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High-Temperature Wavelength-Dependent Properties of a Heisenberg Paramagnet*

M. F. Collins
Physics Department, McMaster University, Hamilton, Ontario, Canada

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The first four terms of the high-temperature expansion of the wavelength-dependent susceptibility $\chi(\mathbf{k})$ and the spin correlation function $S(\mathbf{k})$ are calculated. Nearest-neighbor exchange interactions are assumed with general spin values and a Bravais lattice. From these results the first four terms in the high-temperature expansion of the effective range of the spin correlation are obtained for both ferromagnets and antiferromagnets. The terms are slightly different according to whether the range is defined from $\chi(\mathbf{k})$ or from $S(\mathbf{k})$, though they become identical limitingly close to the critical ordering temperature. The calculated properties can all be measured by current neutron scattering techniques.

I. INTRODUCTION

In a recent paper Fisher and Burford[1] have given a comprehensive study of the wavelength-dependent properties of the Ising model. It would be desirable if we could get similar insight into the Heisenberg model with general spin value S, since this corresponds to a case more commonly occurring in nature. The purpose of this paper is to investigate the high-temperature expansion of the wavelength-dependent properties of this Heisenberg model.

An immediate extra complication arises for the Heisenberg Hamiltonian over the Ising case because there are two distinct wavelength-dependent properties of interest. These are $\chi(\mathbf{k})$, the susceptibility, and $S(\mathbf{k})$, the spatial Fourier transform of the two-spin correlation function, where $\mathbf{k}$ is a general vector in reciprocal space. For the Ising model these two quantities are the same, to within a constant factor; the differences arise essentially because the Heisenberg Hamiltonian does not commute with S^z on any particular atom while the Ising Hamiltonian does. For the Heisenberg Hamiltonian the two quantities only become the same at $\mathbf{k}=0$ or in the limit as S tends to infinity (the "classical model").

This paper deals with the expansion of both $\chi(\mathbf{k})$ and $S(\mathbf{k})$ at high temperatures. For the special cases of $\chi(\mathbf{0})$ and $\chi(\boldsymbol{\tau})$, where $\boldsymbol{\tau}$ is an antiferromagnetic reciprocal-lattice vector of a loose-packed material, Rushbrooke and Wood[2,3] have given the expansions to six terms. This involved a laborious calculation and, seeing that the analogous calculations at general wave vectors are even more laborious, this work only goes as far as the fourth term for a general Bravais lattice of spins.

Given the functions $\chi(\mathbf{k})$ and $S(\mathbf{k})$, an expansion can be made about the point $\mathbf{k}=0$ to give

$$\chi(\mathbf{k})=\chi(0)[1-\xi_x^2k^2+O(k^4)] \tag{1}$$

and

$$S(\mathbf{k})=S(0)[1-\xi_s^2k^2+O(k^4)]. \tag{2}$$

The quantity ξ as defined by the above equations is known as the effective range of spin correlation.[1] For the Heisenberg Hamiltonian there are two such effective ranges ξ_s and ξ_x defined from the functions $S(\mathbf{k})$ and $\chi(\mathbf{k})$, respectively. It can be shown that the effective ranges become equal in the limit as the temperature approaches the critical ordering temperature from above so that the parameters have the same critical exponent ν_1.

The quantities $S(\mathbf{k})$, $\chi(\mathbf{k})$, ξ_x, and ξ_s which are calculated in this paper are all observable quantitatively by current neutron scattering techniques. The physical principles involved in such experiments have been reviewed recently by Marshall and Lowde[4] and will not be discussed in the present paper. In fact, hardly any measurements of spin correlations have been made in the range of temperature from about twice the critical temperature upwards. Only at these temperatures do the terms in the high-temperature expansion that have been calculated give sufficient convergence for quantitative comparison with experiments. It is hoped that the expansions given in this paper will stimulate further experimental work.

II. FORMAL THEORY

Firstly formal expressions will be given for $S(\mathbf{k})$ and $\chi(\mathbf{k})$ and for their high-temperature expansions.

The Fourier transform of the two-spin correlation function is given by

$$S^\alpha(\mathbf{k})=N^{-1}\sum_{\mathbf{n},\mathbf{m}} e^{i\mathbf{k}\cdot(\mathbf{n}-\mathbf{m})}\langle S_\mathbf{n}^\alpha S_\mathbf{m}^\alpha\rangle_T, \qquad (3)$$

where the superscript $\alpha=x$, y, or z, N is the number of atoms in the crystal, $\mathbf{n}$ and $\mathbf{m}$ are atomic positions, and $S_\mathbf{n}^\alpha$ is the α component of spin of the atom at $\mathbf{n}$. The expectation value is to be evaluated at the temperature T of the crystal. Where no ambiguity arises the superscript α in the function $S^\alpha(\mathbf{k})$ will be dropped.

Following the method of Opechowski[5] and of Rushbrooke and Wood,[2] the high-temperature expansion for $S^\alpha(\mathbf{k})$ in powers of β $[=(k_BT)^{-1}]$ is given by

$$S^\alpha(\mathbf{k})=N^{-1}\sum_{\mathbf{n},\mathbf{m}} e^{i\mathbf{k}\cdot(\mathbf{n}-\mathbf{m})}\sum_{r=0}^{\infty}\frac{(-\beta)^r}{r!}\langle S_\mathbf{n}^\alpha S_\mathbf{m}^\alpha \mathcal{H}_0^r\rangle_\infty, \qquad (4)$$

where $\mathcal{H}_0$ is the Hamiltonian of the spin system and the expectation value is to be evaluated at temperature $T=\infty$.

The wavelength-dependent susceptibility $\chi(\mathbf{k})$ is formally defined by the equation

$$M^\alpha(\mathbf{k})=\chi^\alpha(\mathbf{k})H^\alpha(\mathbf{k}), \qquad (5)$$

where $M^\alpha(\mathbf{k})$ is the $\mathbf{k}$ Fourier transform of magnetization in the α direction produced by the $\mathbf{k}$ Fourier transform of the applied field $H^\alpha(\mathbf{k})$ in the α direction.

The Hamiltonian of the system $\mathcal{H}$ is given by

$$\mathcal{H}=\mathcal{H}_0-N^{-1}\sum_{\mathbf{k},\alpha} M^\alpha(\mathbf{k})H^\alpha(-\mathbf{k}), \qquad (6)$$

where $\mathcal{H}_0$ is the Hamiltonian in zero applied field.

High-temperature expansion, following the methods of Opechowski[5] and of Rushbrooke and Wood,[2,3] gives

$$\chi^\alpha(\mathbf{k})=g^2\mu^2\beta\sum_{\mathbf{n},\mathbf{m}} e^{i\mathbf{k}\cdot(\mathbf{n}-\mathbf{m})}\sum_{r=0}^{\infty}\frac{(-\beta)^r}{(r+1)!}\langle S_\mathbf{n}^\alpha\{\mathcal{H}_0^r S_\mathbf{m}^\alpha\}\rangle_\infty, \qquad (7)$$

where the curly brackets symbol $\{\cdots\}$ denotes a sum over the terms inside the bracket taken in all distinct sequences; μ is the Bohr magneton and g the gyromagnetic ratio.

If $\mathcal{H}_0$ commutes with $S_\mathbf{m}^\alpha$ the expressions (4) and (7) for $S^\alpha(\mathbf{k})$ and $\chi^\alpha(\mathbf{k})$ are identical except for the constant factor $Ng^2\mu^2\beta$. This is the case for the Ising Hamiltonian with $\alpha=z$ and also for the Heisenberg Hamiltonian if $\mathbf{k}=0$. The latter condition arises because the Heisenberg Hamiltonian does commute with $\sum_\mathbf{m} S_\mathbf{m}^\alpha$. It is shown in Appendix A that for the Heisenberg antiferromagnet $\chi^\alpha(\boldsymbol{\tau})$ and $Ng^2\mu^2\beta S^\alpha(\boldsymbol{\tau})$ become equal in the limit $T\rightarrow T_N^+$, where $\boldsymbol{\tau}$ is an antiferromagnetic reciprocal-lattice vector.

Rushbrooke and Wood[2,3] have evaluated the expansion of $S^\alpha(\mathbf{k})$ [Eq. (4)] up to $r=6$ for the special case $\mathbf{k}=0$, and also the expansion of $\chi^\alpha(\mathbf{k})$ [Eq. (7)] to the same order for the special cases of simple-cubic and body-centered-cubic lattices with just antiferromagnetic nearest-neighbor interactions at $\mathbf{k}=\boldsymbol{\tau}$, the antiferromagnetic reciprocal-lattice vector. In this paper, $S^\alpha(\mathbf{k})$ and $\chi^\alpha(\mathbf{k})$ are evaluated for general $\mathbf{k}$ values and any Bravais lattice with just nearest-neighbor interactions up to $r=4$. The method used is essentially the same as that of Rushbrooke and Wood, though in the calculation of the susceptibility expansion it is necessary to calculate also terms arising from so-called "even graphs" and also to calculate terms involving pairs of nearest-neighbor atoms to a given atom which are also nearest neighbors to each other ($p_1\neq 0$ in Rushbrooke and Wood's notation).

Except for these two types of terms, the agreement which is found between the present calculations and those of Rushbrooke and Wood should constitute a sufficient checking procedure for the results. A necessary, but not quite sufficient, check on the other terms has been provided by using the calculated expectation values also to determine the fourth moment of the neutron scattering cross section. This is an evaluation of the quantity

$$\sum_{\mathbf{n},\mathbf{m}} e^{i\mathbf{k}\cdot(\mathbf{n}-\mathbf{m})}\langle S_\mathbf{n}^\alpha \mathcal{L}^4 S_\mathbf{m}^\alpha\rangle_\infty,$$

where $\mathcal{L}$ is the Liouville operator representing a commutation with the Hamiltonian. The expression obtained agrees with that given by Collins and Marshall[6] using a different approach.

The actual terms in the expansion rapidly become lengthy as r increases; they are written out in Appendix B.

Two other calculations in this area have appeared in the literature. Dwight, Menyuk, and Kaplan[7] calculated the second term of $S(\mathbf{k})$ for the relatively complicated spin system found in ferrites. Since our calculation was completed, Tahir-Kheli and McFadden[8] published an expansion of $\chi(\mathbf{k})$ up to $r=3$. Their expressions for $r=2$ and for $r=3$ do not agree completely with those given in the present paper. Their work also does not agree with the susceptibility expansion of Rushbrooke and Wood[2,3] and of earlier workers; it is believed that there is an error in the calculation of Tahir-Kheli and McFadden for a certain set of diagrams. The correct

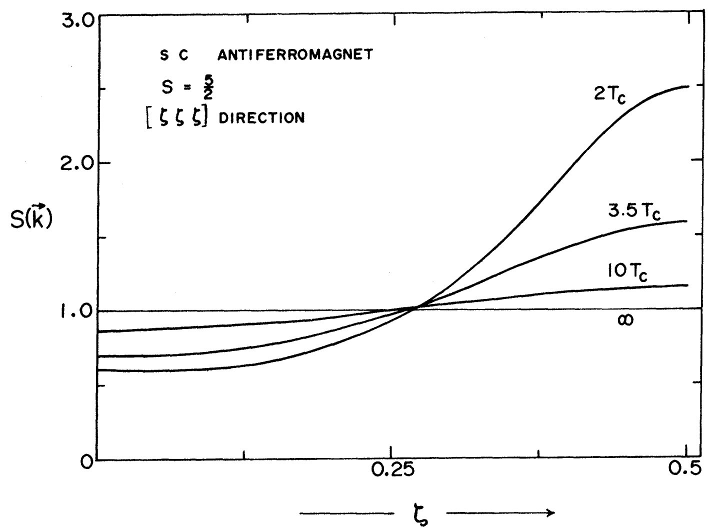

FIG. 1. Neutron scattering $S(\mathbf{k})$ at high temperatures for a simple-cubic antiferromagnet with spin $\frac{5}{2}$ plotted from the origin (0, 0, 0) to the antiferromagnetic reciprocal-lattice vector $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. The scale is such that for a noninteracting spin system $S(\mathbf{k})=1$. As the temperature approaches the ordering temperature T_N the scattering becomes peaked about the point $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$.

treatment of these diagrams follows from Theorem II of Ref. 2.

III. EFFECTIVE RANGE OF CORRELATION

The expression for $\chi(\mathbf{k})$ given in Appendix B can be expanded at small wave vector $\mathbf{k}$ to give

$$\chi(\mathbf{k})=\chi(0)-Ak^2+O(k^4), \tag{8}$$

where for cubic symmetry the parameter A is independent of the direction of $\mathbf{k}$. Comparison with Eq. (1) shows that

$$\xi_x^2=A/\chi(0).$$

The power series in β for $\chi(0)$ can be inverted to give a power series for $\chi(0)^{-1}$. Multiplication of this series with that for A and collection of terms of the same order in β gives a power series for ξ_x^2 in powers of β. The leading term is of order β, so that ξ_x tends to zero as β tends to zero, as is to be expected on physical grounds.

Analogous expansions can be made for ξ_s^2 and for both ξ_x^2 and ξ_s^2 in the antiferromagnetic case. The numerical values of the terms in these expansions are given in Appendix C.

The series for ξ_x^2 and for ξ_s^2 are not the same; the differences between them become smaller as S increases, as expected. Arguments on the same lines as those given in Appendix A can be constructed for the limiting behavior as the critical ordering temperature is approached from above. The necessary generalization of the arguments of Appendix A is that the quantity $\langle\omega^2\rangle$ must be expressed as a function of $\mathbf{k}$ as well as of ξ. This is done using dynamic scaling for the hydro-

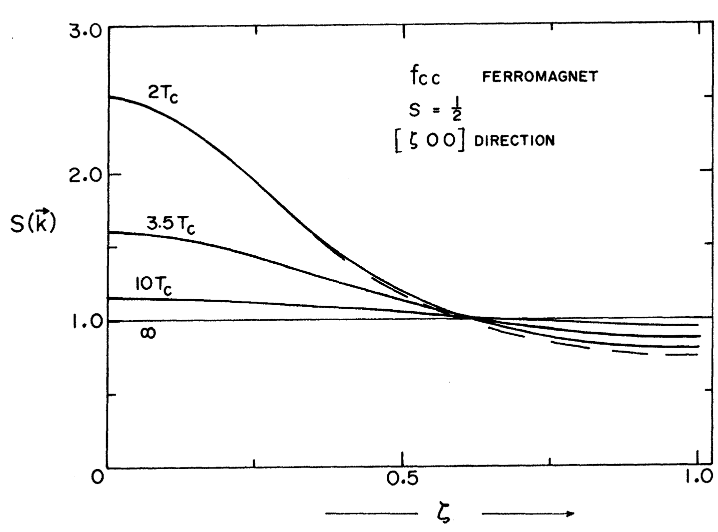

FIG. 2. Neutron scattering $S(\mathbf{k})$ at high temperatures from a face-centered cubic ferromagnet with spin $\frac{1}{2}$. The scattering is plotted from the origin to the [100] zone boundary. As the temperature approaches the ordering temperature T_c, the scattering becomes peaked about the origin. The dashed line shows the analogous plot for $\chi(\mathbf{k})$ at temperature $2T_c$; it should be within the scope of current neutron scattering techniques to observe the differences between $\chi(\mathbf{k})$ and $S(\mathbf{k})$.

dynamic region with the result that

$$\lim_{T\to T_c+}(\xi_s^2-\xi_x^2)=0.$$

Thus the two series become asymptotically the same, with the same critical index ν_1. This is not apparent in the limited number of terms given in Appendix C, especially for small values of the spin S.

Though all the terms in the series are positive, it does not appear to be quite as regular as the series for χ given by Rushbrooke and Wood.[2,3] Use of the ratio technique indicates divergence at the same temperature as the corresponding series for χ to within the uncertainties of the extrapolations (about 10%). The series are too short to enable any good estimate of the critical exponent ν_1 to be made; our best estimate is 0.68 ± 0.07. Fisher and Burford[9] have obtained the first six terms in the series for the ferromagnet, though their actual results do not appear to be quoted in the literature. They claim that for the face-centered-cubic lattice with $S=\infty$ the critical exponent is 0.692 ± 0.012.

IV. NUMERICAL VALUES

In this section some numerical values of $\chi(\mathbf{k})$ and $S(\mathbf{k})$ are given. The series are too short to be of value in determining properties at temperature close to T_c, but for temperatures greater than about $2T_c$ the convergence is satisfactory. Thus the series investigates the short-range order of a spin system at temperatures greater than $2T_c$. In Fig. 1 the function $S(\mathbf{k})$ is plotted for a simple-cubic antiferromagnet with spin $\frac{5}{2}$ at four temperatures. The data are plotted with $\mathbf{k}$ in the [111] direction, with $\mathbf{k}$ varying from the origin (0, 0, 0) to the antiferromagnetic reciprocal-lattice vector $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$. The scale is such that for a noninteracting paramagnetic spin $S(\mathbf{k})=1$. Around the origin $S(\mathbf{k})$ is less than 1, but around the point $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ $S(\mathbf{k})$ is greater than 1, as would be expected qualitatively. The short-range order persists at temperatures well above T_c.

It is clear that the curvature of the plots at the point $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ increases as the temperature decreases. This shows the increasing range of correlation as T_N is approached.

There appear to be no careful quantitative measurements of $S(\mathbf{k})$ except in the critical region. Windsor *et al.*[10] have reported some qualitative data at $3.5T_c$ for rubidium manganese fluoride arising from an investigation of the frequency spectrum of $S(\mathbf{k})$. Their results have the same qualitative features as Fig. 1, but the degree of short-range order observed is only about two-thirds of what is predicted in this paper.

It should perhaps also be noted that the calculations here show that corrections for short-range order must be made to paramagnetic form-factor data taken by neutron scattering techniques. For example, Erickson[11] typically took data at $8T_c$, a temperature where corrections of up to 20% must be applied.

Figure 2 shows the analogous plot to Fig. 1 for a ferromagnet. This time the [100] direction of face-centered-cubic lattice has been chosen with $S=\frac{1}{2}$. The plot is similar to that for the antiferromagnet given in Fig. 1 with no significant alteration of the main features.

At the temperature $2T_c$ the dashed line indicates the analogous plot for $\chi(\mathbf{k})$. The differences between the plot of $S(\mathbf{k})$ and of $\chi(\mathbf{k})$ are small but it should be within the scope of present-day neutron scattering techniques to observe these differences, at least for $S=\frac{1}{2}$.

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This work has benefitted from a number of discussions with Dr. D. A. Goodings.

APPENDIX A

It can be shown that,[4] above T_c,

$$\chi^\alpha(\mathbf{k})=N\beta g^2\mu^2\int_{-\infty}^{\infty}\frac{1-e^{-\hbar\omega\beta}}{\hbar\omega\beta}S^\alpha(\mathbf{k},\omega)\,d\omega$$

and

$$S^\alpha(\mathbf{k})=\int_{-\infty}^{\infty}S^\alpha(\mathbf{k},\omega)\,d\omega,$$

where

$$S^\alpha(\mathbf{k},\omega)=N^{-1}\sum_{\mathbf{n},\mathbf{m}}e^{i\mathbf{k}\cdot(\mathbf{n}-\mathbf{m})}(2\pi)^{-1}\times\int_{-\infty}^{\infty}e^{-i\omega t}\langle S_{\mathbf{n}}^\alpha(0)S_{\mathbf{m}}^\alpha(t)\rangle_T dt,$$

with $S_{\mathbf{n}}^\alpha(t)$ the α component of spin on the atom at $\mathbf{n}$ at time t.

At $T\leq T_N$ and $\mathbf{k}=\boldsymbol{\tau}$ for Heisenberg antiferromagnet the function $S^\alpha(\mathbf{k},\omega)$ tends to a δ function in ω. Just above T_c it would appear to be satisfactory to expand the function $e^{-\hbar\omega\beta}$ as a power series in $\hbar\omega\beta$, and we assume this to be the case. Then we have

$$\chi^\alpha(\boldsymbol{\tau})=N\beta g^2\mu^2\int_{-\infty}^{\infty}\{1-\tfrac{1}{2}\hbar\omega\beta+\tfrac{1}{6}[(\hbar\omega\beta)^2]\}S^\alpha(\boldsymbol{\tau},\omega)\,d\omega.$$

For a centrosymmetric lattice the principle of detailed balance shows that[4]

$$S^\alpha(\boldsymbol{\tau},-\omega)=e^{-\beta\hbar\omega}S^\alpha(\boldsymbol{\tau},\omega).$$

Expanding the exponent also, we obtain

$$\chi^\alpha(\boldsymbol{\tau})=N\beta g^2\mu^2\int_{-\infty}^{\infty}[1-(\tfrac{1}{12}\hbar^2\beta^2\omega^2)+O(\omega^4)]S^\alpha(\boldsymbol{\tau},\omega)\,d\omega$$
$$=N\beta g^2\mu^2S^\alpha(\boldsymbol{\tau})(1-\tfrac{1}{12}\hbar^2\beta^2\langle\omega^2\rangle+O\langle\omega^4\rangle),$$

where

$$\langle\omega^n\rangle=\int_{-\infty}^{\infty}\omega^nS^\alpha(\boldsymbol{\tau},\omega)\,d\omega/S^\alpha(\boldsymbol{\tau}).$$

TABLE I. First four expansion coefficients of ξ^2 for the simple-cubic lattice and spin values S of $\frac{1}{2}$, 1, $\frac{3}{2}$, 2, $\frac{5}{2}$, and 3. The coefficients are expressed in units of the lattice constant a.

Spin S	v	v'	w	w'
$\frac{1}{2}$	0.5	0.5	0.5	0.5
	1.41667	1.58333	1.25	1.75
	2.91667	3.91667	3.5	2.5
	5.49167	8.625	4.91667	7.33333
1	1.333	1.333	1.333	1.3333
	10.4444	10.8889	10.0	11.3333
	65.5556	72.6667	69.3333	62.2222
	390.0148	457.0370	381.5062	452.2716
$\frac{3}{2}$	2.5	2.5	2.5	2.5
	37.0833	37.9167	36.25	38.75
	451.9167	476.9167	464.8333	439.8333
	5 267.9583	5 724.9583	5 215.4167	5 730.8333
2	4.0	4.0	4.0	4.0
	95.333	96.667	94.0	98.0
	1 886.267	1 950.267	1 918.933	1 854.933
	35 834.933	37 734.133	35 625.467	37 824.133
$\frac{5}{2}$	5.833	5.833	5.833	5.833
	203.194	205.139	201.25	207.083
	5 907.806	6 043.917	5 976.833	5 840.722
	165 246.440	171 181.662	164 605.756	171 568.272
3	8.0	8.0	8.0	8.0
	382.667	385.333	380.0	388.0
	15 326.667	15 582.667	15 456.0	15 200.0
	591 237.867	606 616.0	589 598.667	607 777.333

Now dynamic scaling[12] predicts that ω will scale as $\xi^{-1.5}$. This implies that the terms in $S^\alpha(\boldsymbol{\tau})\langle\omega^2\rangle$ and higher on the right-hand side of the above equation tend to zero as the temperature approaches T_N. Since both $\chi^\alpha(\boldsymbol{\tau})$ and $S^\alpha(\boldsymbol{\tau})$ become infinite at T_N it is clear that their critical exponents must be the same and that

$$\lim_{T\to T_N+}[N\beta g^2\mu^2 S^\alpha(\boldsymbol{\tau})-\chi^\alpha(\boldsymbol{\tau})]=0.$$

APPENDIX B

In this Appendix formal expressions are given for $\chi(\mathbf{k})$ and for $S(\mathbf{k})$. Following Rushbrooke and Wood,[2,3] it is convenient to work in terms of the spin variable X, equal to $S(S+1)$, and in terms of the dimensionless reduced temperature θ, given by k_BT/J where J is the Heisenberg exchange parameter. Then

$$\chi(\mathbf{k})=Ng^2\mu^2\beta\tfrac{1}{3}X\sum_{r=0}^{\infty}x_r\theta^{-r}.$$

At $\mathbf{k}=0$, or at $\mathbf{k}=\boldsymbol{\tau}$ for the antiferromagnet, the coefficients x_r reduce to the coefficients a_r and a_r' given by Rushbrooke and Wood.

It is found that

$$x_0=1,$$

$$x_1=\tfrac{2}{3}X\Sigma_1,$$

$$x_2=\tfrac{1}{9}X[4X\Sigma_2-\Sigma_1-2z],$$

$$x_3=(X/27)[\tfrac{3}{5}(12X^2-8X+3)\Sigma_1+3z+8X^2\Sigma_3-4X\Sigma_2+8X(z-1)(3X-1)\Sigma_1-12X^2(2z-1)\Sigma_1-4Xp_1z],$$

$$x_4=(2X/81)\{-0.9[(6X^2-6X+2)\Sigma_1+(4X^2-4X+3)z]-3X(2X-1)(z-1)\times(3\Sigma_1+2z)+0.6p_1X[(56X^2-24X+3)\Sigma_1+6z]-4X^2p_2z+8X^2(3X-1)(z-2)p_1\Sigma_1+8X^2(3X-1)\Sigma_2[(z-1)^2-p_1](z-1)^{-1}+1.2X(12X^2-8X+3)\Sigma_2+4X^2(3X-1)\times(z-2)\Sigma_2-6X^2\Sigma_3+8X^3\Sigma_4+3X^2(2z-1)\times(3\Sigma_1+2z)-24X^3(z-1)p_1\Sigma_1-12X^3\Sigma_2\times(3z-2)-0.6X[z(z-1)-\Sigma_2]\}.$$

TABLE II. First four expansion coefficients of ξ^2 for the body-centered-cubic lattice with spin values S of $\frac{1}{2}$, 1, $\frac{3}{2}$, 2, $\frac{5}{2}$, and 3. The coefficients are expressed in units of the lattice constant a.

Spin S	v	v'	w	w'
$\frac{1}{2}$	0.5	0.5	0.5	0.5
	1.91667	2.08333	1.75	2.25
	5.83333	7.16667	6.58333	5.25
	17.025	23.13056	16.0	21.66667
1	1.3333	1.3333	1.3333	1.3333
	14.0	14.4444	13.5556	14.8889
	124.8148	134.2963	129.7778	120.2963
	1 082.8938	1 209.5704	1 066.2469	1 208.5185
$\frac{3}{2}$	2.5	2.5	2.5	2.5
	49.5833	50.4167	48.75	51.25
	849.8333	883.1667	866.9167	833.5833
	14 233.7917	15 090.0972	14 128.3333	15 133.3333
2	4.0	4.0	4.0	4.0
	127.333	128.667	126.0	130.0
	3 528.933	3 614.267	3 572.267	3 486.933
	95 742.667	99 288.089	95 317.733	99 553.733
$\frac{5}{2}$	5.833	5.833	5.833	5.833
	271.25	273.194	269.306	275.139
	11 023.315	11 204.796	11 115.028	10 933.546
	438 953.785	450 012.079	437 646.914	450 980.185
3	8.0	8.0	8.0	8.0
	510.667	513.333	508.0	516.0
	28 553.333	28 894.667	28 725.333	28 384.0
	1 565 230.4	1 593 849.422	1 561 876.0	1 596 566.667

The notation is the same as that of Rushbrooke and Wood, with z the number of nearest neighbors to any given atom, and p_1 and p_2 the number of closed nonintersecting circuits of three and four neighboring atoms, respectively, involving both a given atom and a particular one of its nearest neighbors. For the simple-cubic, body-centered-cubic, and face-centered-cubic lattices z equals 6, 8, and 12; p_1 equals 0, 0, and 4; and p_2 equals 4, 12, and 22, respectively.

Σ_r is the sum of the cosines of the scalar product of $\mathbf{k}$ and the end points of all the nonintersecting walks of r steps between neighboring atoms, starting from the origin. Formally

$$\Sigma_1=\sum_{\rho}\cos(\mathbf{k}\cdot\boldsymbol{\rho})$$

$$\Sigma_2=\sum_{\rho}\sum_{\rho'}\cos[\mathbf{k}\cdot(\boldsymbol{\rho}+\boldsymbol{\rho}')](1-\delta_{-\rho,\rho'})$$

$$\Sigma_3=\sum_{\rho}\sum_{\rho'}\sum_{\rho''}\cos[\mathbf{k}\cdot(\boldsymbol{\rho}+\boldsymbol{\rho}'+\boldsymbol{\rho}'')](1-\delta_{-\rho,\rho'})\times(1-\delta_{-\rho',\rho''})(1-\delta_{-\rho,\rho'+\rho''}),$$

etc.

In an analogous way $S(\mathbf{k})$ can be expanded to give

$$S(\mathbf{k})=\tfrac{1}{3}X\sum_{r=0}^{\infty}s_r\theta^{-r}.$$

It follows from Theorem II of Ref. 2 that all the terms in $S(\mathbf{k})$ for $r>0$ must be $\mathbf{k}$ dependent. At $\mathbf{k}=0$ the coefficients s_r are identical to the coefficients a_r given by Rushbrooke and Wood. The expansion gives

$$s_0=1,$$

$$s_1=\tfrac{2}{3}X\Sigma_1,$$

$$s_2=\tfrac{1}{9}X[4X\Sigma_2-3\Sigma_1],$$

$$s_3=(X/27)\{[2.4(3X^2-2X+2)+8X(z-1)(3X-1)-4Xp_1-12X^2(2z-1)]\Sigma_1-4X\Sigma_2+8X^2\Sigma_3\},$$

$$s_4=(X/81)([-9(2X^2-2X+1)-30X(2X-1)(z-1)+1.2Xp_1(56X^2-24X+9)-8X^2p_2+16X^2\times(3X-1)(z-2)p_1+30X^2(2z-1)-48X^3\times(z-1)p_1]\Sigma_1+\{16X^2(3X-1)[(z-1)^2-p_1]\times(z-1)^{-1}+2.4X(12X^2-8X+3)+8X^2\times(3X-1)(z-2)-24X^3(3z-2)\}\times\Sigma_2-12X^2\Sigma_3+16X^3\Sigma_4).$$

TABLE III. First four expansion coefficients of ξ^2 for the face-centered cubic lattice with spin values S of $\frac{1}{2}$, 1, $\frac{3}{2}$, 2, $\frac{5}{2}$, and 3. The coefficients are expressed in units of the lattice constant a.

Spin S	v	v'	w	w'
$\frac{1}{2}$	0.5	0.5	0.5	0.5
	2.9167	3.0833	2.75	3.25
	14.6667	16.6667	15.4167	13.4167
	68.1492	86.2659	68.6742	90.0076
1	1.3333	1.3333	1.3333	1.3333
	21.1111	21.5556	20.6667	22.0
	300.2222	314.4444	305.1852	290.9630
	4 031.8537	4 376.2882	4 043.0191	4 490.2783
$\frac{3}{2}$	2.5	2.5	2.5	2.5
	74.5833	75.4167	73.75	76.25
	2 020.6667	2 070.6667	2 037.75	1 987.75
	51 994.4735	54 127.7235	52 069.4318	54 959.4318
2	4.0	4.0	4.0	4.0
	191.333	192.667	190.0	194.0
	8 350.267	8 478.267	8 393.6	8 265.6
	346 945.285	354 828.485	347 254.218	358 376.618
$\frac{5}{2}$	5.833	5.833	5.833	5.833
	407.361	409.306	405.417	411.25
	26 018.222	26 290.444	26 109.935	25 837.713
	1 584 043.227	1 605 108.712	1 585 004.188	1 616 343.377
3	8.0	8.0	8.0	8.0
	766.67	769.33	764.0	772.0
	67 294.67	67 806.67	67 466.67	66 954.67
	5 634 609.12	5 678 462.98	5 637 091.52	5 707 792.85

APPENDIX C

In this Appendix the high-temperature expansions for ξ^2 are given. Using the methods described in the text, we can express ξ^2 as

$$\xi_x^2 = \sum_{r=1}^{\infty} v_r \theta^{-r} \qquad \text{and} \qquad \xi_s^2 = \sum_{r=1}^{\infty} w_r \theta^{-r}.$$

The coefficients of the analogous expansions for the antiferromagnet about the point $\mathbf{k}=\boldsymbol{\tau}$ will be called v_r' and w_r'.

The expressions for the higher terms in these expansions are rather long and cumbersome; only the first two terms will be quoted explicitly. Numerical values for the higher terms are given later in this Appendix.

The expansion gives

$$v_1 = v_1' = w_1 = w_1' = \tfrac{1}{9} X z \langle \rho_1^2 \rangle,$$

$$v_2 = (Xz/54)[4X(z-1)\langle \rho_2^2 \rangle - (4Xz+1)\langle \rho_1^2 \rangle],$$

$$w_2 = (Xz/54)[4X(z-1)\langle \rho_2^2 \rangle - (4Xz+3)\langle \rho_1^2 \rangle],$$

$$v_2' = (Xz/54)[4X(z-1)\langle \rho_2^2 \rangle - (4Xz-1)\langle \rho_1^2 \rangle],$$

$$w_2' = (Xz/54)[4X(z-1)\langle \rho_2^2 \rangle - (4Xz-3)\langle \rho_1^2 \rangle],$$

where $\langle \rho_r^2 \rangle$ is the mean-square distance between an atom and the set of atoms described in the cosine function of the definition of Σ_r (Appendix B).

Tables I–III give the values of these coefficients for the simple-cubic, body-centered-cubic, and face-centered-cubic lattices, respectively.

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