AD-777 766

FIRST ORDER TEMPERATURE CORRECTIONS TO THE FREQUENCY MOMENTS OF THE SPECTRAL FUNCTION OF ANISOTROPIC HEISENBERG SPIN SYSTEMS.

Frederick A. Malinoski

Frankford Arsenal
Philadelphia, Pennsylvania

June 1973

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UNCLASSIFIED
Security Classification

AD777766

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author)

FRANKFORD ARSENAL
Philadelphia, PA 19137

2a. REPORT SECURITY CLASSIFICATION

Unclassified

2b. GROUP

N/A

3. REPORT TITLE

FIRST-ORDER TEMPERATURE CORRECTIONS TO THE FREQUENCY MOMENTS OF THE SPECTRAL FUNCTION OF ANISOTROPIC HEISENBERG SPIN SYSTEMS

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

Technical Research Report

5. AUTHOR(S) (First name, middle initial, last name)

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*Partially supported by a Long Term Training Grant, Department of the Army.

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
June 1973	40	13

8a. CONTRACT OR GRANT NO.

AMCMS Code: 662604.11.232

b. PROJECT NO.

c. DA Project: 1J562604A010

d.

9a. ORIGINATOR'S REPORT NUMBER(S)

F.A. Report R-2083

9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)

10. DISTRIBUTION STATEMENT

Approved for public release; distribution unlimited.

11. SUPPLEMENTARY NOTES

12. SPONSORING MILITARY ACTIVITY

SASA

13. ABSTRACT

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.

DD FORM 1 NOV 65 1473 REPLACES DD FORM 1473, 1 JAN 64, WHICH IS OBSOLETE FOR ARMY USE.

UNCLASSIFIED
Security Classification

i

14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Exchange Anisotropy						
Uniaxial Anisotropy						
Spin Correlation Function						
Spectral Function						
Frequency Moments						
High Temperature Expansions						
Density Matrix						
Short-Range-Order						
Spin Hamiltonian of Heisenberg Model						
Statistical Thermal Average						
Traces of Spin Operator Products						
Equation of Motion of Spin Operators						
Transverse and Longitudinal Moments						
Fourier Representations						
Lattice Structures						
HCP Lattice						
Linear Chain						
Two-Dimensional Net						
Simple Cubic Lattice						
Neutron Scattering Data						
Terbium						
Next Nearest Neighbor Interactions						
Wave Vectors						

FIRST-ORDER TEMPERATURE CORRECTIONS TO THE FREQUENCY MOMENTS OF THE SPECTRAL FUNCTION OF ANISOTROPIC HEISENBERG SPIN SYSTEMS

by

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AMCMS Code: 662604.11.232
DA Project: 1J562604A010

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June 1973

*Partially supported by a Long Term Training Grant, Department of the Army.

ACKNOWLEDGEMENT

Many thanks are due to Dr. R.A. Tahir-Kheli of the Department of Physics of Temple University and to Dr. D.G. McFadden for their helpful discussions and suggestions. I am also indebted to the Temple University Computer Activity for a generous allotment of computer time on the CDC-6400.

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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 non-vanishing 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 was 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,

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1. R.A. Tahir-Kheli, Phys. Rev. 159, 439 (1967); B1, No. 7, 3163 (1970).
2. R.A. Tahir-Kheli, J. Appl. Phys. 40, 1550 (1967).
3. R.A. Tahir-Kheli and D.G. McFadden, Phys. Rev. 178, 800 (1969); 182, 604 (1969).
4. R.A. Tahir-Kheli and D.G. McFadden, Phys. Rev. B 1, 3178 (1970).
5. R.A. Tahir-Kheli and D.G. McFadden, Phys. Rev. B 1, 3649 (1970).
6. V.F. Sears, Can. J. Phys. 45, 2923 (1967).
7. J. Als-Nielsen, O.W. Dietrich, W. Marshall, and P.A. Lindgård, Solid State Comm. 5, 607 (1967); J. Appl. Phys. 39, 1229 (1968).
8. T.M. Holden, B.M. Powell, and A.D.B. Woods, J. Appl. Phys. 39, 457 (1968).
9. O.W. Dietrich and J. Als-Nielsen, in Neutron Inelastic Scattering, 2 (I.A.E.A., 1968), 63.
10. J.W. Cable, M.F. Collins, and A.D.B. Woods, Proc. of the Sixth Rare Earth Res. Conf., Gatlinburg, Tennessee, p. 297, 1967.

observed by Cable et al.,[10] showed that the uniaxial anisotropy energy yielded a significant contribution to the widths of the energy distributions 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 Reference 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 follows. The section on Formulation covers the basic concept of the frequency moments and a description of the Hamiltonian of the spin system considered. In the section First Order Temperature Corrections to the Frequency Moments the corrections 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 to several lattice structures. In the Applications section the moments are applied to the hexagonal close-packed (hcp) lattice of terbium with nearest neighbor, isotropic exchange and to hypothetical simple cubic structures which reduce to that of $RbMnF_3$ under isotropic conditions. The Concluding Remarks section contains a discussion of the derived results and some final statements. In the Appendix, some details of the calculation of the exchange terms in the hcp structure are presented.

FORMULATION

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

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

4. R.A. Tahir-Kheli and D.G. McFadden, Phys. Rev. B1, 3178 (1970).
6. V.F. Sears, Can. J. Phys. 45, 2923 (1967).
10. J.W. Cable, M.F. Collins, and A.D.B. Woods, Proc. of the Sixth Rare Earth Res. Conf., Gatlinburg, Tennessee, p. 297, 1967.

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

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

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 the 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

$$\left\langle \omega^{n+1} \right\rangle_{\vec{K}}^{\alpha\alpha} = \sum_{\vec{g}-\vec{w}} e^{-i\vec{K}\cdot(\vec{g}-\vec{w})} \left[\left(i \frac{d}{dt} \right)^r \left(-i \frac{d}{dt'} \right)^{n-r} F^{\alpha\alpha}(g-w, t-t') \right]_{t=t'} \tag{2.3}$$

where $\vec{K}$ 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 $\vec{g}-\vec{w}$ includes the origin. It has been shown that the odd moments, i.e., $\langle \omega \rangle_{\vec{K}}^{\alpha\alpha}$, $\langle \omega^3 \rangle_{\vec{K}}^{\alpha\alpha}$, etc., vanish

5. R.A. Tahir-Kheli and D.G. McFadden, Phys. Rev. B 1, 3649 (1970).

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-dependent susceptibility is not determined by Equation (2.3), but involves a special case calculated in Reference 3. The interchangeability of the symmetric spatial variables g and w in the correlation function and the statistical mechanical identity

$$\left\langle B(t)C(t')\right\rangle = \left\langle C(t' - i\beta)\, B(t)\right\rangle \tag{2.4}$$

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

$$\langle\omega^{o}\rangle^{aa}_{\vec{K}} = \sum_{\vec{g}-\vec{w}} e^{i\vec{K}\cdot(\vec{g}-\vec{w})} \int_0^{\beta} d\mu \left\langle S^{a}_{g}(0)\, e^{-\mu H}\, S^{a}_{w}(0)\, e^{\mu H}\right\rangle \tag{2.5}$$

It is noted that the spin operators in Equations (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

$$i\frac{d}{dt} S^{a}_{g}(t) = \left[S^{a}_{g}(t), H\right] \tag{2.6}$$

Next 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

$$\begin{aligned}\langle Q\rangle &= Tr\left\{e^{-\beta H} Q\right\}/Tr\left\{e^{-\beta H}\right\} \\ &= Tr\left\{Q - \beta HQ + \tfrac{1}{2}\beta^2 H^2 Q - \ldots\right\}\Big(1 + Tr\left\{\beta H\right\}/Tr\left\{1\right\} \\ &\quad + \left[(Tr\left\{\beta H\right\})^2 - \tfrac{1}{2} Tr\left\{\beta H\right\}^2\right]/Tr\left\{1\right\} + \ldots\Big)/Tr\left\{1\right\}\end{aligned} \tag{2.7}$$

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1. R.A. Tahir-Kheli, Phys. Rev. 159, 439 (1967); B1, No. 7, 3163 (1970).
2. R.A. Tahir-Kheli, J. Appl. Phys. 40, 1550 (1967).
3. R.A. Tahir-Kheli and D.G. McFadden, Phys. Rev. 178, 800 (1969); 182, 604 (1969).
4. R.A. Tahir-Kheli and D.G. McFadden, Phys. Rev. B 1, 3178 (1970).
5. R.A. Tahir-Kheli and D.G. McFadden, Phys. Rev. B 1, 3649 (1970).

The zeroth moment to second-order becomes

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

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 Equation (2.2) are interpreted as "+-" and extended to Equations (2.3) and (2.8), it can be shown that the transverse moments are

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

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

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

$$i \frac{d}{dt} S_g^+(t) = 2AS_g^z S_g^+ - AS_g^+ - 2\sum_f I_+(fg) S_g^z S_f^z + 2\sum_f I_0(fg) S_f^z S_g^+ \qquad (2.11)$$

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

$$\langle Q \rangle = -\beta \frac{Tr\{HQ\}}{Tr\{1\}} + \tfrac{1}{2}\beta^2 \frac{Tr\{H^2 Q\}}{Tr\{1\}} - \beta^2 \frac{[Tr\{HQ\}][Tr\{H\}]}{[Tr\{1\}]^2} \qquad (2.12)$$

---

11. E. Ambler, J.C. Eisenstein, and J.F. Schooley, J. Math. Phys. 3, 118 (1962).

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

$$\mathrm{Tr}\left\{1\right\} = 2S + 1 \tag{2.13}$$

where S is the spin magnitude. If a spin parameter, a, is defined as

$$a = \left(\frac{1}{3}\right) S\,(S+1) \tag{2.14}$$

it can be seen that the Tr (H) is finite for the axial anisotropic Hamiltonian of Equation (2.1) and that it equals - aAN, where N = the number of spin sites.

FIRST-ORDER TEMPERATURE CORRECTIONS TO THE FREQUENCY MOMENTS

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

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

where the spin separation vector $\vec{g}$-$\vec{w}$ has been replaced by $\vec{R}$. If $\vec{U}$ 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 Equation (2.3)

$$\begin{aligned}\frac{\left\langle \omega^2 \right\rangle_{\vec{K}}^{zz}}{16\beta a^2} = {} & \tfrac{1}{2} \sum_{\vec{R}} I_+^2(\vec{R})\,(1 - e^{i\vec{K}\cdot\vec{R}}) \\ & + \beta a \left\{-\frac{2}{5} A \left(1 - \frac{1}{4a}\right) \sum_{\vec{R}} I_+^2(\vec{R})\,(1 - e^{i\vec{K}\cdot\vec{R}})\right\} \\ & + \beta a \sum_{\vec{R}\,\vec{U}} I_+(\vec{R})\, I_+(\vec{U})\, I_+(\vec{R} - \vec{U})\,(1 - e^{i\vec{K}\cdot\vec{R}}) \\ & + \frac{\beta}{4} \sum_{\vec{R}} I_+^2(\vec{R})\, I_0\,(\vec{R})\,(e^{i\vec{K}\cdot\vec{R}} - 1)\end{aligned} \tag{3.2}$$

With the same expansions, the zeroth transverse moment is

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

and the second transverse moment becomes

$$\frac{\langle \omega^2 \rangle^{xx}_{\vec{K}}}{16\beta a^2} = \frac{3}{20} A^2 \left(1 - \frac{1}{4a}\right) + \beta a A^3 \left(\frac{3}{70} - \frac{9}{140a} + \frac{3}{224a^2} \right)$$

$$+ \frac{\beta a A}{5} \left(1 - \frac{1}{4a}\right) \sum_{\vec{R}} \left\{ - e^{i\vec{K}\cdot\vec{R}} I_+(\vec{R}) I_0(\vec{R}) + \frac{7}{2} I_0^2(\vec{R}) - \frac{5}{2} I_+^2(\vec{R}) \right\}$$

$$+ \frac{1}{4} \sum_{\vec{R}} \left[I_+^2(\vec{R}) + I_0^2(\vec{R}) - 2e^{i\vec{K}\cdot\vec{R}} I_+(\vec{R}) I_0(\vec{R}) \right]$$

$$+ \frac{\beta a}{2} \sum_{\vec{R}\,\vec{U}} \left\{ - e^{i\vec{K}\cdot\vec{R}} \left[I_+(\vec{R}) I_0(\vec{U}) I_0(\vec{R}-\vec{U}) + I_0(\vec{R}) I_+(\vec{U}) I_+(\vec{R}-\vec{U}) \right] \right.$$

$$\left. + I_0(\vec{R}) I_0(\vec{U}) I_0(\vec{R}-\vec{U}) + I_+(\vec{R}) I_+(\vec{R}-\vec{U}) I_+(\vec{U}) \right\}$$

$$+ \frac{\beta}{8} \sum_{\vec{R}} \left\{ e^{i\vec{K}\cdot\vec{R}} \left[I_+^3(\vec{R}) + I_0^2(\vec{R}) I_+(\vec{R}) \right] - 2 I_0(\vec{R}) I_+^2(\vec{R}) \right\} \tag{3.4}$$

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

In the notation of Reference 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 Equations (3.2) and (3.4) is missing in Sears' equivalent expression. Since this discrepancy was also noted in Reference 4, it is probable that Sears' 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. In order to particularize these results to a certain structure with interactions extending to next nearest neighbors, it is convenient to introduce a few new definitions. 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, $I_0(1) = I_0$, as follows

$$G = \frac{I_+(1)}{I_0} \; ; \; H = \frac{I_0(2)}{I_0} \; ; \; L = \frac{I_+(2)}{I_0} \; ; \; D = \frac{A}{I_0} \qquad (3.5)$$

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

$$u_1 = \cos(K) \; ; \; v_1 = \cos(2K) \qquad (3.6)$$

By simple geometrical considerations, the one-dimensional, longitudinal and transverse frequency moments are then

$$\frac{\langle \omega^0 \rangle^{zz}_{\vec{K}}}{\beta a} = 1 + \beta I_0\, a \left[4(u_1 + v_1 H) + \frac{4}{5} D \left(1 - \frac{1}{4a}\right) \right] \qquad (3.7)$$

$$\frac{\langle\omega^2\rangle^{zz}_{\vec{K}}}{16\beta a^2 I_0^2} = (1-u_1)G^2 + (1-v_1)L^2$$

$$+ \beta I_0 a\left\{-\frac{4}{5}D\left(1-\frac{1}{4a}\right)\left[(1-u_1)G^2 + (1-v_1)L^2\right]\right.$$

$$\left. + 2G^2L(3-2u_1-v_1)\right\}$$

$$- \tfrac{1}{2}\beta I_0\left[(1-u_1)G^2 + (1-v_1)L^2 H\right] \tag{3.8}$$

$$\frac{\langle\omega^0\rangle^{xx}_{\vec{K}}}{\beta a} = 1 + \beta I_0 a\left[4(u_1G + v_1L) - \frac{2}{5}D\left(1-\frac{1}{4a}\right)\right] \tag{3.9}$$

$$\frac{\langle\omega^2\rangle^{xx}_{\vec{K}}}{16\beta a^2 I_0^2} = \frac{3}{20}D^2\left(1-\frac{1}{4a}\right) - u_1G - v_1 LH + \tfrac{1}{2}(1+H^2+G^2+L^2)$$

$$+ \beta I_0 a\left\{-2u_1 G(H+L) - v_1(L+G^2H) + 3(H+G^2L)\right\}$$

$$+ \beta I_0\left\{\tfrac{1}{4}(u_1G^3 + v_1L^3) - \tfrac{1}{2}(G^2+HL^2) + \tfrac{1}{4}(u_1G + v_1H^2L)\right\}$$

$$+ \beta I_0 a D\left(1-\frac{1}{4a}\right)\left[-\frac{2}{5}(u_1G + v_1LH) + \frac{7}{5}(1+H^2) - (G^2+L^2)\right]$$

$$+ \beta I_0 a D^3\left(\frac{3}{70}\right)\left[1 - \frac{3}{2a} + \frac{5}{16a^2}\right] \tag{3.10}$$

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

$$u_2 = \cos(K_x) + \cos(K_y);\; v_2 = \cos(K_x)\cos(K_y) \tag{3.11}$$

the two-dimensional moments become

$$\frac{\langle\omega^0\rangle_{\vec{K}}^{zz}}{\beta a} = 1 + \beta I_0 a\left[4(u_2 + 2v_2 H) + \frac{4}{5}D\left(1 - \frac{1}{4a}\right)\right]$$

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

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

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

$$+ \tfrac{1}{2}\beta I_0\left[G^2(u_2 - 2) + 2(v_2 - 1)L^2 H\right] \qquad \textbf{(3-12)}$$

$$\frac{\langle\omega^0\rangle_{\vec{K}}^{zz}}{\beta a} = 1 + \beta I_0 a\left[4(u_2 G + 2v_2 L) - \frac{2}{5}D\left(1 - \frac{1}{4a}\right)\right] \qquad \textbf{(3-13)}$$

$$\frac{\langle\omega^2\rangle_{\vec{K}}^{xx}}{16\beta a^2 I_0^2} = \frac{3}{20}D^2\left(1 - \frac{1}{4a}\right) - u_2 G - 2v_2 LH + 1 + H^2 + G^2 + L^2$$

$$+ \beta I_0 a\left[-4(u_2 G[H + L] + v_2[L + GH]) + 12(H^3 + G^2 L)\right]$$

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

$$+ \beta I_0 D\left(1 - \frac{1}{4a}\right)\left[-\frac{2}{5}(u_2 G + 2v_2 HL) + \frac{14}{5}(1 + H^2) - 2(G^2 + L^2)^2\right]$$

$$+ \beta I_0 a D^3\left(\frac{3}{70}\right)\left[1 - \frac{3}{2a} + \frac{5}{16a^2}\right] \qquad \textbf{(3-14)}$$

For a simple cubic lattice with

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

$$v_3 = \cos(K_x)\cos(K_y) + \cos(K_x)\cos(K_z) + \cos(K_y)\cos(K_z) \qquad (3.15)$$

the frequency moments are

$$\frac{\langle\omega^0\rangle^{zz}_{\vec{K}}}{\beta a} = 1 + \beta I_0 a\left[4(u_3 + 2v_3 H) + \frac{4}{5}D\left(1 - \frac{1}{4a}\right)\right] \qquad (3.16)$$

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

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

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

$$+ \tfrac{1}{2}\beta I_0\left[G^2(3 - u_3) + 2(3 - v_3)L^2 H\right] \qquad (3.17)$$

$$\frac{\langle\omega^0\rangle^{xx}_{\vec{K}}}{\beta a} = 1 + \beta I_0 a\left[4(u_3 G + 2v_3 L) - \frac{2}{5}D\left(1 - \frac{1}{4a}\right)\right] \qquad (3.18)$$

$$\frac{\langle\omega^2\rangle^{xx}_{\vec{K}}}{16\beta a^2 I_0^2} = \frac{3}{20}D^2\left(1 - \frac{1}{4a}\right) - (u_3 G + 2v_3 LH) + \frac{3}{2}(1 + 2H^2 + G^2 + 2L^2)$$

$$+ 4\beta I_0 a\Big\{- u_3 G(H + 2L) - v_3 L(1 + H^2) - v_3 HG^2$$

$$- 2v_3 HL^2 + 3(3H + 2H^2 + 3G^2 L + 2L^3)\Big\}$$

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

$$+ \beta I_0 a D\left(1 - \frac{1}{4a}\right)\left[-\frac{2}{5}(u_3 G + 2v_3 LH) + \frac{21}{5}(1 + 2H^2) - 3(G^2 + 2L^2)\right]$$

$$+ \beta I_0 a D^3\left(\frac{3}{70}\right)\left[1 - \frac{3}{2a} + \frac{5}{16a^2}\right] \qquad (3.19)$$

The frequency moments are now applied to paramagnetic terbium, an hexagonal close-packed structure with a large negative uniaxial anisotropy constant D and with principally nearest neighbor, isotropic exchange, so that $I_+ = I_0 = I$ in Equations (3.1) to (3.4). The moments isotropic in the exchange are therefore

$$\frac{\langle\omega^0\rangle^{zz}_{\vec{K}}}{\beta} = a + \beta a^2\left[\frac{4}{5}A\left(1 - \frac{1}{4a}\right) + 2\sum_{\vec{R}} e^{i\vec{K}\cdot\vec{R}}\, I(\vec{R})\right] \tag{3.20}$$

$$\begin{aligned}\frac{\langle\omega^2\rangle^{zz}_{\vec{K}}}{\beta} = {} & 8a^2\sum_{\vec{R}}\left(1 - e^{i\vec{K}\cdot\vec{R}}\right) I^2(\vec{R}) + \beta\Bigg\{- 4a^2\sum_{\vec{R}}\left(1 - e^{i\vec{K}\cdot\vec{R}}\right) I^3(\vec{R}) \\ & + 16a^3\sum_{\vec{R}\vec{U}}\left(1 - e^{i\vec{K}\cdot\vec{R}}\right) I(\vec{R})\,I(\vec{U})\,I(\vec{R}-\vec{U}) - \frac{32}{5}a^3 A\left(1 - \frac{1}{4a}\right) \\ & \times\sum_{\vec{R}}\left(1 - e^{i\vec{K}\cdot\vec{R}}\right) I^2(\vec{R})\Bigg\}\end{aligned} \tag{3.21}$$

$$\frac{\langle\omega^0\rangle^{xx}_{\vec{K}}}{\beta} = a + \beta a^2\left[-\frac{2}{5}A\left(1 - \frac{1}{4a}\right) + 2\sum_{\vec{R}} e^{i\vec{K}\cdot\vec{R}}\, I(\vec{R})\right] \tag{3.22}$$

$$\begin{aligned}\frac{\langle\omega^2\rangle^{xx}_{\vec{K}}}{\beta} = {} & 8a^2\sum_{\vec{R}}\left(1 - e^{i\vec{K}\cdot\vec{R}}\right) I^2(\vec{R}) + \frac{12}{5}a^2 A^2\left(1 - \frac{1}{4a}\right) \\ & + \beta\Bigg\{16a^3\sum_{\vec{R}\vec{U}}\left(1 - e^{i\vec{K}\cdot\vec{R}}\right) I(\vec{R})\,I(\vec{U})\,I(\vec{R}-\vec{U}) - 4a^2\sum_{\vec{R}}\left(1 - e^{i\vec{K}\cdot\vec{R}}\right) I^3(\vec{R}) \\ & + \frac{16}{5}a^3 A\left(1 - \frac{1}{4a}\right)\sum_{\vec{R}}\left(1 - e^{i\vec{K}\cdot\vec{R}}\right) I^2(\vec{R}) + 16a^3 A^3\left(\frac{3}{70} - \frac{9}{140a} + \frac{3}{224a^2}\right)\Bigg\}\end{aligned} \tag{3.23}$$

In the hexagonal close-packed structure, there are three distinct kinds of exchanges. 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 $\vec{c}$-axis. If we consider the fundamental translation vectors $\vec{a}$, $\vec{b}$, and $\vec{c}$ to be along the x-axis, 30° counter-clockwise from the y-axis, and along the z-axis, then it can be shown (see the Appendix) that the moments can be written as

$$\frac{\langle\omega^0\rangle^{zz}_{\vec{K}}}{\beta} = a + \beta a^2\left[\frac{4}{5}A\left(1 - \frac{1}{4a}\right) + 4(I_b C_1 + I_u C_2 + I_c C_3)\right] \tag{3.24}$$

$$\begin{aligned}\frac{\langle\omega^2\rangle^{zz}_{\vec{K}}}{\beta} = {} & 16a^3\left[I_b^2(3 - C_1) + I_u^2(3 - C_2) + I_c^2(1 - C_3)\right] \\ & + \beta 8a^2\Big\{(4a - 1)I_b^3(3 - C_1) - I_u^3(3 - C_2) - I_c^3(1 - C_3) \\ & - \frac{8}{5}aA\left(1 - \frac{1}{4a}\right)\left[I_b^2(3 - C_1) + I_u^2(3 - C_2) + I_c^2(1 - C_3)\right]\Big\}\end{aligned} \tag{3.25}$$

$$\frac{\langle\omega^0\rangle^{xx}_{\vec{K}}}{\beta} = a + \beta a^2\left[-\frac{2}{5}A\left(1 - \frac{1}{4a}\right) + 4(I_b C_1 + I_u C_2 + I_c C_3)\right] \tag{3.26}$$

$$\begin{aligned}\frac{\langle\omega^2\rangle^{xx}_{\vec{K}}}{\beta} = {} & 16a^2\left[I_b^2(3 - C_1) + I_u^2(3 - C_2) + I_c^2(1 - C_3) + \frac{3}{20}A^2\left(1 - \frac{1}{4a}\right)\right] \\ & + \beta 8a^2\Big\{(4a - 1)I_b^3(3 - C_1) - I_u^3(3 - C_2) - I_c^3(1 - C_3) \\ & + \frac{4}{5}aA\left(1 - \frac{1}{4a}\right)\left[I_b^2(3 - C_1) + I_u^2(3 - C_2) + I_c^2(1 - C_3)\right] \\ & + 2aA^3\left(\frac{3}{70} - \frac{9}{140a} + \frac{3}{224a^2}\right)\Big\}\end{aligned} \tag{3.27}$$

Here we have used the notation

$$C_1 = \cos(K_x a_0) + 2\cos\left(\frac{K_x a_0}{2}\right)\cos\left(\frac{\sqrt{3}\,K_y a_0}{2}\right) \quad (3.28)$$

$$C_2 = \cos\left(\frac{K_z C}{2}\right)\left\{e^{i\frac{\sqrt{3}}{6}K_y a_0}\left[2\cos\left(\frac{K_x a_0}{2}\right)\right] + e^{-i\frac{\sqrt{3}}{3}K_y a_0}\right\} \quad (3.29)$$

$$C_3 = \cos(K_z C) \quad (3.30)$$

$$a_0 = |\vec{a}| = |\vec{b}| \quad (3.31)$$

$$C = |\vec{c}| \quad (3.32)$$

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

$$\sigma_0(\vec{K}) = \left[1 + \left(\frac{K_z}{K}\right)^2\right]\frac{\langle\omega^0\rangle_{\vec{K}}^{xx}}{\beta} + \left[1 - \left(\frac{K_z}{K}\right)^2\right]\frac{\langle\omega^0\rangle_{\vec{K}}^{zz}}{\beta} \quad (3.33)$$

and

$$\sigma_2(\vec{K}) = \left[1 + \left(\frac{K_z}{K}\right)^2\right]\frac{\langle\omega^2\rangle_{\vec{K}}^{xx}}{\beta} + \left[1 - \left(\frac{K_z}{K}\right)^2\right]\frac{\langle\omega^2\rangle_{\vec{K}}^{zz}}{\beta} \quad (3.34)$$

The r.m.s. energy transfer is therefore

$$E_{rms} = \left[\sigma_2(\vec{K}) / \sigma_0(\vec{K})\right]^{1/2} \quad (3.35)$$

The following required data for terbium are given in Reference 6: $S=6$, $a_0=3.59 \mathring{A}$, $c/a_0=1.58$. 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 r.m.s. energy transfer versus wave-vector is calculated in the $\vec{a}$ direction at a temperature of 660 °K. and displayed in Figure 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' 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.

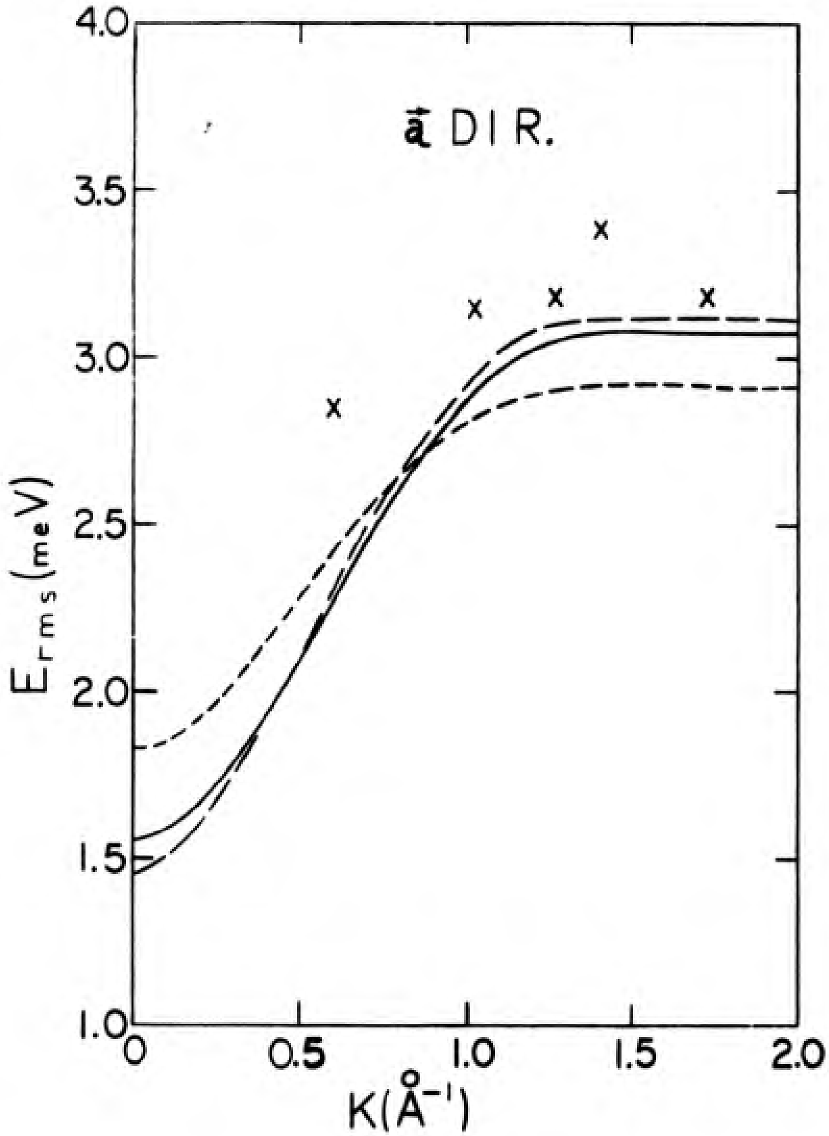
Figure 2 displays the same information as Figure 1, except that the wave-vector is in the $\vec{c}$ direction. Most of the above comments apply here, except that now the present calculations at 660 °K. (solid curve) show an improvement over Sears' results in fitting the experimental data at small wave-vectors.

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.

To evaluate the manner in which these moments can vary with temperature, anisotropy, interaction range, and the wave-vector, we consider a hypothetical simple cubic lattice with variable axial and exchange anisotropy and with variable strength of the next nearest-neighbor exchange. This lattice, in its isotropic nearest-neighbor limit, is assumed to be identical to $RbMnF_3$, a cubic perovskite structure in its antiferromagnetic state so that contact can be made with the results of Reference 4 which, in addition, specified the wave vector directions, in accordance with neutron scattering data of Windsor et al.[12] For this antiferromagnet the lattice constant is 4.24Å, $I_0=-0.28$ meV, and $S=5/2$. The temperature ratio T/T_c was introduced by a relation[13] for the critical (Curie) temperature, T_c

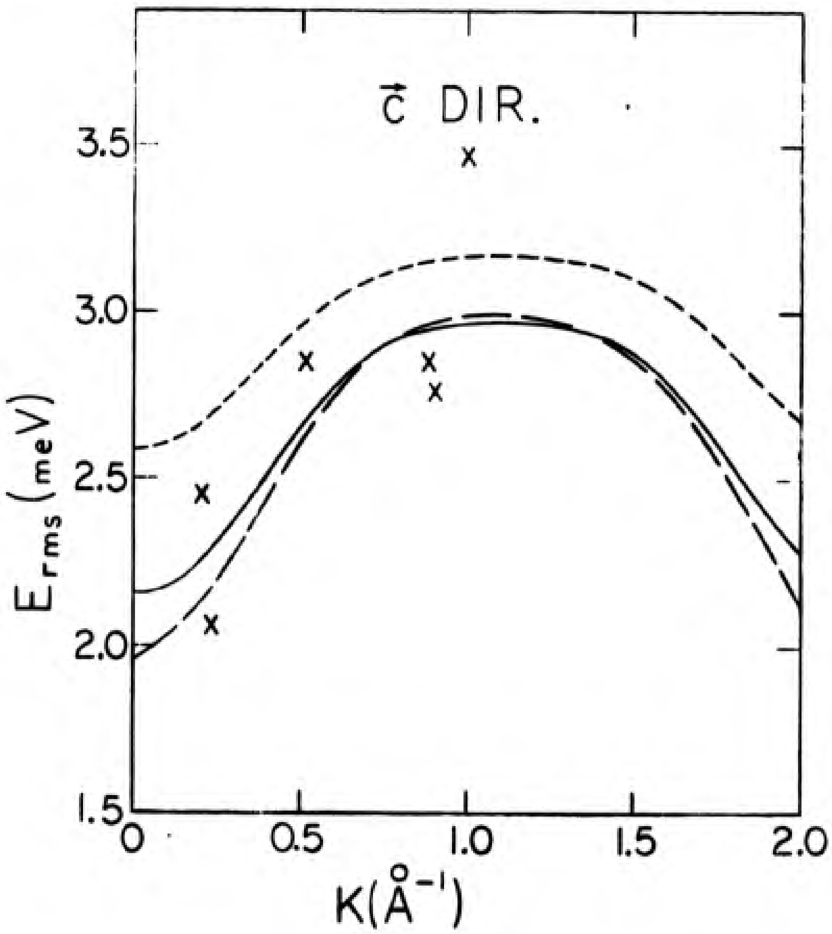
$$k_B T_c = \frac{2}{3F} S(S+1) I_0 z \tag{4.1}$$

12. C.G. Windsor, G.A. Briggs, and M. Kestigan, J. Phys. C (Proc. Phys. Soc.) Ser. 2, 1, 940 (1968).
13. S.V. Tyablikov, Methods in the Quantum Theory of Magnetism, Plenum Press, p. 269, 1967.



Note: 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.

Figure 1. The rms Energy Transfer of Scattered Neutrons vs the Wave-vector for Terbium at 660°K in the $\vec{a}$-Direction of the Hexagonal Close-packed Structure



Note: 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 calaculation. 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.

Figure 2. The Same as in Figure 1 Except That the Wave-vector is in the c-Direction of the Hexagonal Close-packed Terbium

where k_B = Boltzmann constant, z = number of nearest neighbors (=6 for the simple cubic); and F = 1.52 for the simple cubic case. Then in Equations (3.16) – (3.19) for the frequency moments, the dimensionless, temperature factor, βI_0, is given as

$$\beta I_0 \simeq -0.0434/\left(\frac{T}{T_c}\right) \tag{4.2}$$

In Tables I to X we present computations of the percentage changes (relative to the infinite temperature case) in the zeroth and second, longitudinal and transverse, frequency moments as functions of: (1) T/T_c, (2) the magnitude of K_z, (3) the $\vec{K}$ vector plane, (4) the exchange anisctropy G, (5) the axial anisotropy D, and (6) the parameters of the interaction range, H and L. The quantities Δ_0^L, Δ_2^L, Δ_0^T, Δ_2^T are the percentage changes from the infinite temperature results, respectively, of the zeroth and second longitudinal moments, and the zeroth and second transverse moments.

Table I shows the percentage first-order temperature correction for the zeroth longitudinal moment as a function of K_z, the axial anisotropic parameter D, and the value of $\vec{K}$ in the 110 plane, with isotropic nearest-neighbor exchange and a temperature ratio T/T_c=3.89. This value of T/T_c is chosen to to replicate the temperature considered in Reference 4, where $T=3.5T_N$. The wave vector $\vec{K}$ is taken in the 110 plane, such that $K_x=K_y$, $K_{110}=\sqrt{2}\,K_x$, $K_z=|K_z|$expt. (Reference 12). It is seen that: (1) Δ_0^L increases with increasing K_z or K_{110}; (2) Δ_0^L decreases as the axial anisotropic constant D increases; (3) the largest negative Δ_0^L values occur with small K_z and K_{110} and large D; (4) the largest positive Δ_0^L values are obtained with large K_z and K_{110} and small D (i.e., very negative) values. These results are easily verified in Equation (3.16).

Corresponding computations of Δ_0^T are shown in Table II for conditions equivalent to Table I. The results for Δ_0^T are similar to those of Δ_0^L in Table I except that the dependence upon D is, of course, in the opposite sense. See Equation (3.18).

Table III exhibits Δ_2^L as a function of D, for the same conditions of the previous tables. It can be seen from Equation (3.17) that the dependence upon the wave-vector vanishes. For D=0, the Δ_2^L value is simply = $-\frac{1}{2}\beta I_0$ = .56.

TABLE I.

The Percentage First-Order Temperature Corrections, Δ_0^L, to the Zeroth Longitudinal Frequency Moments for Various Values of the z-Component K_z of the Wave Vector, for Values of the Axial Anisotropic Parameter D, and the Value of $\vec{K}$ in the 110 Plane

		Δ_0^L			
K_{110}	D \ K_z	.1	.3	.5	.7
.1	-5	-24.85	-16.81	- 6.18	- .15
	-3	-29.61	-21.58	-10.94	- 4.91
	-1.5	-33.19	-25.15	-14.52	- 8.49
	0	-36.76	-28.72	-18.09	-12.06
	1.5	-40.33	-32.29	-21.66	-15.63
	3	-43.90	-35.87	-25.23	-19.20
	5	-48.67	-40.36	-30.00	-23.97
.3	-5	-16.17	- 8.13	2.50	8.53
	-3	-20.93	-12.89	- 2.26	3.77
	-1.5	-24.50	-16.47	- 5.83	.20
	0	-28.08	-20.04	- 9.41	- 3.38
	1.5	-31.65	-23.61	-12.98	- 6.95
	3	-35.22	-27.18	-16.55	-10.52
	5	-39.98	-31.95	-21.31	-15.28
.5	-5	- 1.83	6.21	16.84	22.87
	-3	- 6.59	1.44	12.08	18.11
	-1.5	-10.17	- 2.13	8.50	14.53
	0	-13.74	- 5.70	4.93	10.96
	1.5	-17.31	- 9.27	1.36	7.39
	3	-20.88	-12.85	- 2.21	3.82
	5	-25.65	-17.61	- 6.98	- .95
.7	-5	13.16	21.20	31.83	37.86
	-3	8.40	16.43	27.07	33.10
	-1.5	4.82	12.86	23.49	29.52
	0	1.25	9.29	19.92	25.95
	1.5	- 2.32	5.72	16.35	22.38
	3	- 5.89	2.14	12.77	18.81
	5	-10.66	- 2.62	8.01	14.04
.9	-5	23.57	31.61	42.24	48.27
	-3	18.81	26.84	37.48	43.51
	-1.5	15.23	23.27	33.90	39.93
	0	11.66	19.70	30.33	36.36
	1.5	8.09	16.13	26.76	32.79
	3	4.52	12.55	23.19	29.21
	5	- .25	7.79	18.42	24.45

NOTE: The system is a simple cubic antiferromagnet with the lattice constant = 4.24A, and the exchange interaction I=-0.28 meV. Exchange isotropy and nearest-neighbor interactions are included, and the temperature ratio T/T_c = 3.89, connecting with the results of Reference 4. G = 1, H = L = 0.

TABLE II.
The Percentage First-Order Temperature Corrections, Δ_0^T, to the Zeroth Transverse Frequency Moments for Various Assumptions

K_{110}	D \ K_z	.1	.3	.5	.7
		Δ_0^T			
.1	-5	-42.71	-34.68	-24.04	-18.01
	-3	-40.33	-32.29	-21.66	-15.63
	-1.5	-38.55	-30.51	-19.87	-13.84
	0	-36.76	-28.72	-18.09	-12.06
	1.5	-34.97	-26.94	-16.30	-10.27
	3	-33.19	-25.15	-14.52	- 8.49
	5	-30.81	-22.77	-12.13	- 6.10
.3	-5	-34.03	-25.99	-15.36	- 9.33
	-3	-31.65	-23.61	-12.98	- 6.95
	-1.5	-29.86	-21.82	-11.19	- 5.16
	0	-28.08	-20.04	- 9.41	- 3.38
	1.5	-26.29	-18.25	- 7.62	- 1.59
	3	-24.50	-16.49	- 5.83	.20
	5	-22.12	-14.08	- 3.45	2.58
.5	-5	-19.69	-11.66	- 1.02	5.01
	-3	-17.31	- 9.27	1.36	7.39
	-1.5	-15.52	- 7.49	3.14	9.18
	0	-13.74	- 5.70	4.93	10.96
	1.5	-11.95	- 3.91	6.72	12.75
	3	-10.17	- 2.13	8.50	14.53
	5	- 7.78	.25	10.89	16.92
.7	-5	- 4.70	3.33	13.97	20.00
	-3	- 2.32	5.71	16.35	22.38
	-1.5	- .54	7.50	18.13	24.16
	0	1.25	9.29	19.92	25.95
	1.5	3.04	11.07	21.71	27.74
	3	4.82	12.86	23.49	29.52
	5	7.20	15.24	25.88	31.90
.9	-5	5.71	13.74	24.38	30.41
	-3	8.09	16.13	26.76	32.79
	-1.5	9.87	17.91	28.55	34.57
	0	11.66	19.70	30.33	36.36
	1.5	13.45	21.48	32.12	38.15
	3	15.23	23.27	33.90	39.93
	5	17.61	25.65	36.29	42.32

NOTE: The same set of assumptions found in Table I apply here.

TABLE III.

The Percentage First-Order Temperature Corrections, Δ_2^L, to the Second Longitudinal Frequency Moments with the Same Conditions as in the Previous Tables

D	Δ_2^L
-5	-11.35
-3	- 6.59
-1.5	- 3.01
0	.56
1.5	4.13
3	7.70
5	12.47

TABLE IV.

The Percentage First-Order Temperature Corrections, Δ_2^T, of the Second Transverse Frequency Moments, for the Same Conditions of the Previous Tables

		Δ_2^T					
K_{110}	K_z \ D	-5	-3	-1.5	1.5	3	5
.1	.1	2.85	1.91	1.36	- .95	-1.77	-2.79
	.3	3.38	2.59	1.91	-1.10	-2.15	-3.17
	.5	3.89	3.03	2.10	-1.16	-2.40	-3.53
	.7	4.11	3.18	2.14	-1.17	-2.48	-3.69
.3	.1	3.42	2.62	1.93	-1.11	-2.17	-3.20
	.3	3.81	2.97	2.07	-1.15	-2.36	-3.48
	.5	4.20	3.24	2.16	-1.18	-2.52	-3.75
	.7	4.37	3.34	2.19	-1.18	-2.57	-3.87
.5	.1	4.05	3.15	2.13	-1.17	-2.46	-3.65
	.3	4.31	3.31	2.18	-1.18	-2.55	-3.83
	.5	4.57	3.45	2.21	-1.19	-2.63	-4.02
	.7	4.69	3.51	2.23	-1.20	-2.67	-4.10
.7	.1	4.49	3.41	2.20	-1.19	-2.61	-3.96
	.3	4.66	3.50	2.23	-1.20	-2.66	-4.08
	.5	4.85	3.59	2.25	-1.20	-2.71	-4.22
	.7	4.94	3.63	2.25	-1.20	-2.73	-4.28
.9	.1	4.71	3.52	2.23	-1.20	-2.67	-4.11
	.3	4.85	3.59	2.25	-1.20	-2.71	-4.21
	.5	5.00	3.65	2.26	-1.20	-2.75	-4.32
	.7	5.08	3.69	2.27	-1.21	-2.77	-4.38

TABLE V.

The Temperature Dependence of Δ_0^L for Various Values of $\vec{K}$ and D. The Wave Vector Direction is the 111 Plane and the Lattice is Isotropic in its Exchange and is Dominated by Only Nearest-neighbor Interactions

K_z	D \ T/T_c	Δ_0^L 2	3	4	5
.1	-5	-46.11	-30.74	-23.05	-18.44
	-3	-55.37	-36.91	-27.69	-22.14
	-1.5	-62.32	-41.55	-31.16	-24.93
	0	-69.27	-46.18	-34.64	-27.71
	1.5	-76.22	-50.81	-38.11	-30.49
	3	-83.17	-55.44	-41.58	-33.27
	5	-92.43	-66.62	-46.22	-36.97
.3	-5	.79	.53	.39	.32
	-3	- 8.48	- 5.65	- 4.24	- 3.39
	-1.5	-15.42	-10.28	- 7.71	- 6.17
	0	-22.37	-14.91	-11.19	- 8.95
	1.5	-29.32	-19.55	-14.66	-11.73
	3	-36.27	-24.18	-18.13	-14.51
	5	-45.53	-30.36	-22.77	-18.21
.5	-5	62.83	41.89	31.42	25.13
	-3	53.57	35.71	26.78	21.43
	-1.5	46.62	31.08	23.31	18.65
	0	39.67	26.45	19.84	15.87
	1.5	32.72	21.82	16.36	13.09
	3	25.78	17.18	12.89	10.31
	5	16.51	11.01	8.26	6.60
.7	-5	98.02	65.35	49.01	39.21
	-3	88.75	59.17	44.38	35.50
	-1.5	81.81	54.54	40.90	32.72
	0	74.86	49.91	37.43	29.94
	1.5	67.91	45.27	33.95	27.16
	3	60.96	40.64	30.48	24.38
	5	51.70	34.46	25.84	20.68

Note: G = 1, H = L = O, K_{111} direction.

TABLE VI.
The Behavior of Δ_0^T with Temperature, as in Table V

K_z	D \ T/T_c	2	3	4	5
		Δ_0^T			
.1	-5	-80.85	-53.90	-40.43	-32.34
	-3	-76.22	-50.81	-38.11	-30.49
	-1.5	-72.74	-48.50	-36.37	-29.10
	0	-69.27	-46.18	-34.64	-27.71
	1.5	-65.80	-43.86	-32.90	-26.32
	3	-62.32	-41.55	-31.16	-24.93
	5	-57.69	-38.46	-28.84	-23.08
.3	-5	-33.96	-22.64	-16.98	-13.58
	-3	-29.32	-19.55	-14.66	-11.73
	-1.5	-25.85	-17.23	-12.92	-10.34
	0	-22.37	-14.91	-11.19	- 8.95
	1.5	-18.90	-12.60	- 9.45	- 7.56
	3	-15.42	-10.28	- 7.71	- 6.17
	5	-10.79	- 7.19	- 5.40	- 4.32
.5	-5	28.09	18.73	14.05	11.24
	-3	32.72	21.82	16.36	13.09
	-1.5	36.20	24.13	18.10	14.48
	0	39.67	26.45	19.84	15.87
	1.5	43.15	28.76	21.57	17.26
	3	46.62	31.08	23.31	18.65
	5	51.25	34.17	25.63	20.50
.7	-5	63.28	42.18	31.64	25.31
	-3	67.91	45.27	33.96	27.16
	-1.5	71.38	47.59	35.69	28.55
	0	74.86	49.91	37.43	29.94
	1.5	78.33	52.22	29.17	31.33
	3	81.81	54.54	40.90	32.72
	5	86.44	57.63	43.22	34.58

TABLE VII.

The Dependence of Δ_2^L Upon Only Temperature and the Axial Anisotropy. The Conditions of Table V Apply

D \ T/T_c	Δ_2^L 2	3	4	5
-5	-22.08	-14.72	-11.04	-8.83
-3	-12.81	- 8.54	- 6.41	-5.12
-1.5	- 5.86	- 3.91	- 2.93	-2.35
0	1.09	.72	.54	.43
1.5	8.03	5.36	4.02	3.21
3	14.98	9.99	7.49	5.99
5	24.25	16.17	12.12	9.70

TABLE VIII.

The Effects of T, D, and $\vec{K}$ Upon Δ_2^T for the Conditions of Table V

K_z	D \ T/T_c	Δ_2^T 2	3	4	5
.1	-5	5.71	3.81	2.85	2.28
	-3	3.98	2.65	1.99	1.59
	-1.5	2.94	1.96	1.47	1.18
	1.5	-1.94	-1.29	- .96	- .78
	3	-3.59	-2.39	-1.80	-1.43
	5	-5.55	-3.70	-2.78	-2.22
.3	-5	8.03	5.35	4.02	3.21
	-3	6.22	4.15	3.11	2.49
	-1.5	4.18	2.78	2.09	1.67
	1.5	-2.28	-1.52	-1.14	- .91
	3	-4.84	-3.23	-2.42	-1.94
	5	-7.20	-4.80	-3.60	-2.88
.5	-5	9.45	6.30	4.73	3.78
	-3	6.98	4.66	3.49	2.79
	-1.5	4.37	2.91	2.18	1.75
	1.5	-2.34	-1.56	-1.17	- .93
	3	-5.28	-3.52	-2.64	-2.11
	5	-8.21	-5.47	-4.11	-3.28
.7	-5	9.93	6.62	4.96	3.97
	-3	7.19	4.79	3.59	2.88
	-1.5	4.41	2.94	2.21	1.76
	1.5	-2.34	-1.56	-1.17	- .94
	3	-5.39	-3.59	-2.69	-2.16
	5	-8.55	-5.70	-4.27	-3.42

TABLE IX.
The Additional Effects of the Exchange Anisotropy G and Next Nearest Neighbor Interactions, H and L, Upon Δ_0^L and Δ_2^L, with the Other Conditions Equivalent to Those in Table V

H	G	K_z	D \ T/T_c	Δ_0^L 2	Δ_0^L 5	Δ_2^L 2	Δ_2^L 5
0	1	.1	-5	- 46.11	-18.44	- 22.08	- 8.83
			0	- 69.27	-27.71	1.09	.43
			5	- 92.43	-36.97	24.25	9.70
		.7	-5	98.02	39.21	- 22.08	- 8.83
			0	74.86	29.94	1.09	.43
			5	51.70	20.68	24.25	9.70
	5	.1	-5	- 46.11	-18.44	- 22.08	- 8.83
			0	- 69.27	-27.71	1.09	.43
			5	- 92.43	-36.97	24.25	9.70
		.7	-5	98.02	39.21	- 22.08	- 8.83
			0	74.86	29.94	1.09	.43
			5	51.70	20.67	24.25	9.70
1	1	.1	-5	-172.38	-68.95	-103.33	-41.33
			0	-195.54	-78.22	- 80.17	-32.07
			5	-218.71	-87.48	- 57.01	-22.80
		.7	-5	- 49.45	-19.78	-122.67	-49.07
			0	- 72.61	-29.04	- 99.51	-39.80
			5	- 95.77	-38.31	- 76.35	-30.54
	5	.1	-5	-172.38	-68.95	-200.69	-80.28
			0	-195.54	-78.22	-177.53	-71.01
			5	-218.71	-87.48	-154.37	-61.75
		.7	-5	- 49.46	-19.78	-124.11	-49.64
			0	- 72.61	-29.04	-100.95	-40.38
			5	- 95.77	-38.31	- 77.79	-31.11

Note: H = L, K_{111} direction.

TABLE X.
The Additional Effects of the Exchange Anisotropy G and Next Nearest Neighbor Interactions, H and L, Upon Δ_0^T and Δ_2^T, with the Other Conditions Equivalent to Those in Table V

H	G	K_z	D \ T/T_c	Δ_0^T 2	Δ_0^T 5	Δ_2^T 2	Δ_2^T 5
0	1	.1	-5	- 80.85	- 32.34	5.71	2.28
			0	- 69.27	- 27.71	1.09	.43
			5	- 57.69	- 23.08	- 5.55	- 2.22
		.7	-5	63.28	25.31	9.93	3.97
			0	74.86	29.94	1.09	.43
			5	86.44	34.58	- 8.55	- 3.42
	5	.1	-5	-357.93	-143.17	- 80.05	-32.02
			0	-346.35	-138.54	- 4.40	- 1.76
			5	-334.77	-133.91	72.29	28.92
		.7	-5	362.71	145.08	- 27.46	-10.99
			0	374.29	149.72	5.39	2.16
			5	385.87	154.35	37.60	15.04
1	1	.1	-5	-207.12	- 82.85	- 43.01	-17.20
			0	-195.54	- 78.22	-183.51	-73.41
			5	-183.96	- 73.58	- 56.83	-22.73
		.7	-5	- 84.19	- 33.67	- 54.43	-21.77
			0	- 72.61	- 29.04	- 99.33	-39.73
			5	- 61.03	- 24.41	- 72.99	-29.20
	5	.1	-5	-484.21	-193.68	-189.81	-75.92
			0	-472.62	-189.05	-131.81	-52.72
			5	-461.04	-184.42	- 43.45	-17.38
		.7	-5	215.24	86.10	-118.25	-47.30
			0	226.82	90.73	- 91.31	-36.52
			5	238.40	95.36	- 53.46	-21.38

Note: H = L, K_{111} direction.

In Table IV, Δ_2^T is shown for the same conditions existing in the previous tables. The behavior of Δ_2^T with K_z, K_{110}, and D is qualitatively similar to that of Δ_0^L in Table I, but the magnitude of Δ_2^T is much smaller, in general, and the variability of Δ_2^T is quite small over the indicated ranges of K_z, K_{110}, and D. Since the values of Δ_2^T, in the complete isotropic limit (i.e., when D=0, in addition to the assumed isotropic exchange), are equivalent to the single longitudinal value of .56 independent of K_z or K_{110} and given in Table III, this quantity is not included in Table IV.

It should be noted that the D=0 values of the moments in Tables I to IV correspond very closely to the results in Reference 4.

The temperature behavior of Δ_0^L is listed in Table V for the isotropic nearest-neighbor exchange. However, the wave-vector direction is the 111 plane, with $K_x=K_y=K_z$. It is observed that Δ_0^L decreases as temperature decreases, as obviously required, and that Δ_0^L increases as K_z becomes larger and decreases as D becomes larger. It is further noted that the variation of Δ_0^L with K_z is considerably greater in the case of the 111 plane than in the 110 plane. At low temperatures Δ_0^L varies quite rapidly with K_z and D.

Table VI shows a corresponding listing of Δ_0^T. Again the results are similar to those for Δ_0^L, except for the reversed dependence upon D. The variation of Δ_0^T with K_z is also greatly enhanced in the 111 plane, in comparison with the 110 plane. The low-temperature large variations of Δ_0^T with K_z and D are again noted.

The corresponding calculations of Δ_2^L are given in Table VII. Since, under the assumed restrictions, Δ_2^L does not depend upon the wave-vector, this quantity is given as a function of only D and T/T_c. It may be observed that the value of Δ_2^L for D=0, T/T_c=4, i.e., .54, is close to the aforementioned value in Table III.

In Table VIII the Δ_2^T calculations for the 111 plane display the expected insensitivity to K_z and D, relative to the other moments.

Table IX explores the additional effects of exchange anisotropy and next nearest-neighbor interactions upon the longitudinal moments. Here we consider the 111 plane and take H=L=0, 1; G=1, 5; K_z=.1, .7; D=-5, 0, +5; and T/T_c=2, 5. It can be seen that: (1) Δ_0^L is not a function of G; (2) Δ_2^L is not a function of G for H=0 but is, for H=1; (3) Δ_0^L and Δ_2^L are very large for H=1.

The similar calculations for Δ_0^T and Δ_2^T are displayed in Table X. It is observed that: (1) Δ_0^T and Δ_2^T are functions of G, even in the nearest-neighbor limit; (2) these transverse temperature corrections are generally larger than the longitudinal corrections for corresponding cases.

CONCLUDING REMARKS

The second-order temperature terms of the zeroth and second, longitudinal and transverse, wave-vector-dependent frequency moments have 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 r.m.s. energy transfer of magnetically scattered neutrons in terbium[10] and previous nearest-neighbor exchange-isotropic moment calculations[6] provided a basis of comparison to 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

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10. J.W. Cable, M.F. Collins, and A.D.B. Woods, Proc. of the Sixth Rare Earth Res. Conf., Gatlinburg, Tennessee, p. 297, 1967.

of magneto-vibrational 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 generic form of the moments were also applied to several dimensionalities, including a variable hypothetical simple cubic structure, based on $RbMnF_3$ in its isotropic nearest-neighbor limit. This consideration was engendered by the dearth of appropriate experimental data for anisotropic, uniaxial structures. Calculations were made for the variation of the moments with temperature, uniaxial anisotropy, exchange anisotropy, interaction range, and the wave-vector direction and magnitude. Hopefully, 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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APPENDIX

EXCHANGE TERMS OF HCP STRUCTURE

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 Equations (3.24) to (3.32) is used, then the spin separation vectors in the basal plane are

$$\vec{v}_1 = \vec{a} \qquad \vec{v}_4 = -\vec{a}$$
$$\vec{v}_2 = \vec{b} \qquad \vec{v}_5 = -\vec{b}$$
$$\vec{v}_3 = \vec{a} + \vec{b} \qquad \vec{v}_6 = -(\vec{a} + \vec{b})$$

The unit cell vectors are

$$\vec{r}_1 = \frac{2}{3}\vec{a} + \frac{1}{3}\vec{b} + \frac{1}{2}\vec{c} \qquad \vec{r}_4 = \frac{2}{3}\vec{a} + \frac{1}{3}\vec{b} - \frac{1}{2}\vec{c}$$
$$\vec{r}_2 = -\frac{1}{3}\vec{a} + \frac{1}{3}\vec{b} + \frac{1}{2}\vec{c} \qquad r_5 = -\frac{1}{3}\vec{a} + \frac{1}{3}\vec{b} - \frac{1}{2}\vec{c}$$
$$\vec{r}_3 = -\frac{1}{3}\vec{a} - \frac{2}{3}\vec{b} + \frac{1}{2}\vec{c} \qquad \vec{r}_6 = -\frac{1}{3}\vec{a} - \frac{2}{3}\vec{b} - \frac{1}{2}\vec{c}$$

The separation vectors in the $\vec{c}$ direction are $\vec{w}_1 = \vec{c}$ and $\vec{w}_2 = -\vec{c}$.

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 as

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

$$= 2I_b^2 C_1 + 2I_u^2 C_2 + 2I_c^2 C_3$$

where C_1, C_2, and C_3 are defined in Equations (3.28) to (3.32). Here we have defined the crystal axes as

$$\vec{a} = a_0 \vec{i}$$

$$\vec{b} = \left(\frac{\sqrt{3}}{2}\vec{j} - \frac{1}{2}\vec{i}\right) a_0$$

$$\vec{c} = c\vec{k}$$

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

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

and

$$\sum_{\vec{R}\vec{U}} e^{i\vec{K}\cdot\vec{R}}\, I(\vec{R})\, I(\vec{U})\, I(\vec{R}-\vec{U}) = 2I_b^3\, C_1$$

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