Paramagnetic Scattering of Neutrons from a Heisenberg System*

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(Received 9 October 1967)

The neutron paramagnetic scattering function for a Heisenberg system is evaluated in the high-temperature limit and in the cluster model. The numerical results for several different cluster configurations are presented for various values of scattering vector. The scattering function for a simple cubic lattice with spin $\frac{1}{2}$ is compared with the frequency distribution functions derived from the method of moments by de Gennes and by Collins and Marshall.

RECENTLY there has been much work, theoretical as well as experimental, on the scattering of neutrons from paramagnetic media. Van Vleck[1] introduced the method of moments in the frequency distribution function to calculate the form factor for forward scattering of neutrons by paramagnetic media. This work was later related by de Gennes[2] to the time-dependent spin-correlation functions introduced by Van Hove.[3] Collins and Windsor[4] were able to evaluate the scattering function for an Ising system from the spin dynamics at high temperatures, and their results for a simple cubic lattice with spin $\frac{5}{2}$ checked favorably with the de Gennes's Gaussian distribution derived from the method of moments. Still later, Collins and Marshall[5] modified de Gennes's Gaussian distribution by expanding in terms of moments of frequency up to fourth moment for the Heisenberg Hamiltonian and for the high temperature. Their new expression for the scattering function showed an improvement over the Gaussian distribution for the Ising system evaluated previously.[4]

Inasmuch as an Ising system represents an idealized mathematical model, while a Heisenberg model is generally thought to describe paramagnetic insulators reasonably well, a reasonable test for the neutron-scattering theory seems to lie in a Heisenberg system. This paper aims to provide this test by evaluating the neutron-scattering function for a Heisenberg paramagnet in a manner different from the method of moments advanced hitherto.

The Heisenberg Hamiltonian per pair of atoms in a paramagnetic insulator is given by

$$H=-2J\sum_{i<j}\mathbf{S}_i\cdot\mathbf{S}_j\cong-2J\mathbf{S}_0\mathbf{S}_1, \tag{1}$$

where J is the exchange integral, $\mathbf{S}_i$ is the spin operator for the ith atom, and the second expression on the right is the result of the cluster-model approximation.[6] Here $\mathbf{S}_0$ is the spin of the central atom and $\mathbf{S}_1$ is the total spin of the z nearest-neighbor atoms to the central atoms to the central atom (that these nearest neighbor atoms are not nearest neighbors to each other is assumed). The cluster model as described by Eq. (1) reduces a Heisenberg system effectively to a two-spin system, and for a two-spin system there exists a complete quantum-mechanical description. That is, the eigenvalue problem can be stated as

$$H\mid s_0s_1sm\rangle=E(s_0s_1sm)\mid s_0s_1sm\rangle, \tag{2}$$

with the eigenvalues given by

$$E(s_0s_1sm)=-J[s(s+1)-s_1(s_1+1)-s_0(s_0+1)], \tag{3}$$

where s_0 is the total spin of the central atom, s_1 can assume the values of 0, 1, 2, $\cdots$, zs_0, and s takes on quantized values between $\mid s_0-s_1\mid$ and s_0+s_1. The eigenfunctions $\mid s_0s_1sm\rangle$ are given in terms of individual spinors by the relation[7]

$$\mid s_0s_1sm\rangle=\sum_{m_0m_1}\mid s_0s_1m_0m_1\rangle\langle s_0s_1m_0m_1\mid s_0s_1sm\rangle, \tag{4}$$

where m_0, m_1, and m are the magnetic quantum numbers corresponding, respectively, to s_0, s_1, and s.

The magnetic neutron cross section for a rigid crystal at high temperatures, according to Van Hove,[3] can be written as

$$d^2\sigma/d\Omega d\omega=A(k_i,k_f)\,S(\mathbf{K},\omega), \tag{5}$$

where $A(k_i, k_f)$ is a known function of scattering parameters and $S(\mathbf{K}, \omega)$ is the scattering function defined by

$$S(\mathbf{K},\omega)=[2\pi s_0(s_0+1)]^{-1}\times\int_{-\infty}^{\infty}dte^{-i\omega t}\sum_R\exp(i\mathbf{K}\cdot\mathbf{R})\,\langle\mathbf{S}_0(0)\cdot\mathbf{S}_R(t)\rangle. \tag{6}$$

Here $\mathbf{K}=\mathbf{k}_f-\mathbf{k}_i$ and $\omega=E_f-E_i$ are the momentum

* Supported in part by the National Science Foundation.
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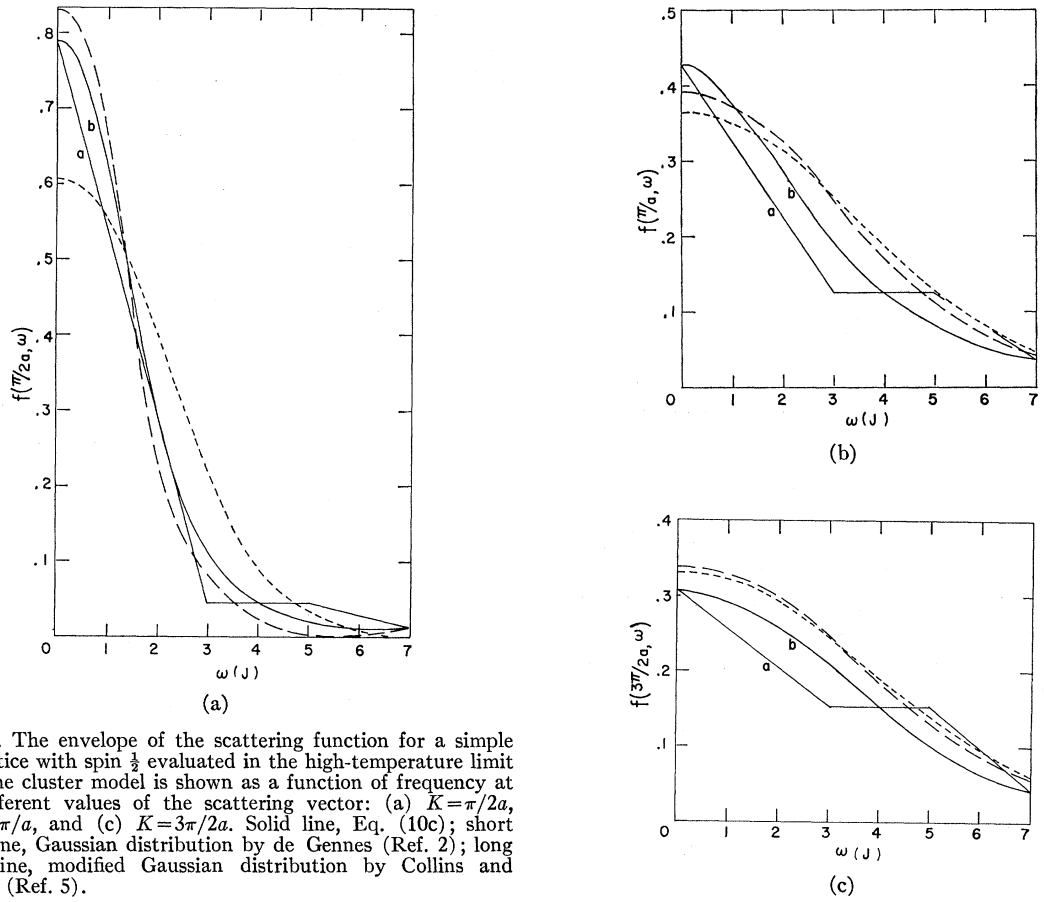

FIG. 1. The envelope of the scattering function for a simple cubic lattice with spin $\frac{1}{2}$ evaluated in the high-temperature limit and in the cluster model is shown as a function of frequency at three different values of the scattering vector: (a) $K=\pi/2a$, (b) $K=\pi/a$, and (c) $K=3\pi/2a$. Solid line, Eq. (10c); short dashed line, Gaussian distribution by de Gennes (Ref. 2); long dashed line, modified Gaussian distribution by Collins and Marshall (Ref. 5).

and energy transfers from neutron to crystal in a system of units for which $\hbar$ is unity, and the brackets around the spin vectors signify thermal average. In the cluster model under consideration Eq. (6) may be rewritten in a more workable form as

$$S(\mathbf{K},\omega)=[Zs_0(s_0+1)]^{-1}\sum_{ij}\exp(-E_i/k_BT)\times[S_{ij}(0)+S_{ij}(\mathbf{K})]\delta(\omega+E_i-E_j), \quad (7)$$

with

$$S_{ij}(0)=|\langle i|S_0^z(0)|j\rangle|^2+\tfrac{1}{2}\langle i|S_0^+(0)|j\rangle\times\langle j|S_0^-(0)|i\rangle+\tfrac{1}{2}\langle i|S_0^-(0)|j\rangle\langle j|S_0^+(0)|i\rangle,$$

$$S_{ij}(\mathbf{K})=[(\sin Ka)/Ka]\{\langle i|S_0^z(0)|j\rangle\langle j|S_1^z(0)|i\rangle+\langle i|S_0^+(0)|j\rangle\langle j|S_1^-(0)|i\rangle\}. \quad (8)$$

The states i and j are the stationary states determined

TABLE I. The amplitude of the δ-function singularities $f(\mathbf{K},\omega)$ of the scattering function $S(\mathbf{K},\omega)$ evaluated for several different cluster configurations.

Cluster configuration	ω	$f(0,\omega)$	$f(\pi/2a,\omega)$	$f(\pi/a,\omega)$	$f(3\pi/2a,\omega)$
$z=2$ linear chain	0	1.00	0.839	0.556	0.461
	$\pm3J$	0	0.081	0.222	0.270
$z=4$ quadratic layer	0	1.00	0.807	0.467	0.357
	$\pm3J$	0	0.060	0.167	0.202
	$\pm5J$	0	0.049	0.010	0.121
$z=6$ simple cubic	0	1.00	0.793	0.428	0.307
	$\pm3J$	0	0.045	0.125	0.152
	$\pm5J$	0	0.045	0.125	0.152
	$\pm7J$	0	0.013	0.036	0.043

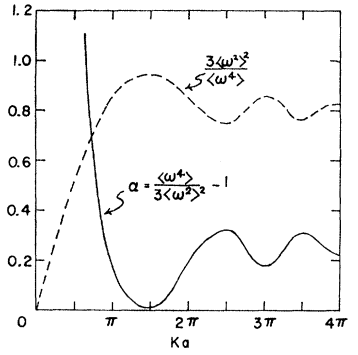

FIG. 2. The parameter $\alpha=[(\langle\omega^4\rangle/3\langle\omega^2\rangle^2)-1]$ from second and fourth moments is shown as a function of scattering vector for a simple cubic lattice with spin $\frac{1}{2}$.

in Eq. (2), and Z is the partition function. The scattering vector was averaged over angular variables as it appears in Eq. (8). In the high-temperature limit the Boltzmann factor in Eq. (7) reduces to unity and the partition function merely represents the number of states for the system including the degeneracy of cluster quantum number s_1 given by the expression (for $s_0=\frac{1}{2}$)

$$\omega(s_1)=\frac{(2n)!}{(n-s_1)!(n+s_1)!}-\frac{(2n)!}{(n-s_1-1)!(n+s_1+1)!}, \tag{9}$$

where $2n=z$.

The scattering function as given by Eq. (7) is evaluated in the high-temperature limit for several different cluster configurations. These results are summarized as follows:

For $z=2$ (linear chain)

$$S(\mathbf{K},\omega)=\tfrac{1}{9}[5+4((\sin Ka)/Ka)]\delta(\omega)+\tfrac{2}{9}[1-((\sin Ka)/Ka)]\delta(\omega\pm 3J); \tag{10a}$$

for $z=4$ (quadratic layer)

$$S(\mathbf{K},\omega)=\tfrac{1}{15}[7+8((\sin Ka)/Ka)]\delta(\omega)+\tfrac{1}{6}(1-((\sin Ka)/Ka)]\delta(\omega\pm 3J)+\tfrac{1}{10}[1-((\sin Ka)/Ka)]\delta(\omega\pm 5J); \tag{10b}$$

for $z=6$ (simple cubic)

$$S(\mathbf{K},\omega)=\tfrac{1}{21}[9+12((\sin Ka)/Ka)]\delta(\omega)+\tfrac{1}{8}[1-((\sin Ka)/Ka)]\delta(\omega\pm 3J)+\tfrac{1}{8}[1-((\sin Ka)/Ka)]\delta(\omega\pm 5J)+\tfrac{1}{28}[1-((\sin Ka)/Ka)]\delta(\omega\pm 7J). \tag{10c}$$

The numerical results for the coefficients of the δ functions in Eq. (10), $f(K,\omega)$, for various values of K are tabulated in Table I. The result for a simple cubic lattice with spin $\frac{1}{2}$ is compared graphically in Fig. 1 with the results derived from the method of moments—the Gaussian distribution by de Gennes[2] and its modification by Collins and Marshall.[5] Here in Fig. 1 curve a represents connection of points with straight lines, while curve b represents the smoothed-out version of curve a. The unsmoothness of curve a, argues Van Vleck,[1] is due to the over-simplified character of the idealized cluster model; namely, all states with same s (total spin quantum number) have the same energy with a high degree of degeneracy given by Zeeman factor $2s+1$ plus the degeneracy of s_1 (cluster quantum number) given by Eq. (9). If, however, the influence of the atoms outside the cluster is taken into account, this degeneracy will be lifted—resulting in a "smearing-out" of these states and thus in a "smoothing-out" of curve a not too different from curve b.

Several observations can be made from Table I and Fig. 1. As the scattering vector K increases, the frequency distribution changes its shape from that of Lorentzian to that of Gaussian, in complete agreement with de Gennes[2] and with Collins and Marshall.[5] Furthermore, the modification by Collins and Marshall appears to give a better description for small K, but for large K the correction seems negligible. This seems natural from the fact that (a) the factor $\langle\omega^4\rangle/3\langle\omega^2\rangle^2$ is not sensibly different from unity for large K ($K\geq\pi$), as pointed out by de Gennes, and (b) the parameter $\alpha=[(\langle\omega^4\rangle/3\langle\omega^2\rangle^2)-1]$ determines the effect of the correction term due to Collins and Marshall (see Fig. 2).