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Dynamic Properties of a One-Dimensional Heisenberg Magnet*

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A form for the relaxation shape function for a Heisenberg paramagnet is proposed which has the merit of satisfying certain sum rules and limits. It is shown by comparison with exact infinite-temperature calculations by Carboni and Richards, and inelastic neutron scattering measurements by Hutchings *et al.* on $(CD_3)_4NMnCl_3$, to provide a very good description of an antiferromagnetic linear chain at all temperatures.

Various measurements on one-dimensional magnetic systems[1,2] have created renewed interest in the static and dynamic properties of a Heisenberg linear-chain magnet. Most previous theories have been for either zero or infinite temperature. Exceptions are the exact calculation by Fisher[3] of static two-spin correlation functions for a classical Heisenberg chain, and McClean and Blume's[4] study of dynamic properties, which is based on an integro-differential equation for the relaxation function [of Eq. (4), see below] discussed previously by several authors[5] for simple-cubic Heisenberg magnets. Numerical calculations have been performed for finite-length spin-$\frac{1}{2}$ chains by Carboni and Richards[6] and for long classical Heinsenberg chains by Blume, Watson, and Vineyard.[7] Also, Richards[8] has recently found a spectrum for the collective mode by linearizing the equations of motion in second order. Here we propose a new, simple theory which gives both good agreement with the calculations of Carboni and Richards and with neutron scattering measurements by Hutchings *et al.*[2] on the linear-chain antiferromagnet $(CD_3)_4$-$NMnCl_3$ at temperatures between 1.9 and 40°K. Apart from a multiplicative factor of $\sqrt{\frac{3}{2}}$, Richards's dispersion relation is found to compliment our calculation.

The magnetic energy of the system is described by the Heisenberg exchange Hamiltonian

$$\mathcal{H}=\tfrac{1}{2}J\sum_{i\delta}\vec{S}_i\cdot\vec{S}_{i+\delta}, \tag{1}$$

where J is the exchange parameter between adjacent spins.

The inelastic partial-differential neutron cross section for scattering from a paramagnet with wave-vector change $\vec{k}$ and energy change ω is determined by[9]

$$S(\vec{k},\omega)=(2\pi)^{-1}\int_{-\infty}^{\infty}dt\,e^{-i\omega t}\langle S_{\vec{k}}^{z}(0)S_{-\vec{k}}^{z}(t)\rangle \tag{2a}$$

$$\equiv k_BT\,\chi_{\vec{k}}[\omega\beta/(1-e^{-\omega\beta})]F(\vec{k},\omega), \tag{2b}$$

where $S_{\vec{k}}^{z}=\sum_j \exp(-i\vec{k}\cdot\vec{R}_j)S_j^{z}$ and $T=1/k_B\beta$ is the absolute temperature.

In (2b), $F(\vec{k},\omega)$ is the relaxation shape function,

$$F(\vec{k},\omega)=(2\pi)^{-1}\int_{-\infty}^{\infty}dt\,e^{-i\omega t}[R_{\vec{k}}(t)/R_{\vec{k}}(0)], \tag{3}$$

where the relaxation function $R_{\vec{k}}(t)$ is

$$R_{\vec{k}}(t)=\int_0^{\beta}d\lambda\,\langle S_{\vec{k}}^{z}(-i\lambda)S_{-\vec{k}}^{z}(t)\rangle, \tag{4}$$

and the isothermal susceptibility $\chi_{\vec{k}}=R_{\vec{k}}(0)$. To extract the dominant energy dependence of $F(\vec{k},\omega)$, we adopt Mori's[10] approach and construct a generalized Langevin equation, which for $R_{\vec{k}}(t)$ gives the equation

$$\dot{R}_{\vec{k}}(t)=-\int_0^t d\bar{t}\,K_{\vec{k}}(t-\bar{t})R_{\vec{k}}(\bar{t}). \tag{5}$$

$K_{\vec{k}}(t)$ describes the fluctuating forces in the equation of motion for $S_{\vec{k}}^{z}$ and is akin to the relaxation function (4) but with a different temporal development. However, it satisfies an equation of the same form as (5), which can therefore be regarded as the first in a chain of coupled equations for $R_{\vec{k}}(t)$. In terms of the Laplace transform of the relaxation function, $R_{\vec{k}}(s)$, this chain of equations is equivalent to the continued fraction representation

$$R_{\vec{k}}[s]=\chi_{\vec{k}}\{s+\delta_1/[s+\delta_2/(s+\delta_3/s+\cdots)]\}^{-1}. \tag{6}$$

The static correlation functions δ_i are related to the moments $\langle\omega^n\rangle$ of F; explicitly,

$$\delta_1=\langle\omega^2\rangle,\quad \delta_2=\langle\omega^4\rangle/\langle\omega^2\rangle-\langle\omega^2\rangle.$$

At infinite temperatures we have the exact results[11]

$$\langle\omega^2\rangle_\infty=\Omega_0^{2}(1-\cos ak), \tag{7a}$$

$$\langle\omega^4\rangle_\infty=\Omega_0^{2}\tfrac{1}{2}\langle\omega^2\rangle_\infty[5-3\cos ak-3/4S(S+1)], \tag{7b}$$

where $\Omega_0=2J[S(S+1)/3]^{1/2}$. The k dependence of δ_2 is much weaker than that of δ_1, and δ_3 is a constant to a good approximation.[12] Hence we propose a three-pole approximation for $R_{\vec{k}}[s]$, namely,

$$R_{\vec{k}}[s]=\frac{\chi_{\vec{k}}(s^2+s/\tau+\delta_2)}{s^3+s^2/\tau+s(\delta_1+\delta_2)+\delta_1/\tau}, \tag{8}$$

where the termination function $\tau=(\pi\delta_2/2)^{-1/2}$. The merits of this type of approximation for $R_{\vec{k}}$ to describe density fluctuations in simple classical liquids have been discussed in detail by one of us (S.W.L.).[13] For simple-cubic Heisenberg paramagnets we recover at small k the diffusion constant calculated by Mori and Kawasaki,[9] Windsor,[14] and Blume and Hubbard.[5] The relaxation shape function corresponding to the approximate form (8) for $R_{\vec{k}}[s]$ is

$$\pi F(\vec{k},\omega)=(1/\chi_{\vec{k}})R_{\vec{k}}'[i\omega]$$

$$=\frac{\tau\delta_1\delta_2}{[\tau\omega(\omega^2-\delta_1-\delta_2)]^2+(\omega^2-\delta_1)^2}, \tag{9}$$

which is compared in Fig. 1 for $S=\frac{1}{2}$ to the exact calculations by Carboni and Richards[6] for a nine-spin linear chain. For the smallest wave vector there is excellent (absolute) agreement. For larger k our peak at nonzero ω is more pronounced than that found for the finite chain but the peak positions agree. This good overall agreement gives confidence in our approximate form for $F(\vec{k},\omega)$ at infinite temperatures.

At finite temperatures we estimate the moments $\langle\omega^2\rangle$, $\langle\omega^4\rangle$ by replacing the two- and four-spin correlation functions in the exact expressions[11] by their exact values for a classical

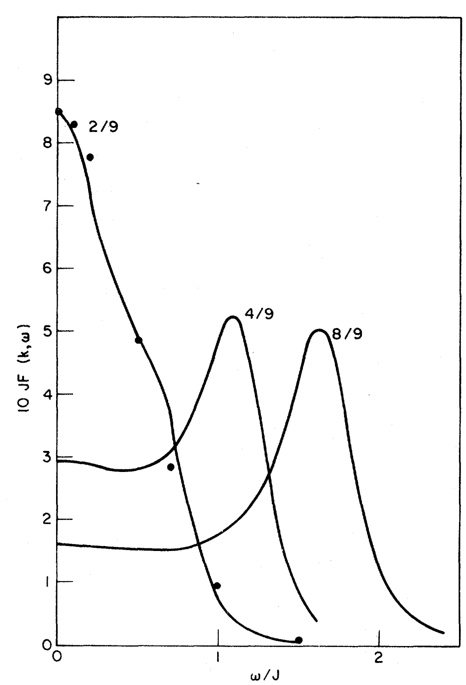

FIG. 1. $10\Omega_0F(\vec{k},\omega)$ at infinite temperature and $S=\frac{1}{2}$, shown as a function of ω for various values of ak/π. The calculations by Carboni and Richards for $ak=2\pi/9$ are represented by $8.5\exp[-2.23(\omega/J)^2]$ and are denoted in the figure by solid circles.

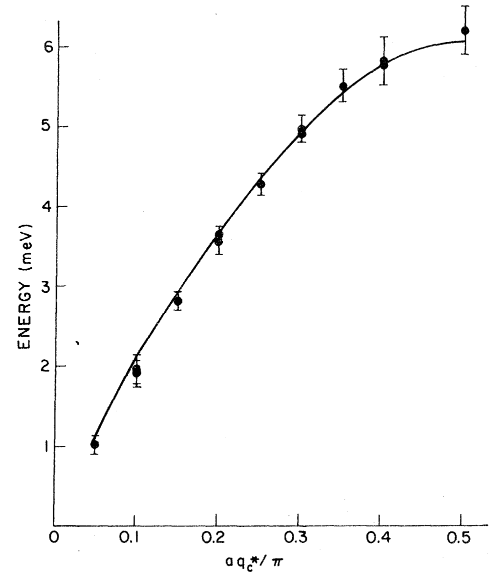

FIG. 2. Energy of the collective excitation at 4.4 °K, as calculated from $S(\vec{k},\omega)$, plotted as a function of the wave-vector displacement from the super-lattice peak. The points represent the measurements of Hutchings *et al.* (Ref. 2) on $(CD_3)_4NMnCl_3$.

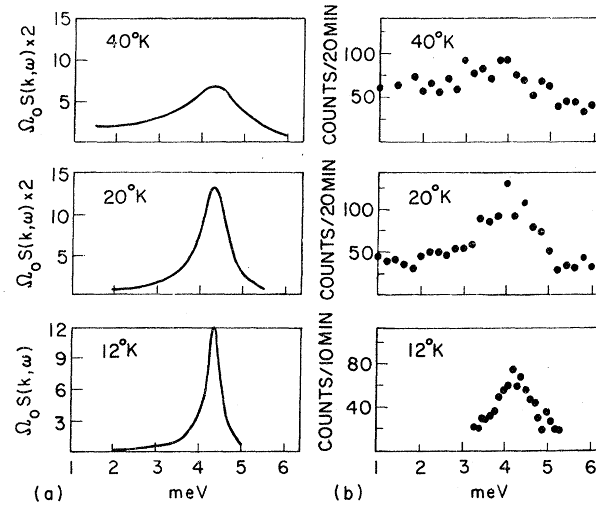

FIG. 3. $\Omega_0 S(\vec{k},\omega)$ for $ak=\frac{3}{4}\pi$, $S=\frac{5}{2}$, and various temperatures, plotted in (a) for comparison with the scattered intensity (b) measured by Hutchings *et al.* for $(CD_3)_4NMnCl_3$. For 20 and 40°K the calculated values have been multiplied by 2 to account for the change in the experimental counting time, and this is given alongside the respective diagrams.

Heisenberg linear chain. The results are

$$\langle\omega^2\rangle = \Omega_0^2(1-\cos ak)(1+u^2+2u\cos ak) \times 3u/K(1-u^2), \tag{10a}$$

$$\langle\omega^4\rangle = \Omega_0^2\tfrac{1}{2}\langle\omega^2\rangle\{5-3\cos ak+v(1-3\cos ak - 3/u)+u[6\cos ak-(2+v)\cos ak]\}, \tag{10b}$$

where $u=\coth K - 1/K$, $v=1-3u/K$, and $K=JS(S+1)\beta$. At infinite temperatures, $\langle\omega^2\rangle$ coincides with the exact quantum result (7a), and $\langle\omega^4\rangle$ differs from (7b) by the term $3/8S(S+1)$ as expected. For $T=0$, we find

$$\langle\omega^2\rangle_0 = 3\Omega_0^2\sin^2 ak, \quad \langle\omega^4\rangle_0/\langle\omega^2\rangle_0 = 3\Omega_0^2\sin^2 ak.$$

Consequently $\delta_2=0$, and $F(\vec{k},\omega)$ consists of a pair of δ functions at $\omega=\pm\omega_k$, where

$$\omega_k = 2J[S(S+1)]^{1/2}|\sin ak|, \tag{11}$$

which is in complete accord with the analysis by Hutchings *et al.*[2] of their measurements on $(CD_3)_4$-$NMnCl_3$ at 1.9 K. In general, a collective excitation, i.e., a maximum in $F(\vec{k},\omega)$ at nonzero ω, is guaranteed when the condition $\delta_1/\delta_2 \geq \frac{1}{2}$ is satisfied.

Figure 2 shows the good agreement between the calculated dispersion of the collective excitation at 4.4°K and the measurements of Hutchings *et al.*[2] on $(CD_3)_4NMnCl_3$. The energy constant $\Omega_0=3.53$ meV, found by using the value $\frac{1}{2}J=6.0$°K. The observed intensity at $ak=\frac{3}{4}\pi$, for temperatures 12, 20, and 40°K, and $S(\vec{k},\omega)$ calculated from (2b) and (9) are compared in Fig. 3 with a favorable result. Comparable agreement between theory and experiment is found at other values of the wave vector.

A similar analysis to that discussed above has been made for three-dimensional paramagnets, and it will be reported in a separate paper. At infinite temperatures we find that $F(\vec{k},\omega)$ is very similar at all wave vectors to that calculated by Blume and Hubbard[5]; indeed at $\omega=0$ there is complete agreement. For finite temperatures (estimating $\langle\omega^2\rangle$ and $\langle\omega^4\rangle$ with the spherical model) we have good agreement with the high-resolution measurements by Tucciarone, Corliss, and Hastings[15] on the simple antiferromagnet $RbMF_3$.

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Investigation of γ-Ray Emission Preceding Isomeric Fission of ^{236}U†

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Measurements were made to detect γ rays preceding isomeric fission in ^{236}U induced by eV-range neutrons captured in ^{235}U. A limit of $\leq 6\times10^{-5}$ was placed on the ratio of the rate of isomeric fission events with prefission γ rays to the rate for prompt fission events. This experiment provides direct evidence that the penetration of the outer barrier is much greater than that for the inner barrier for 3^- and 4^- states in ^{236}U.

About thirty fission isomers have been identified in transuranium nuclei to date. These isomers can be considered as shape isomers associated by Strutinsky[1] with a double-humped fission barrier. This model has provided a satisfactory explanation for many aspects of fission isomerism such as the isomeric half-life, excitation energy of the isomer, and the height of the fission barrier. With the hypothesis of a double-humped fission barrier, the formation of the isomeric state is assumed to proceed in many reactions by penetration of the first barrier from an excited state near the static equilibrium deformation followed by γ decay to the isomeric state in the second minimum. The detection of the prefission γ rays is a vital test of the Strutinsky model. However, these γ rays have not yet been observed. The purpose of this experiment, therefore, was to detect the γ decay to an isomeric state followed by the subsequent fission of this state.

Several experiments[2-5] have identified a fission isomer in ^{236}U with a half-life approximately 100 nsec. Elwyn and Ferguson[4] populated this fission isomer by bombarding ^{235}U with 0.5- and 2.2-MeV neutrons. In the present experiment, a 90-mg ^{235}U sample was bombarded with neutrons from the pulsed neutron source of the Livermore linac in an attempt to detect γ-ray emission prior to fission events corresponding to the 100-nsec ^{236m}U isomer. The neutron energy was determined by the time of flight of the neutrons to the ^{235}U sample located at 14.5 m from the neutron source. Measurements were made for the neutron energy range between 1 and 100 eV. Fission fragments were detected using an ionization chamber. The γ rays were detected by a pair of deuterated-benzene (C_6D_6) scintillators which subtended a fractional solid angle ($\Omega/4\pi$) of approximately 0.8. The outputs of the C_6D_6 scintillators were summed resulting in an efficiency of approximately 15% (including solid angle) with a