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Short-Range-Order Effects on the Frequency Moments of Heisenberg Spin Systems with Uniaxial and Exchange Anisotropy

Frederick A. Malinoski*
Frankford Arsenal, Philadelphia, Pennsylvania 19137

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The first-order temperature corrections to the zeroth and second, longitudinal and transverse wave-vector-dependent frequency moments are calculated for a Heisenberg spin system with uniaxial and exchange anisotropy and with an arbitrary interaction range. These moments are applied to several lattices including the hexagonal close-packed structure of terbium, which possesses a large uniaxial anisotropy constant. The calculated energy transfer of magnetically scattered neutrons is shown to be in good agreement with experimental data for terbium and also gives evidence of a significant short-range-order effect.

I. INTRODUCTION

The method of wave-vector-dependent frequency moments[1-5] has been successfully used in studying the dynamical properties of Heisenberg spin systems by means of approximate phenomenological representations of the spectral line shapes of spin correlations. The knowledge of the three lowest-order nonvanishing, frequency moments (zeroth, second, and fourth) establishes a phenomenological approximation based on a two-parameter Gaussian diffusivity. However, the knowledge of only the zeroth and second moments is still useful in that contact can be made with the energy distributions of neutrons, scattered inelastically by pertinent magnetic materials.

Most of the frequency-moment calculations have been conducted at elevated temperatures,[1-3,5] where the spin system can be considered to be almost completely random. As the temperature is lowered, the component spins of the magnetic system experience the presence of short-range order, e.g., as exhibited by the frequency wave-vector-dependent susceptibilities measured by inelastic neutron scattering experiments. Since these experiments are often performed at temperatures not much higher than several times the magnetic critical temperatures for strongly exchange-coupled systems, the existence of the short-range order could be expected to have significant effects.

For an isotropic Heisenberg paramagnet, Tahir-Kheli and McFadden[4] have computed the temperature dependence of the frequency moments. The series expansion of the moments in powers of T_C/T was calculated to the order $(T_C/T)^4$ for the zeroth moment, $(T_C/T)^3$ for the second moment, and $(T_C/T)^2$ for the fourth moment, and were specialized to the case of a simple cubic lattice. In addition, for a spin system isotropic in the exchange coupling but with uniaxial anisotropy, i.e., a static crystalline field, Sears[6] has calculated the zeroth and second moments to the order equivalent to $(T_C/T)^2$.

Terbium and other rare earths are examples of uniaxially anisotropic spin structure and have been the subjects of experimental investigation, particularly neutron scattering studies.[7-10] Paramagnetic exchange broadening in terbium, observed by Cable *et al.*,[10] showed that the uniaxial anisotropy energy yielded a significant contribution to the widths of the energy distribution of magnetically scattered neutrons. Moreover, since the experiment was performed at a temperature approximately three times the Néel temperature, a significant departure in the broadening from that calculated on the basis of infinite temperature indicated the presence of short-range order in the spin system. The study by Sears[6] showed that much of the departure of the infinite-temperature theory from the experimental results could be eliminated by the first-order temperature correction to the second moment. However, a discrepancy in the second moment of Sears has been reported in Ref. 4 and is also noted in this study.

The principal objectives of this paper are to derive and assess the first-order temperature corrections of the zeroth and second, longitudinal and transverse frequency moments of a Heisenberg spin system with both exchange and uniaxial (crystal-field) anisotropy and to investigate the importance of the aforementioned discrepancy in the second moments, in particular, as applied to the available data on the energy of scattered neutrons in terbium.

The organization of this paper is as follows: In Sec. II the basic concept of the frequency moments is formulated, and a description of the Hamiltonian of the spin system considered is presented. In Sec. III the first-order temperature corrections to the moments are calculated in a generic form with exchange and uniaxial anisotropy and with an arbitrary range of the exchange interactions. These moments are then particularized in Sec. IV to the hexagonal-close-packed (hcp) lattice of terbium with nearest-neighbor isotropic exchange. Section V contains a discussion of the derived results and some final statements. In Appendix A, some details of the calculation of the exchange terms in the hcp structure are presented. Appendix B contains the anisotropic frequency moments particularized to several other lattice structures, the linear chain, the two-dimensional net, and the simple cubic lattice, with next-nearest-neighbor interactions.

II. FORMULATION

The Hamiltonian of the anisotropic Heisenberg spin system can be written in the form

$$H=-\sum_{gw}\left[I_{+}(gw)S_g^{+}S_w^{-}+I_0(gw)S_g^{z}S_w^{z}\right]-A\sum_{j}(S_j^{z})^2 . \tag{2.1}$$

This Heisenberg model assumes both exchange anisotropy, i.e., $I_{+}\neq I_0$, and uniaxial anisotropy by the A term. Here $I(gw)$ is the exchange interaction integral between two spins at sites g and w. The z-z exchange, I_0, is considered to be distinct from the x-x or y-y exchange, I_{+}. The operator S_g^{α} is the α component of the spin vector associated with the lattice point g, and A is the uniaxial anisotropy constant, characterizing the strength of the crystalline field. This latter type of anisotropy is also referred to as axial, crystal-field, or single-ion anisotropy. The exchange integrals are assumed to depend only upon the spatial separation of the sites and to vanish as the separation goes to zero. The range of the exchange interactions includes next nearest neighbors. The spacing of the spins is assumed to be uniform for the one-dimensional linear chain, the two-dimensional simple net, and the three-dimensional simple cubic lattices. Periodicity in the boundary conditions is considered to be applicable, as is the absence of all magnetovibrational contributions to the scattering. The Dirac system of units with $\hbar=1$ is used throughout this study.

The space-time-dependent spin correlation function is defined as

$$F^{\alpha\alpha'}(g-w,\,t-t')=\langle[S_g^{\alpha}(t),\,S_w^{\alpha'}(t')]\rangle , \tag{2.2}$$

where α and $\alpha'=x, y, z$. Because cylindrical symmetry is assumed, correlations with $\alpha\neq\alpha'$ vanish and the transverse correlations with $\alpha=x$ and with $\alpha=y$ are equivalent. In the general anisotropic case, the longitudinal correlations with $\alpha=z$ differ from the transverse correlations. The time dependence of the spin operators is in the Heisenberg representation with respect to the Hamiltonian. The angular brackets denote a statistical thermal average over a canonical ensemble, and the straight brackets denote a commutator.

The wave-vector-dependent frequency moments can be defined[5] by determining the time derivatives of a Fourier representation of the spin correlation function. These moments are

$$\langle\omega^{n+1}\rangle_{\vec{K}}^{\alpha\alpha}=\sum_{\vec{g}-\vec{w}}e^{-i\vec{K}\cdot(\vec{g}-\vec{w})}\left[\left(i\frac{d}{dt}\right)^{r}\left(-i\frac{d}{dt'}\right)^{n-r}\right.$$

$$\times F^{\alpha\alpha}(g-w,\, t-t')\Big]_{t=t'}\,, \qquad (2.3)$$

where $\vec{\mathrm{K}}$ is the wave vector, the indices n and r are non-negative integers such that $n \geq r$, and the sum over all position vectors $\vec{\mathrm{g}}$-$\vec{\mathrm{w}}$ includes the origin. It has been shown that the odd moments, i.e., $\langle\omega\rangle_{\vec{\mathrm{K}}}^{\alpha\alpha}$, $\langle\omega^3\rangle_{\vec{\mathrm{K}}}^{\alpha\alpha}$, etc., vanish, by considering their alternate, more basic definition in terms of the spectral function, which is the space-time Fourier representation of the spin correlation function.[1-5]

The zeroth moment or wave-vector-dependent susceptibility is not determined by Eq. (2.3), but involves a special case calculated in Ref. 3. The interchangeability of the symmetric spatial variables g and ω in the correlation function and the statistical mechanical identity,

$$\langle B(t)C(t')\rangle = \langle C(t'-i\beta)\,B(t)\rangle\,, \qquad (2.4)$$

where $\beta = (k_B T)^{-1}$, $k_B =$ Boltzmann's constant, and $T =$ absolute temperature, lead to the following result for the zeroth moment:

$$\langle\omega^0\rangle_{\vec{\mathrm{K}}}^{\alpha\alpha} = \sum_{\vec{\mathrm{g}}-\vec{\mathrm{w}}} e^{i\vec{\mathrm{K}}\cdot(\vec{\mathrm{g}}-\vec{\mathrm{w}})} \int_0^{\beta} d\mu \langle S_g^{\alpha}(0)\, e^{-\mu H} S_w^{\alpha}(0)\, e^{\mu H}\rangle\,. \qquad (2.5)$$

It is noted that the spin operators in Eqs. (2.3) and (2.5) are specified for equal times.

The method for the computation of the frequency moments involves first the application of the equation of motion of the spin operators,

$$i\frac{d}{dt} S_g^{\alpha}(t) = [S_g^{\alpha}(t),\, H]\,. \qquad (2.6)$$

The time-independent statistical thermal averages $\langle Q\rangle$, where Q is a generalized spin operator product or commutator, are computed by the usual high-temperature expansion procedure with the density matrix $\rho = e^{-\beta H}$. Then the generalized thermal average, to second order in inverse temperature (i.e., to the first-order temperature-correction term), is

$$\begin{aligned}\langle Q\rangle &= \frac{\mathrm{Tr}\{e^{-\beta H} Q\}}{\mathrm{Tr}\{e^{-\beta H}\}} \\ &= \mathrm{Tr}\{Q - \beta HQ + \tfrac{1}{2}\beta^2 H^2 Q - \cdots\}\bigg(1 + \frac{\mathrm{Tr}\{\beta H\}}{\mathrm{Tr}\{1\}} \\ &\quad + \frac{(\mathrm{Tr}\{\beta H\})^2 - \frac{1}{2}\mathrm{Tr}\{\beta H\}^2}{\mathrm{Tr}\{1\}} + \cdots\bigg)\bigg/\mathrm{Tr}\{1\}\,.\end{aligned} \qquad (2.7)$$

The zeroth moment to second order becomes

$$\langle\omega^0\rangle_{\vec{\mathrm{K}}}^{\alpha\alpha} = \sum_{\vec{\mathrm{g}}-\vec{\mathrm{w}}} e^{i\vec{\mathrm{K}}\cdot(\vec{\mathrm{g}}-\vec{\mathrm{w}})} \left[\beta\langle S_g^{\alpha} S_w^{\alpha}\rangle + \tfrac{1}{2}\beta^2\langle S_g^{\alpha}[S_w^{\alpha},\, H]\rangle\right], \qquad (2.8)$$

where the equal-time specification is now omitted. This procedure also requires the evaluation of traces of products of spin operators with highly variable permutations of the site indices. These traces have been tabulated by Ambler *et al.*[11]

If the components $\alpha\alpha'$ in Eq. (2.2) are interpreted as "+ −" and extended to Eqs. (2.3) and (2.8), it can be shown that the transverse moments are

$$\langle\omega^n\rangle_{\vec{\mathrm{K}}}^{xx} = \langle\omega^n\rangle_{\vec{\mathrm{K}}}^{yy} = \tfrac{1}{2}\langle\omega^n\rangle_{\vec{\mathrm{K}}}^{+-}\,. \qquad (2.9)$$

To calculate the second longitudinal and transverse moments, the following time derivatives are required:

$$i\frac{d}{dt} S_g^{z}(t) = \sum_f I_+(gf)[S_f^+ S_g^- - S_g^+ S_f^-]\,, \qquad (2.10)$$

$$\begin{aligned} i\frac{d}{dt} S_g^{+}(t) &= 2AS_g^z S_g^+ - AS_g^+ - 2\sum_f I_+(fg) S_g^+ S_f^z \\ &\quad + 2\sum_f I_0(fg) S_f^z S_g^+\,. \end{aligned} \qquad (2.11)$$

In the second moments as defined in Eq. (2.3), the thermal averages of spin commutators are considered. Then Eq. (2.7) with Q considered as a commutator becomes

$$\begin{aligned}\langle Q\rangle &= -\beta\frac{\mathrm{Tr}\{HQ\}}{\mathrm{Tr}\{1\}} + \frac{\beta^2\mathrm{Tr}\{H^2 Q\}}{2\,\mathrm{Tr}\{1\}} \\ &\quad - \beta^2\frac{[\mathrm{Tr}\{HQ\}][\mathrm{Tr}\{H\}]}{[\mathrm{Tr}\{1\}]^2}\,.\end{aligned} \qquad (2.12)$$

Thus this is a suitable form for the first-order temperature correction to the second- and higher-order frequency moments. Note that

$$\mathrm{Tr}\{1\} = 2S+1\,. \qquad (2.13)$$

If a spin parameter a is defined as

$$a = \tfrac{1}{3}S(S+1)\,, \qquad (2.14)$$

it can be seen that the $\mathrm{Tr}(H)$ is finite for the axial anisotropic Hamiltonian of Eq. (2.1) and that it equals $-aAN$, where N equals the number of spin sites.

III. FIRST-ORDER TEMPERATURE CORRECTIONS TO FREQUENCY MOMENTS

With the thermal expansion of Eq. (2.8), the zeroth longitudinal moment becomes

$$\frac{\langle\omega^0\rangle_{\vec{\mathrm{K}}}^{zz}}{\beta a} = 1 + \beta a\left[\tfrac{4}{5}A\left(1 - \frac{1}{4a}\right) + 2\sum_{\vec{\mathrm{R}}} e^{i\vec{\mathrm{K}}\cdot\vec{\mathrm{R}}} I_0(\vec{\mathrm{R}})\right], \qquad (3.1)$$

where the spin separation vector $\vec{\mathrm{g}} - \vec{\mathrm{w}}$ has been replaced by $\vec{\mathrm{R}}$. If $\vec{\mathrm{U}}$ is similarly defined as the lattice vector between site g and all sites other than w, which are included within the range of the exchange interactions, then the second longitudinal moment becomes, by Eq. (2.3),

$$\frac{\langle\omega^2\rangle_{\vec{\mathrm{K}}}^{zz}}{16\beta a^2} = \frac{1}{2}\sum_{\vec{\mathrm{R}}} I_+^2(\vec{\mathrm{R}})(1 - e^{i\vec{\mathrm{K}}\cdot\vec{\mathrm{R}}})$$

$$+\beta a\left[-\tfrac{2}{5}A\left(1-\frac{1}{4a}\right)\sum_{\vec{R}} I_+^2(\vec{R})(1-e^{i\vec{K}\cdot\vec{R}})\right]$$

$$+\beta a\sum_{\vec{R}\vec{U}} I_+(\vec{R})I_+(\vec{U})I_+(\vec{R}-\vec{U})(1-e^{i\vec{K}\cdot\vec{R}})$$

$$+\frac{\beta}{4}\sum_{\vec{R}} I_+^2(\vec{R})I_0(\vec{R})(e^{i\vec{K}\cdot\vec{R}}-1). \quad (3.2)$$

With the same expansions, the zeroth transverse moment is

$$\frac{\langle\omega^0\rangle_{\vec{K}}^{xx}}{\beta a}=1+\beta a\left[-\tfrac{2}{5}A\left(1-\frac{1}{4a}\right)+2\sum_{\vec{R}} e^{i\vec{K}\cdot\vec{R}}I_+(\vec{R})\right], \quad (3.3)$$

and the second transverse moment becomes

$$\frac{\langle\omega^2\rangle_{\vec{K}}^{xx}}{16\beta a^2}=\tfrac{3}{20}A^2\left(1-\frac{1}{4a}\right)+\beta aA^3\left(\frac{3}{70}-\frac{9}{140a}+\frac{3}{224a^2}\right)+\frac{\beta aA}{5}\left(1-\frac{1}{4a}\right)\sum_{\vec{R}}\{-e^{i\vec{K}\cdot\vec{R}}I_+(\vec{R})I_0(\vec{R})+\tfrac{7}{2}I_0^2(\vec{R})-\tfrac{5}{2}I_+^2(\vec{R})\}$$
$$+\frac{1}{4}\sum_{\vec{R}}[I_+^2(\vec{R})+I_0^2(\vec{R})-2e^{i\vec{K}\cdot\vec{R}}I_+(\vec{R})I_0(\vec{R})]+\frac{\beta a}{2}\sum_{\vec{R}\vec{U}}\{-e^{i\vec{K}\cdot\vec{R}}[I_+(\vec{R})I_0(\vec{U})I_0(\vec{R}-\vec{U})+I_0(\vec{R})I_+(\vec{U})I_+(\vec{R}-\vec{U})]$$
$$+I_0(\vec{R})I_0(\vec{U})I_0(\vec{R}-\vec{U})+I_+(\vec{R})I_+(\vec{R}-\vec{U})I_+(\vec{U})\}+\frac{\beta}{8}\sum_{\vec{R}}\{e^{i\vec{K}\cdot\vec{R}}[I_+^3(\vec{R})+I_0^2(\vec{R})I_+(\vec{R})]-2I_0(\vec{R})I_+^2(\vec{R})\}. \quad (3.4)$$

It should be mentioned that the necessary conditions attesting to the possible correctness of these results are satisfied, namely, (a) the cancellation of all terms in N, the number of spin sites, and (b) the vanishing of the A terms, when $S=\frac{1}{2}$. In addition, these results for the moments transmute to the results of Ref. 4, in the isotropic limit, i.e., when both $A=0$ and $I_+=I_0$. In this latter case, it is further observed that the longitudinal and transverse moments become identical, as required. The axial-exchange anisotropic dependence of these moments in their temperature expansion is entirely new.

In the notation of Ref. 6, the moments essentially involve a linear combination of the present longitudinal and transverse moments. However, the term in the second-order temperature expansion of the second moments, involving

$$\sum_{\vec{R}} I^3(\vec{R})(e^{i\vec{K}\cdot\vec{R}}-1)$$

in the exchange—isotropic forms of Eqs. (3.2) and (3.4), is missing in Sears's equivalent expression. Since this discrepancy was also noted in Ref. 4, it is probable that Sears's result is in error.

These forms of the frequency moments are general, in the sense that they apply to any lattice structure, for arbitrary range of the exchange integrals, and for any spin magnitude. The frequency moments are now applied to paramagnetic terbium, an hcp structure with a large negative uniaxial anisotropy constant A and with principally nearest neighbor, isotropic exchange, so that $I_+=I_0=I$ in Eqs. (3.1) to (3.4). The moments isotropic in the exchange are therefore

$$\frac{\langle\omega^0\rangle_{\vec{K}}^{zz}}{\beta}=a+\beta a^2\left[\tfrac{4}{5}A\left(1-\frac{1}{4a}\right)+2\sum_{\vec{R}} e^{i\vec{K}\cdot\vec{R}}I(\vec{R})\right], \quad (3.5)$$

$$\frac{\langle\omega^2\rangle_{\vec{K}}^{zz}}{\beta}=8a^2\sum_{\vec{R}}(1-e^{i\vec{K}\cdot\vec{R}})I^2(\vec{R})+\beta\left[-4a^2\sum_{\vec{R}}(1-e^{i\vec{K}\cdot\vec{R}})I^3(\vec{R})+16a^3\sum_{\vec{R}\vec{U}}(1-e^{i\vec{K}\cdot\vec{R}})I(\vec{R})I(\vec{U})I(\vec{R}-\vec{U})\right.$$
$$\left.-\tfrac{32}{5}a^3A\left(1-\frac{1}{4a}\right)\sum_{\vec{R}}(1-e^{i\vec{K}\cdot\vec{R}})I^2(\vec{R})\right], \quad (3.6)$$

$$\frac{\langle\omega^0\rangle_{\vec{K}}^{xx}}{\beta}=a+\beta a^2\left[-\tfrac{2}{5}A\left(1-\frac{1}{4a}\right)+2\sum_{\vec{R}} e^{i\vec{K}\cdot\vec{R}}I(\vec{R})\right], \quad (3.7)$$

$$\frac{\langle\omega^2\rangle_{\vec{K}}^{xx}}{\beta}=8a^2\sum_{\vec{R}}(1-e^{i\vec{K}\cdot\vec{R}})I^2(\vec{R})+\tfrac{12}{5}a^2A^2\left(1-\frac{1}{4a}\right)$$
$$+\beta\left[16a^3\sum_{\vec{R}\vec{U}}(1-e^{i\vec{K}\cdot\vec{R}})I(\vec{R})I(\vec{U})I(\vec{R}-\vec{U})-4a^2\sum_{\vec{R}}(1-e^{i\vec{K}\cdot\vec{R}})I^3(\vec{R})\right.$$
$$\left.+\tfrac{16}{5}a^3A\left(1-\frac{1}{4a}\right)\sum_{\vec{R}}(1-e^{i\vec{K}\cdot\vec{R}})I^2(\vec{R})+16a^3A^3\left(\frac{3}{70}-\frac{9}{140a}+\frac{3}{224a^2}\right)\right]. \quad (3.8)$$

In the hexagonal close-packed structure, there are three distinct kinds of exchange. Let I_b denote the coupling constant for nearest-neighbor spins in the basal plane, I_u the corresponding exchange for the spins in a unit cell, and I_c that for neighboring spins along the c axis. If we consider the fundamental translation vectors $\vec{a}$, $\vec{b}$, and $\vec{c}$ to be along the x axis, 30° counterclockwise from the y axis, and along the z axis, then it can be shown (see Appendix A) that the moments can be written

$$\frac{\langle\omega^0\rangle_{\vec{K}}^{zz}}{\beta}=a+\beta a^2\left[\tfrac{4}{5}A\left(1-\frac{1}{4a}\right)+4(I_bc_1+I_uc_2+I_cc_3)\right], \tag{3.9}$$

$$\begin{aligned}\frac{\langle\omega^2\rangle_{\vec{K}}^{zz}}{\beta}=16a^3[I_b^2(3-c_1)+I_u^2(3-c_2)+I_c^2(1-c_3)]+8\beta a^2\Big\{&(4a-1)I_b^3(3-c_1)-I_u^3(3-c_2)-I_c^3(1-c_3)\\&-\tfrac{8}{5}aA\left(1-\frac{1}{4a}\right)[I_b^2(3-c_1)+I_u^2(3-c_2)+I_c^2(1-c_3)]\Big\},\end{aligned} \tag{3.10}$$

$$\frac{\langle\omega^0\rangle_{\vec{K}}^{xx}}{\beta}=a+\beta a^2\left[-\tfrac{2}{5}A\left(1-\frac{1}{4a}\right)+4(I_bc_1+I_uc_2+I_cc_3)\right], \tag{3.11}$$

$$\begin{aligned}\frac{\langle\omega^2\rangle_{\vec{K}}^{xx}}{\beta}=16a^2\left[I_b^2(3-c_1)+I_u^2(3-c_2)+I_c^2(1-c_3)+\tfrac{3}{20}A^2\left(1-\frac{1}{4a}\right)\right]+8\beta a^2\Big\{&(4a-1)I_b^3(3-c_1)-I_u^3(3-c_2)-I_c^3(1-c_3)\\&+\tfrac{4}{5}aA\left(1-\frac{1}{4a}\right)[I_b^2(3-c_1)+I_u^2(3-c_2)+I_c^2(1-c_3)]+2aA^3\left(\frac{3}{70}-\frac{9}{140a}+\frac{3}{224a^2}\right)\Big\}.\end{aligned} \tag{3.12}$$

Here we have used the notation

$$c_1=\cos(K_xa_0)+2\cos(\tfrac{1}{2}K_xa_0)\cos(\tfrac{1}{2}\sqrt{3}K_ya_0), \tag{3.13}$$

$$c_2=\cos(\tfrac{1}{2}K_zc)\{e^{i\sqrt{3}K_ya_0/6}[2\cos(\tfrac{1}{2}K_xa_0)]+e^{-i\sqrt{3}K_ya_0/6}\}, \tag{3.14}$$

$$c_3=\cos(K_zc), \tag{3.15}$$

$$a_0=|\vec{a}|=|\vec{b}|, \tag{3.16}$$

$$c=|\vec{c}|. \tag{3.17}$$

In the notation of Ref. 6, the zeroth moment $\sigma_0(\vec{K})$ and second moment $\sigma_2(\vec{K})$ are, in terms of the present moments,

$$\sigma_0(\vec{K})=\left[1+\left(\frac{K_z}{K}\right)^2\right]\frac{\langle\omega^0\rangle_{\vec{K}}^{xx}}{\beta}+\left[1-\left(\frac{K_z}{K}\right)^2\right]\frac{\langle\omega^0\rangle_{\vec{K}}^{zz}}{\beta} \tag{3.18}$$

and

$$\sigma_2(\vec{K})=\left[1+\left(\frac{K_z}{K}\right)^2\right]\frac{\langle\omega^2\rangle_{\vec{K}}^{xx}}{\beta}+\left[1-\left(\frac{K_z}{K}\right)^2\right]\frac{\langle\omega^2\rangle_{\vec{K}}^{zz}}{\beta}. \tag{3.19}$$

The rms energy transfer is therefore

$$E_{\mathrm{rms}}=[\sigma_2(\vec{K})/\sigma_0(\vec{K})]^{1/2}. \tag{3.20}$$

The generic form of the frequency moments in Eqs. (3.1) to (3.4) is also particularized to several other lattice structures with exchange interactions extending to next-nearest neighbors. These results are given in Appendix B for a linear chain, a two-dimensional net, and a simple cubic lattice.

IV. APPLICATIONS

The following required data for terbium are given in Ref. 6: $S=6$, $a_0=1.58$ Å. The exchange constants I_b, I_u, I_c are 0.057, 0.050, and 0.038 meV, respectively. The axial anisotropy constant A $=-0.45$ meV.

With this data, the rms energy transfer versus wave vector is calculated in the $\vec{a}$ direction at a temperature of 660 °K and displayed in Fig. 1. Here the solid curve is the present result; the long dashed curve is the Sears calculation; the short dashed curve is the present result at infinite temperature. The crosses denote the data obtained by Cable *et al.* at 660 °K. The present result at infinite temperature agrees with that of Sears, since the theoretical disagreement does not involve the infinite-temperature term. It is seen that the present calculation at 660 °K agrees fairly well with the Sears calculation so that the likely error in Sears's calculation is a relatively minor one in practice. The present result, however, does not give a better fit to the experimental data, but does show a significant presence of short-range-order effects.

Figure 2 displays the same information as in Fig. 1, except that the wave vector is in the $\vec{c}$ direction. Most of the above comments apply here, except that now the present calculations at 660 °K (solid curve) show an improvement over Sears's results in fitting the experimental data at small wave vectors.

The continuing slight disagreement with the experimental results could be due to the neglect of extending the interaction range to include next-nearest neighbors or to the exclusion of higher-order temperature-expansion terms in the moments.

V. CONCLUDING REMARKS

The second-order temperature terms of the zeroth and second, longitudinal and transverse, wave-vector-dependent frequency moments have

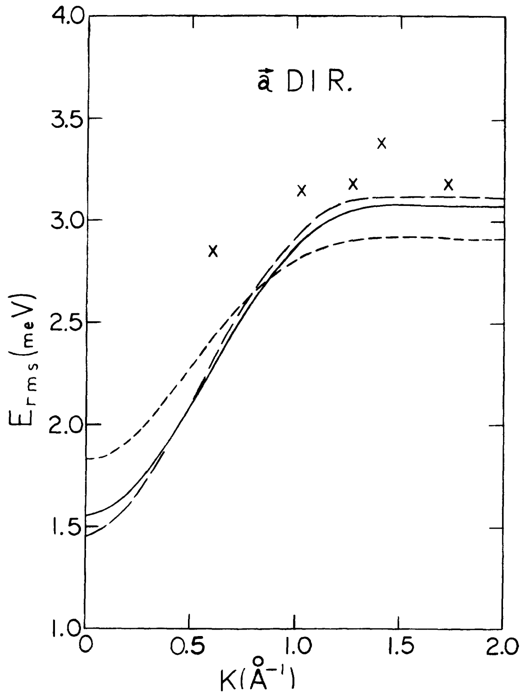

FIG. 1. rms energy transfer of scattered neutrons versus the wave-vector for terbium at 660 °K in the $\vec{a}$ direction of the hexagonal close-packed structure. The solid curve is the present result based on the derived moments, which disagree somewhat in the first-order temperature correction with the previous derivation of Sears. The long dashed curve is the Sears calculation. The short dashed curve is the infinite temperature result common to both the present calculation and the Sears calculation. The crosses denote the neutron scattering data obtained by Cable *et al.*

been derived for a Heisenberg spin system with uniaxial and exchange anisotropy and with an arbitrary interaction range. These generic moments were applied to terbium, an hcp structure with a large negative uniaxial anisotropy. Available data on the rms energy transfer of magnetically scattered neutrons in terbium[10] and previous nearest-neighbor exchange-isotropic moment calculations[6] provided a basis of comparison with the present results. Although an error was found in the previous moment calculations, this error was assessed to be rather small for the case of terbium. In fact, the calculations of the neutron energy transfer by the present moments showed no improvement over those based on the prior moments in matching the experimental data for the wave vector in the $\vec{a}$ direction but did result in a slight improvement for the wave vector in the $\vec{c}$ direction. The short-range-order effects were found to be significant.

The theoretical estimates of these frequency moments are subject to several possible uncertainties, which are difficult to assess. The approximate nature of the system Hamiltonian is one of these, in that the assumption of no coupling between the spin and lattice degrees of freedom neglects the possibility of magnetovibrational scattering. The assumption of nearest-neighbor isotropic exchange and the accuracy of the experimental measures of the input parameters for terbium are also areas of some uncertainty, however small. Finally, the short-range-order effects may be quite significant[4] beyond the limited thermal expansion considered in this paper.

The general form of the moments were also applied to several dimensionalities, including a linear chain, a simple net, and a simple cubic structure. For a hypothetical simple cubic lattice, the variation of the moments with temperature, uniaxial anisotropy, exchange anisotropy, interaction range, and the wave-vector direction and magnitude, were calculated. It is hoped that the temperature dependence, found to be significant, and the anisotropy in the moments can be more completely verified in the future when more pertinent data become available.

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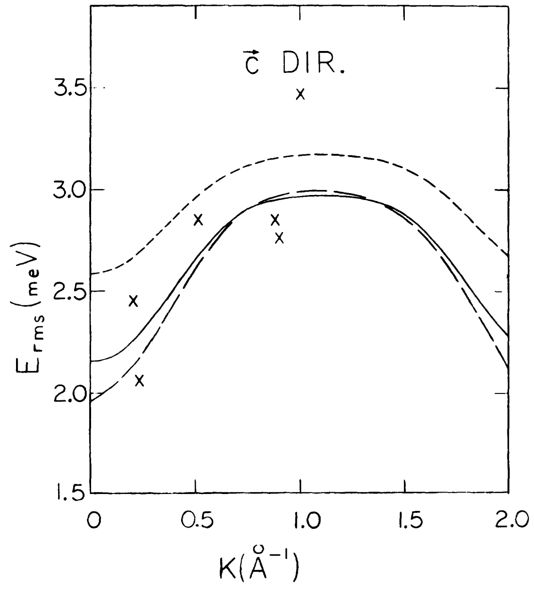

FIG. 2. Same as in Fig. 1 except that the wave-vector is in the $\vec{c}$ direction of the hexagonal close-packed terbium.

APPENDIX A

Nearest-neighbor interactions for the hexagonal close-packed (hcp) structure of terbium are of three distinct varieties, the exchange in the basal plane, in the unit cell, and in the $\vec{c}$ direction. We shall denote these exchanges by I_b, I_u, and I_c. In the geometry of the hcp structure, there are six nearest neighbors in the basal plane, six in the unit cell, and two in the $\vec{c}$ direction. If the notation of Eqs. (3.9) to (3.17) is used, then the spin separation vectors in the basal plane are

$$\vec{v}_1=\vec{a}\ , \qquad \vec{v}_2=\vec{b}\ , \qquad \vec{v}_3=\vec{a}+\vec{b}\ ,$$
$$v_4=-\vec{a}\ , \qquad v_5=-\vec{b}\ , \qquad v_6=-(\vec{a}+\vec{b})\ .$$

The unit cell vectors are

$$\vec{r}_1=\tfrac{2}{3}\vec{a}+\tfrac{1}{3}\vec{b}+\tfrac{1}{2}\vec{c}\ , \qquad \vec{r}_4=\tfrac{2}{3}\vec{a}+\tfrac{1}{3}\vec{b}-\tfrac{1}{2}\vec{c}\ ,$$
$$\vec{r}_2=-\tfrac{1}{3}\vec{a}+\tfrac{1}{3}\vec{b}+\tfrac{1}{2}\vec{c}\ , \qquad \vec{r}_5=-\tfrac{1}{3}\vec{a}+\tfrac{1}{3}\vec{b}-\tfrac{1}{2}\vec{c}\ ,$$
$$\vec{r}_3=-\tfrac{1}{3}\vec{a}-\tfrac{2}{3}\vec{b}+\tfrac{1}{2}\vec{c}\ , \qquad \vec{r}_6=-\tfrac{1}{3}\vec{a}-\tfrac{2}{3}\vec{b}-\tfrac{1}{2}\vec{c}\ .$$

The separation vectors in the $\vec{c}$ direction are

$$\vec{w}_1=\vec{c}\ , \quad \vec{w}_2=-\vec{c}\ .$$

Therefore, an exchange summation term of the type $\sum_{\vec{R}} e^{i\vec{K}\cdot\vec{R}} I^2(\vec{R})$ can be written

$$\sum_{\vec{R}} e^{i\vec{K}\cdot\vec{R}} I^2(\vec{R})=\sum_{i=1}^{6} e^{i\vec{K}\cdot\vec{v}_i} I_b^2+\sum_{i=1}^{6} e^{i\vec{K}\cdot\vec{r}_i} I_u^2 +\sum_{i=1}^{2} e^{i\vec{K}\cdot\vec{w}_i} I_c^2 = 2I_b^2 c_1+2I_u^2 c_2+2I_c^2 c_3\ ,$$

where c_1, c_2, and c_3 are defined in Eqs. (3.13) to (3.17). Here we have defined the crystal axes as

$$\vec{a}=a_0\vec{i}\ , \qquad \vec{b}=(\tfrac{1}{2}\sqrt{3}\,\vec{j}-\tfrac{1}{2}\vec{i})a_0\ , \qquad \vec{c}=c\vec{k}\ ,$$

where $\vec{i}$, $\vec{j}$, and $\vec{k}$ are unit vectors in the x, y, and z directions.

The term $\sum_{\vec{R}} I^2(\vec{R})$ is simply

$$\sum_{\vec{R}} I^2(\vec{R})=6I_b^2+6I_u^2+2I_c^2\ .$$

The term $\sum_{\vec{R}\vec{U}} I(\vec{R})I(\vec{U})I(\vec{R}-\vec{U})$ vanishes everywhere for nearest-neighbor interactions, except in the basal plane. Therefore, we have

$$\sum_{\vec{R}\vec{U}} I(\vec{R})I(\vec{U})I(\vec{R}-\vec{U})=6I_b^3$$

and

$$\sum_{\vec{R}\vec{U}} e^{i\vec{K}\cdot\vec{R}} I(\vec{R})I(\vec{U})I(\vec{R}-\vec{U})=2I_b^3 c_1\ .$$

APPENDIX B

It is convenient to introduce a few new definitions in order to specialize the frequency moments of Eqs. (3.1) to (3.4) to certain lattice structures. With the nearest-neighbor distance denoted as (1) and the next-nearest-neighbor distance as (2), the various energies are normalized by the nearest-neighbor longitudinal exchange $I_0(1)$ as follows:

$$G=I_+(1)/I_0\ , \quad H=I_0(2)/I_0\ , \quad L=I_+(2)/I_0\ , \quad D=A/I_0\ ,$$

where $I_0=I_0(1)$.

In a one-dimensional linear chain, the wave-vector-dependent quantities are introduced as

$$u_1=\cos(K)\ , \quad v_1=\cos(2K)\ ,$$

where K is the reduced wave vector, incorporating the lattice constant. By simple geometrical considerations, the longitudinal and transverse frequency moments of a linear chain then become

$$\frac{\langle\omega^0\rangle_{\vec{K}}^{zz}}{\beta a}=1+\beta I_0 a\left[4(u_1+v_1H)+\tfrac{4}{5}D\left(1-\frac{1}{4a}\right)\right]\ ,$$

$$\frac{\langle\omega^2\rangle_{\vec{K}}^{zz}}{16\beta a^2 I_0^2}=(1-u_1)G^2+(1-v_1)L^2+\beta I_0 a\left\{-\tfrac{4}{5}D\left(1-\frac{1}{4a}\right)[(1-u_1)G^2 +(1-v_1)L^2]+2G^2L(3-2u_1-v_1)\right\}-\tfrac{1}{2}\beta I_0[(1-u_1)G^2+(1-v_1)L^2H]\ ,$$

$$\frac{\langle\omega^0\rangle_{\vec{K}}^{xx}}{\beta a}=1+\beta I_0 a\left[4(u_1G+v_1L)-\tfrac{2}{5}D\left(1-\frac{1}{4a}\right)\right]\ ,$$

$$\frac{\langle\omega^2\rangle_{\vec{K}}^{xx}}{16\beta a^2 I_0^2}=\tfrac{3}{20}D^2\left(1-\frac{1}{4a}\right)-u_1G-v_1LH+\tfrac{1}{2}(1+H^2+G^2+L^2)+\beta I_0 a\{-2u_1G(H+L)-v_1(L+G^2H)+3(H+G^2L)\}$$
$$+\beta I_0\{\tfrac{1}{4}(u_1G^3+v_1L^3)-\tfrac{1}{2}(G^2+HL^2)+\tfrac{1}{4}(u_1G+v_1H^2L)\}$$
$$+\beta I_0 aD\left(1-\frac{1}{4a}\right)[-\tfrac{2}{5}(u_1G+v_1LH)+\tfrac{7}{5}(1+H^2)-G^2-L^2]+\beta I_0 aD^3\left(\frac{3}{70}\right)\left[1-\frac{3}{2a}+\frac{5}{16a^2}\right]\ .$$

In the two-dimensional simple net with the following redefinitions of the wave-vector dependence,

$$u_2=\cos(K_x)+\cos(K_y)\ , \quad v_2=\cos(K_x)\cos(K_y)\ ,$$

the two-dimensional moments become

$$\frac{\langle\omega^0\rangle^{zz}_{\vec{K}}}{\beta a}=1+\beta I_0 a\left[4(u_2+2v_2H)+\tfrac{4}{5}D\left(1-\frac{1}{4a}\right)\right],$$

$$\frac{\langle\omega^2\rangle^{zz}_{\vec{K}}}{16\beta a^2 I_0^2}=(2-u_2)G^2+2(1-v_2)L^2$$

$$-\tfrac{4}{5}\beta I_0 aD\left(1-\frac{1}{4a}\right)[(2-u_2)G^2+2(1-v_2)L^2]$$

$$+8\beta I_0 aG^2L^2(3-u_2-v_2)$$

$$+\tfrac{1}{2}\beta I_0[G^2(u_2-2)+2(v_2-1)L^2H],$$

$$\frac{\langle\omega^0\rangle^{xx}_{\vec{K}}}{\beta a}=1+\beta I_0 a\left[4(u_2G+2v_2L)-\tfrac{2}{5}D\left(1-\frac{1}{4a}\right)\right],$$

$$\frac{\langle\omega^2\rangle^{xx}_{\vec{K}}}{16\beta a^2 I_0^2}=\tfrac{3}{20}D^2\left(1-\frac{1}{4a}\right)-u_2G-2v_2LH+1+H^2$$

$$+G^2+L^2+\beta I_0 a[-4(u_2G[H+L]$$

$$+v_2[L+GH])+12(H^3+G^2L)]+\beta I_0\{\tfrac{1}{4}[u_2G(G^2+1)$$

$$+2v_2L(L^2+1)]-G^2-HL\}$$

$$+\beta I_0 D\left(1-\frac{1}{4a}\right)[-\tfrac{2}{5}(u_2G+2v_2HL)$$

$$+\tfrac{14}{5}(1+H^2)-2(G^2+L^2)^2]$$

$$+\beta I_0 aD^3\left(\frac{3}{70}\right)\left[1-\frac{3}{2a}+\frac{5}{16a^2}\right].$$

For a simple cubic lattice with

$$u_3=\cos(K_x)+\cos(K_y)+\cos(K_z),$$

$$v_3=\cos(K_x)\cos(K_y)+\cos(K_x)\cos(K_z)$$

$$+\cos(K_y)\cos(K_z),$$

the frequency moments are

$$\frac{\langle\omega^0\rangle^{zz}_{\vec{K}}}{\beta a}=1+\beta I_0 a\left[4(u_3+2v_3H)+\tfrac{4}{5}D\left(1-\frac{1}{4a}\right)\right],$$

$$\frac{\langle\omega^2\rangle^{zz}_{\vec{K}}}{16\beta a^2 I_0^2}=G^2(3-u_3)+2L^2(3-v_3)$$

$$-\tfrac{4}{5}\beta I_0 aD\left(1-\frac{1}{4a}\right)[G^2(3-u_3)+2L^2(3-v_3)]$$

$$+8\beta I_0 a[G^2L(9-2u_3-v_3)+2L^3(3-v_3)]$$

$$+\tfrac{1}{2}\beta I_0[G^2(3-u_3)+2(3-v_3)L^2H],$$

$$\frac{\langle\omega^0\rangle^{xx}_{\vec{K}}}{\beta a}=1+\beta I_0 a\left[4(u_3G+2v_3L)-\tfrac{2}{5}D\left(1-\frac{1}{4a}\right)\right],$$

$$\frac{\langle\omega^2\rangle^{xx}_{\vec{K}}}{16\beta a^2 I_0^2}=\tfrac{3}{20}D^2\left(1-\frac{1}{4a}\right)-u_3G-2v_3LH$$

$$+\tfrac{3}{2}(1+2H^2+G^2+2L^2)$$

$$+4\beta I_0 a\{-u_3G(H+2L)$$

$$-v_3L(1+H^2)-v_3HG^2$$

$$-2v_3HL^2+3(3H+2H^2+3G^2L+2L^3)\}$$

$$+\beta I_0\{\tfrac{1}{4}[u_3G(1+G^2)+2v_3L(L^2+H^2)]$$

$$-\tfrac{3}{2}(G^2+2HL^2)\}$$

$$+\beta I_0 aD\left(1-\frac{1}{4a}\right)[-\tfrac{2}{5}(u_3G+2v_3LH)$$

$$+\tfrac{21}{5}(1+2H^2)-3(G^2+2L^2)]$$

$$+\beta I_0 aD^3\left(\frac{3}{70}\right)\left[1-\frac{3}{2a}+\frac{5}{16a^2}\right].$$

For a hypothetical simple cubic lattice, these latter moments were evaluated in their variations with temperature, uniaxial and exchange anisotropy, interaction range and strength, and the wave-vector direction and magnitude. This hypothetical lattice in its isotropic nearest-neighbor limit was assumed to be identical to $RbMnF_3$, a cubic perovskite structure, in its antiferromagnetic state, so that contact could be made with the results of Ref. 4, where, in addition, the wave-vector directions were specified in accordance with the neutron scattering data of Windsor *et al.*[12] Computations of the changes, relative to the infinite temperature case, in the zeroth and second, longitudinal and transverse, frequency moments were conducted as functions of the above-mentioned variables. These calculations (not shown here) indicate the explicit sensitivity of the moments to wide ranges in the above parameters.

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