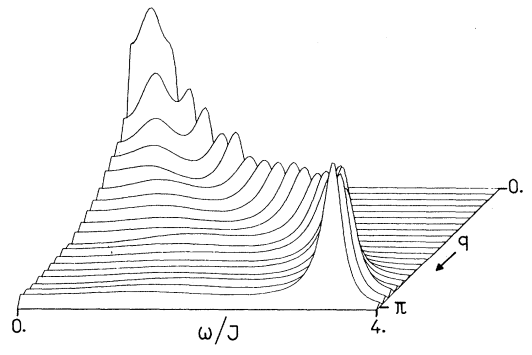

FIG. 3. The frequency spectra of the spin-density correlation function, with an applied field $h^* = 1.0$, plotted as a function of both q and ω. The spectra are for $q = 0.1\pi$ to $q = \pi$ in steps of 0.05π. The vertical scale is arbitrary.

ed that the wave-vector–dependent spin-energy coupling (measured, for example, by the correlation function $\langle E_q S_{-q}^z \rangle$) must fall to zero at the zone boundary. Also, this coupling is much weaker in an antiferromagnetically coupled chain, and one would expect that the second magnon mode would be much weaker, or nonexistent, for this case.[10]

Several authors[11] have discussed the possibility of a second magnon mode in three-dimensional (3D) ferromagnets, based on the idea that, at low temperatures, momentum is approximately conserved, and this additional conservation law admits the possibility of an oscillatory motion of momentum, magnetization, and energy. While the proposed mechanism is suggestive, one should be cautious in assuming that it is necessarily relevant to the 1D magnet for which the physics is, in many respects, fundamentally different from that of 3D magnets. A theoretical study of the dynamics of the classical Heisenberg chain,[12] based on the Mori approach to the generalized Langevin equation, led to results reminiscent of the double-peak structure which we find (see, in particular Fig. 1 of Ref. 12). The structure found was much less marked than the simulation results indicate, but it can be shown[13] that, in the short-time limit, the theory leads to coupled equations of motion for the magnetization and energy densities, with coefficients which are calculated exactly, which yield dispersion relations in satisfactory agreement with those of the two modes found in the computer simulation results. This suggests that the original theory underestimated the relaxation times in the memory-function matrix.

Finally, we turn to the question of whether the second magnon mode could be observed experimentally. The most investigated[1,14,15] quasi-1D ferromagnet is $CsNiF_3$, but this material has a large single-site anisotropy, with an easy plane perpendicular to the c axis. In fact, inelastic neutron scattering measurements on $CsNiF_3$, with an applied field in the easy plane, have been made.[15] The results showed, at low temperatures and with a reduced field $h^* \simeq 0.25$, sharp peaks in the scattering function, with a dispersion relation consistent with the spin-wave result. It is clear that these authors observed only the *transverse* response, and for this component our computer simulation study, for the isotropic system, also shows essentially sharp spin-wave–like oscillations. With the neutron scattering vector in the c direction, the observed cross section has contributions from both the transverse and longitudinal response functions, weighted by the corresponding susceptibilities. Since the ratio of the transverse to longitudinal susceptibility becomes very large in a strong field, very good statistics are needed to see the longitudinal response.

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