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Dynamical Spin Correlations in a Heisenberg Spin Cluster*

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The time- and temperature-dependent spin-spin correlation functions and their wave-vector-dependent frequency transforms are obtained exactly from the Wigner-Eckart theorem in the Bethe-Peierls-Weiss cluster model of Heisenberg spin systems. The expressions obtained are valid for *all* spin values and for *all* temperatures above the transition temperature. The present results for the spin-$\frac{5}{2}$ simple-cubic lattice are compared with the theories of Windsor and of Blume and Hubbard and with the experimental data on $RbMnF_3$.

It has been possible to study dynamical properties of Heisenberg spin systems in terms of time- and temperature-dependent spin-spin correlations and their wave-vector-dependent frequency transforms which are numerically evaluated in the Bethe-Peierls-Weiss (BPW) cluster model for various spin values and temperatures.[1,2] This paper reports exact analytic expressions for these quantities obtained from the Wigner-Eckart theorem and valid for *all* spin values and for *all* temperatures above the transition temperature. The present results for the spin-$\frac{5}{2}$ sc lattice are compared with other theories[3-5] and with $RbMnF_3$ data.[6]

The two quantities of interest in this paper are the dynamical spin correlation function defined by

$$\langle S_0^{\varepsilon}(0) S_n^{\varepsilon}(t)\rangle = C\sum_{ij} e^{-\beta E_i}\,\langle i|S_0^{\varepsilon}|j\rangle\,\langle j|S_n^{\varepsilon}|i\rangle\, e^{i(E_j-E_i)t}, \tag{1}$$

and the wave-vector-dependent frequency transform given by

$$S(\vec{k},\omega) = \sum_n e^{i\vec{k}\cdot\vec{n}} \int_{-\infty}^{\infty} \frac{dt}{2\pi}\, e^{-i\omega t}\,\langle S_0^{\varepsilon}(0) S_n^{\varepsilon}(t)\rangle\,, \tag{2}$$

where $C = 1/s_0(s_0+1)Z$, with Z being the partition function and s_0 the spin value per atom. These quantities can be evaluated exactly in the BPW cluster model of Heisenberg spin systems, which, for temperatures above the transition temperature, is characterized by the effective-spin Hamiltonian[7]

$$H = -J\vec{S}_0\cdot\vec{S}_1\,, \tag{3}$$

where $\vec{S}_0$ represents the central spin of a cluster and $\vec{S}_1$ the total effective spin for the γ_0 nearest neighbors surrounding the central spin. This Hamiltonian is diagonal in a representation characterized by $|s_0 s_1 s m\rangle$, in which S_0^2, S_1^2, S^2, and S^{ε} are diagonal. In this representation the matrix elements of the spin operators that appear in Eq. (1) can be obtained exactly from the Wigner-Eckart theorem,[8] and the nonvanishing elements of these are

$$\begin{aligned}
&\langle s|S_0^{\varepsilon}|s\rangle\langle s|S_n^{\varepsilon}|s\rangle = [s(s+1)+P(s_1)]\\
&\qquad\times[s(s+1)\pm P(s_1)](2s+1)/12s(s+1)\,,\\
&\langle s|S_0^{\varepsilon}|s+1\rangle\langle s+1|S_n^{\varepsilon}|s\rangle = \pm Q(s_1,s)\,,\\
&\langle s|S_0^{\varepsilon}|s-1\rangle\langle s-1|S_n^{\varepsilon}|s\rangle = \pm Q(s_1,s-1)\,,
\end{aligned} \tag{4}$$

where the plus sign goes with $n=0$ and the minus sign with $n=1$ and where

$$\begin{aligned}
P(s_1) &= s_0(s_0+1)-s_1(s_1+1)\,,\\
Q(s_1,s) &= (s_0+s_1+2+s)(s_0+s_1-s)\\
&\quad\times(s_1-s_0+1+s)(s_0-s_1+1+s)/12(s+1)\,.
\end{aligned}$$

From Eqs. (1) and (4) it follows that

$$\begin{aligned}
\langle S_0^{\varepsilon}(0)S_n^{\varepsilon}(t)\rangle = C\sum W(s_1)\exp\{\tfrac{1}{2}J[s(s+1)-s_1(s_1+1)]\beta\}\\
\times\{[s(s+1)+P(s_1)][s(s+1)\pm P(s_1)]\\
\times(2s+1)/12s(s+1)\pm e^{-i(s+1)Jt}Q(s_1,s)\\
\pm e^{iJst}Q(s_1,s-1)\}\,,
\end{aligned} \tag{5}$$

where the summations are over s_1 and s, each in the range of values given by $0\le s_1\le\gamma_0 s_0$ and $|s_0-s_1|\le s\le(s_0+s_1)$. The quantity $W(s_1)$ represents the multiplicity of s_1 values.[7]

A more relevant quantity of interest, directly accessible by inelastic neutron scattering experiments, is the frequency transform given by Eq. (2), which now becomes

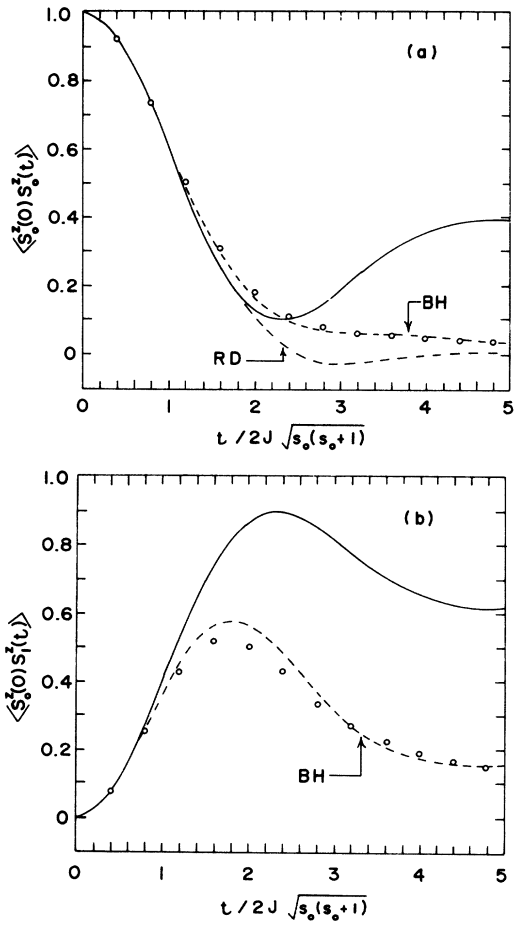


FIG. 1. The autocorrelation (a) and the nearest-neighbor correlation (b) predicted by Eq. (5) at infinite temperature is shown in solid curves in comparison with the computer simulation calculations due to Windsor denoted by open circles and with the theories of Blume and Hubbard (BH) and of Resibois and DeLeener (RD).

$$S(\vec{k},\omega) = (C/\gamma_0)\sum W(s_1)\exp\{\tfrac{1}{2}J[s(s+1)-s_1(s_1+1)]\beta\}$$
$$\times\{\delta(\omega)[s(s+1)+P(s_1)](\gamma_0[s(s+1)+P(s_1)]$$
$$+\gamma_k[s(s+1)-P(s_1)])(2s+1)/12s(s+1)$$
$$+(\gamma_0-\gamma_k)[\delta(\omega+(s+1)J)Q(s_1,s)$$
$$+\delta(\omega-sJ)Q(s_1,s-1)]\}, \quad (6)$$

where $\gamma_k = \sum e^{i\vec{k}\cdot\vec{n}}$ summed over nearest neighbors.

In Fig. 1 the autocorrelation $\langle S_0^z(0)S_0^z(t)\rangle$ and the nearest-neighbor correlation $\langle S_0^z(0)S_1^z(t)\rangle$ predicted by Eq. (5) are shown in solid curves in comparison with the predictions of other theories. The open circles represent the computer simulation calculations due to Windsor,[3] while the dashed curves represent the calculations made by Blume and Hubbard (BH) in their self-consistent-field approach[4] and by Resibois and DeLeener (RD) in their kinetic equation approach.[5] For short times, $t \lesssim 4J[s_0 \times (s_0+1)]^{1/2}$, the predictions of a simple-cluster model appear to be in good agreement with those of other theories, but for longer times the agreement seems less impressive. The autocorrelation in a small cluster tends to oscillate even after a long time, clearly indicating the lack of a damping mechanism in the model, while the nearest-neighbor correlation oscillates in phase with other theories but with higher amplitudes. It is interesting to note that in the present model the sum rule

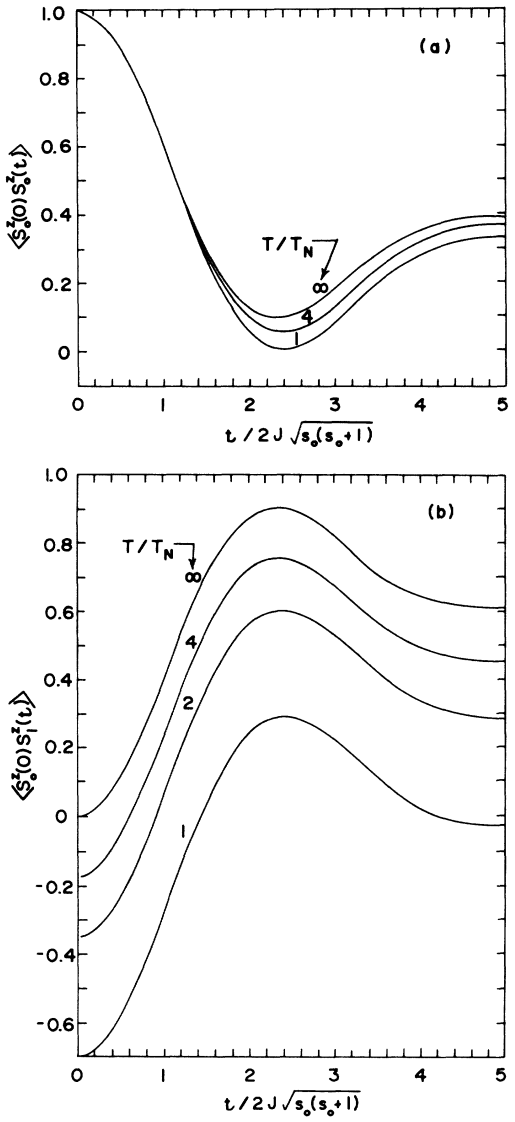
$$\sum_n \langle S_0^z(0) S_n^z(t)\rangle = 1 \quad (7)$$

is strictly conserved at any given time t in the infinite-temperature limit, in spite of the fact that the sum includes only two terms, the self-correlation and the nearest-neighbor correlation. It is for this very reason that the nearest-neighbor correlation oscillates with higher amplitude in the present model than in other models. This may be interpreted as a self-consistent character of the model, in which the nearest-neighbor correlation bears the burden of the more distant neighbors

The temperature dependence of these time correlations are depicted in Fig. 2. The autocorrelations in Fig. 2(a) and the nearest-neighbor correlations in Fig. 2(b) are shown as a function of time for several different values of T/T_N for a typical Heisenberg antiferromagnet. Although the amplitudes of the nearest-neighbor correlations vary greatly with temperature, very little variation is observed in the structures of the self-correlation or the nearest-neighbor correlation. It is, however, more interesting to note the temperature dependence of the sum of these correlations as shown in Fig. 2(c). As stated in Eq. (7), the sum is normalized to unity at infinite temperature, and it is seen to fall off to about 0.3 as the temperature is decreased to T_N.

Comparison with experiments can be made through the frequency transform, to which the inelastic neutron scattering differential cross section is proportional.[9] In Fig. 3 the predictions of Eq. (6) are compared with the $RbMnF_3$ data for three different temperatures and at a fixed value of wave vector.[6] The solid curves represent smooth envelopes of δ-function singularities given in Eq. (6), and the dashed lines near the zero-energy transfer show that the elastic peaks are off the scale. Since the experimental data give only relative counting rates, the theoretical curves are normalized to the experimental values at one point, namely, $\omega = 1.85$ meV.

An interesting feature seen from Fig. 3 is that the simple theory exhibits, in agreement with experiments, a clear resolution in the neutron spectra—an elastic peak at center and two inelastic peaks on both sides. Although the agreement is



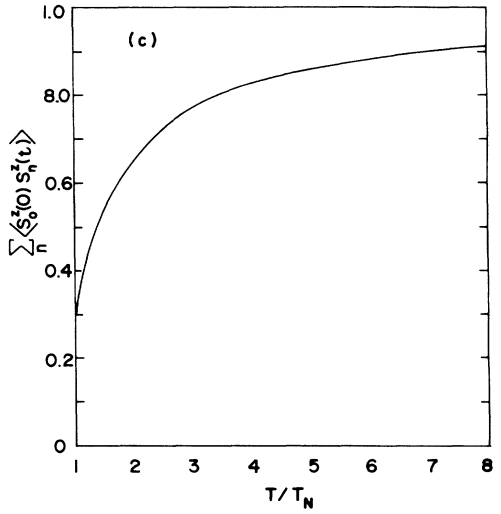


FIG. 2. The autocorrelations (a) and the nearest-neighbor correlations (b) are shown as a function of time for several different values of T/T_N. In (c) the sum of these correlations at any given time t is shown as a function of T/T_N for a typical Heisenberg antiferromagnet.

rather reasonable near T_N, it becomes poorer as the temperature is increased to 84 °K. The reason for this behavior is that even though the frequency transform $S(\vec{k}, \omega)$ was obtained from only two terms in the sum [see Eq. (7)], the fact that the sum rule is obeyed at any given time t is an indication that the effect of more distant neighbor correlations are approximately taken into account in the sum. The persistence of structure in the theory, especially near the zero-energy transfer, even at high temperatures is most probably due to the lack of a spin-diffusion mechanism in the cluster model, the diffusion process being a long-time phenomenon over wide range of lattice space. This is a reflection of the poor long-time behavior in the time correlations shown in Fig. 1, resulting from an over-simplified character of the cluster model. If the influence of the spins outside the cluster were somehow taken into account, the infinitely sharp elastic peaks would be Lorentz broadened. The model, nevertheless, exhibits

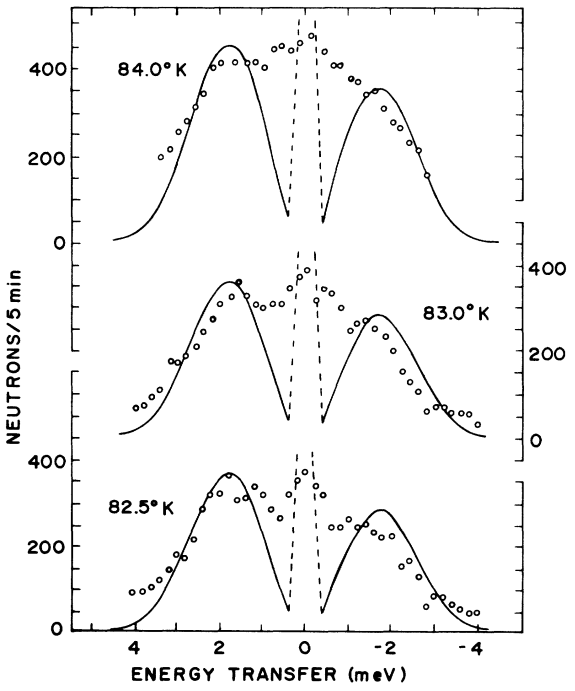


FIG. 3. Frequency spectra predicted by Eq. (6) are shown in solid curves in comparison with the inelastic neutron scattering data on $RbMnF_3$ shown in open circles. The dashed lines near the zero-energy transfer show that the elastic peaks are off the scale.

correct symmetry requirement at the infinite-temperature limit, namely, $S(\vec{k}, \omega) = S(\vec{k}, -\omega)$. Also in this temperature limit the cluster model $S(\vec{k}, \omega)$ gives the correct second frequency moment as first predicted by de Gennes for a complete Heisenberg system.[10]

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Spin Fluctuations in Plutonium and Other Actinide Metals and Compounds*

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It is proposed that the anomalous behavior of the resistivities of Pu and Np and some actinide intermetallic compounds can be explained on the basis of spin fluctuations in narrow 5*f* bands. An initial T^2 increase is observed in the resistivities of all these systems. From radiation-damage data we conclude that there are two distinct regions of 5*f*-electron behavior: (i) a high-temperature region ($\gtrsim 100$ °K), where the 5*f* electrons occupy virtual bound states and the metal resembles a disordered alloy with magnetic impurities such as Pd(U), and (ii) a low-temperature region, where the 5*f* bands are hybridized and a well-defined Fermi surface is formed. Anomalies observed in several properties of Pu at 60 °K may well reflect the temperature at which well-defined 5*f* bands begin to form.

I. INTRODUCTION

The resistivites of several actinide metals exhibit anomalous behavior which has not been adequately explained.[1] There are maxima in the resistivity-temperature curves for α-neptunium and all of the allotropic phases of plutonium stabilized below room temperature, as well as resistivity minima for stabilized δ-Pu above room temperature. However, the magnetic susceptibilities of these phases are nearly temperature independent. We have now observed qualitatively similar behavior in UAl_2 and $PuAl_2$ which form the $MgCu_2$-type cubic Laves phase. We believe, and will try to show, that in all these materials the primary scattering mechanism is spin-flip scattering from paramagnons in fairly narrow 5*f* bands.

Explanations of the α-Pu properties have been based on (i) a subtle phase change at 60 °K, (ii) interband scattering combined with band-structure effects,[2] and (iii) antiferromagnetic ordering near 60 °K.[3,4] The phase change hypothesis is unlikely because of the lack of diffraction evidence[5] or hystereses in physical properties near 60 °K. Interband scattering proposed by Smoluchowski[2] is probably part of the cause for the rapid increase in the resistivity above ~10 °K. However, the explanation of the negative resistivity-temperature slope at higher temperatures in terms of a particular value for the curvature of the density of states does not seem plausible since very similar values would be required for all three allotropic phases of plutonium (monoclinic α, body-centered-monoclinic β, and face-centered-cubic δ), the intermetallic compounds, and orthorhombic neptunium metal. Also the magnetic susceptibilities fail to show the temperature dependence expected from that band picture.

The nearly magnetic behavior of Pu, as shown by the large magnetic susceptibility, plus the expectation of narrow 5*f* bands caused speculation to center primarily on the existence of antiferromagnetism. A number of studies (specific heat,[6] radiation damage,[7] magnetoresistivity,[1] thermoelectric power,[8] elastic constants,[9] Hall effect[10]) have weakly supported the hypothesis of an antiferromagnetic transition. A detailed discussion of such a transition in α-Pu by Rocher[4] is based on a 5*f*