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Series Expansions for High-Temperature Dynamics of Heisenberg Paramagnets*

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High-temperature expansions are given for the second and fourth moments of the frequency in the spin-correlation function and the frequency-dependent susceptibility. The results are applicable to loose-packed cubic Bravais lattices with general spin values and nearest-neighbor interactions. At small wave vectors, for both the ferromagnet and the antiferromagnet, the series for the reciprocal of the second moment shows the expected divergence at the critical temperature. Insufficient terms are available to yield accurate critical indices. There is good agreement between the calculation and neutron-scattering experiments on $RbMnF_3$. Paramagnon peaks are not likely to be present at temperatures of order five to ten times the critical temperature in the antiferromagnets considered, but probably do occur at favorable points in the zone for the corresponding ferromagnets.

I. INTRODUCTION

The problem of describing the dynamics of a Heisenberg paramagnet has not been solved. For infinite temperatures, a number of papers have given different approximate treatments which are substantially consistent with one another.[1-9] In the temperature region near the critical ordering temperature, the theory of Kawasaki[10] and the dynamic scaling laws[11,12] give a description of the dynamics for long wavelengths.

The purpose of the present paper is to investigate the region of temperature intermediate between these two extremes. The method of moments[1,4,5] is used to describe the dynamics in conjunction with a high-temperature series expansion. This type of treatment appears to have been first used by Sears[13] and was developed in more detail by Tahir-Kheli and McFadden.[5] The present paper starts by repeating this latter work to eliminate some algebraic errors (pointed out in Refs. 6 and 14) and to give a further term in the second-moment expansion.

The form of the series at long wavelengths essentially gives a high-temperature expansion for some dynamic critical properties. Use of such expansions (for review, see Ref. 15) has proved to be one of the most powerful tools for investigating static critical properties; this present work appears to be the first application of this technique to dynamic properties.

The properties calculated are observable quantitatively by current neutron-scattering techniques. The physical principles and techniques involved have been described in the literature[16,17] and will not be discussed in the present paper. A few such measurements have been made and these are compared to the theory in Sec. IV of the paper.

II. FORMAL THEORY

First, formal expressions will be given for the dynamic spin-correlation function and the generalized susceptibility. Then the moments of their frequency spectrum are obtained and expressed as a high-temperature series expansion.

The dynamic spin-correlation function $S^{\alpha\alpha}(\vec{k}, \omega)$ is the spatial and temporal Fourier transform of the two-spin-correlation function and is given by

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

where the superscript $\alpha = x$, y, or z, N is the number of atoms in the crystal, $\vec{n}$ and $\vec{m}$ are atomic positions and $S_{\vec{m}}^{\alpha}(t)$ is the α component of spin on the atom at $\vec{m}$ at time t. The expectation value is to be evaluated at the temperature T of the crystal.

It follows that[1]

$$\int_{-\infty}^{\infty} S^{\alpha\alpha}(\vec{k}, \omega) \omega^p \, d\omega = N^{-1} \sum_{\vec{n}, \vec{m}} e^{i\vec{k}\cdot(\vec{n}-\vec{m})} \langle S_{\vec{n}}^{\alpha} \mathcal{L}^p S_{\vec{m}}^{\alpha} \rangle_T \,, \tag{2}$$

where p is a positive integer or zero and $\mathcal{L}$ is the Liouville operator representing a commutation with the Hamiltonian.

Following the method of Opechowski[18] and of Rushbrooke and Wood,[19,20] a high-temperature expansion can be made of the right-hand side of the above equation in powers of β $(=k_B T)^{-1}$ using the result

$$\langle S^{\alpha}_{\vec{n}}\,\mathcal{L}^p S^{\alpha}_{\vec{m}}\rangle_T=\sum_{r=0}^{\infty}\frac{(-\beta)^r}{r!}\langle \mathcal{H}_0^r S^{\alpha}_{\vec{n}}\,\mathcal{L}^p S^{\alpha}_{\vec{m}}\rangle_{\infty}\;, \tag{3}$$

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

It is usual to work in terms of a normalized pth moment of the spin-correlation function $\langle\omega_s^p\rangle$ given by

$$\langle\omega_s^p\rangle=\int_{-\infty}^{\infty}S^{\alpha\alpha}(\vec{k},\omega)\omega^p\,d\omega\Big/\int_{-\infty}^{\infty}S^{\alpha\alpha}(\vec{k},\omega)\,d\omega\;. \tag{4}$$

The denominator of the above expression is equal to the parameter $S(\vec{k})$ whose high-temperature expansion has been given in Ref. 14. {There is a minor error in Ref. 14. In the coefficients x_4 and s_4 of Appendix B, the terms $\Sigma_2[(z-1)^2-p_1](z-1)^{-1}$ should read $(z-1)\Sigma_2-p_1\Sigma_1$. Concomitant errors appear in Table III. The author wishes to thank Professor S. Rushbrooke for bringing this to his attention.} Inversion of this term and multiplication with the series for the numerator and collection of terms of the same order in β gives a power series for $\langle\omega_s^p\rangle$ in powers of β.

In an analogous manner we can make the same expansion for the imaginary part of the generalized susceptibility $\operatorname{Im}\chi^{\alpha\alpha}(\vec{k},\omega)$. It is convenient to take moments of this susceptibility divided by the frequency ω, since then we have the property that at infinite temperature all the moments $\langle\omega_\chi^p\rangle$ are identical with the moments of the correlation function $\langle\omega_s^p\rangle$. Following Marshall and Lowde,[16] Eq. (96), we can write for temperatures above the critical temperature

$$\omega^{-1}\operatorname{Im}\chi^{\alpha\alpha}(\vec{k},\omega)=\pi g^2\mu_B^2 N\beta[(1-e^{-\hbar\omega\beta})/\hbar\omega\beta]\,S^{\alpha\alpha}(\vec{k},\omega)\,,$$

where μ_B is the Bohr magneton and g the gyromagnetic ratio. In taking moments of this function and writing them as a power series in β we take all the same steps as with the spin-correlation function and further expand $e^{-\hbar\omega\beta}$ as a power series in β. This gives the result that the rth temperature coefficient of the pth moment of the susceptibility function contains the same term as in the correlation-function moments and also terms in the expansion of all higher moments of the correlation function up to the $(p+r)$th moment. The existence of such relations between the two sets of moments was first pointed out by Falk and Bruch.[21] The normalization of the susceptibility moments contains the function $\chi^{\alpha}(k)$ rather than $S^{\alpha}(k)$, but Ref. 14 lists this expansion as well as that for $S^{\alpha}(k)$ so no additional problems are introduced.

The main computation problem in determining the rth temperature coefficient of a pth moment is in evaluating the expectation value $\langle S^{\alpha}_{\vec{n}}S^{\alpha}_{\vec{m}}\mathcal{H}_0^{r+p}\rangle_{\infty}$ in all possible orders of arrangement for the two spin terms and the $(r+p)$ terms in $\mathcal{H}_0$. This is the same problem as that originally faced by Rushbrooke and Wood[20] in calculating the high-temperature expansion of the staggered susceptibility. This expansion uses the sum of the expectation values in all possible orders while the moments determination involves more complicated linear combinations arising from the application of the Liouville operator. In many cases, Rushbrooke and Wood were able to evaluate the terms in the susceptibility without in fact calculating all the individual expectation values. Nonetheless, their calculation forms an invaluable checking point for the present calculations. We have calculated all nonzero expectation values for $r+p\leq 5$ and their sums agree with Rushbrooke and Wood's calculation. The expectation values for $r+p\leq 4$ were in fact derived for a previous paper[14] dealing with static properties. The computational method used is essentially the same as that of Rushbrooke and Wood[19] and involves the same restrictions of a loose-packed cubic Bravais lattice with nearest-neighbor interactions only.

A further check on the data is furnished by the constraint that all moments, to all orders of β, go to zero[1] at $\vec{k}=0$. This property can be checked separately for each particular grouping of the $r+p$ lines on the graphs of Rushbrooke and Wood.[19,20]

The detailed forms of the moments obtained are listed in the Appendix. Except for the highest term in the second moment these expressions have also been calculated by Tahir-Kheli and McFadden.[5] Our data disagree with those calculations for the term in β^2 for the second moment and for the term in β for the fourth moment, though lower terms in β agree. The discrepancies in both cases are for the two lowest terms in the moments when expressed as series in powers of $S(S+1)$, while the highest term agrees. This means that in a practical computation of the moments, the differences are of little importance except for the case $S=\frac{1}{2}$. It is believed that the present calculation is correct, however; errors in the earlier work have been noted by the authors of Refs. 6 and 14.

Since submission of this paper for publication, Dr. K. Binder has drawn my attention to a paper[21a] that calculates a parameter equivalent to the spatial Fourier transform of the term c_1 listed in the Appendix. There appear to be some discrepancies between this calculation and the terms listed in the present paper for graphs with two and three vertices, though the four-vertex graphs agree.

III. CRITICAL DYNAMICS

A. Ferromagnets

Ferromagnetic spin properties show critical divergences as the temperature tends to the critical temperature and the wave vector tends to zero.

Weak singularities in properties may occur at general wave vectors,[22] but these are not likely to be apparent in short series such as are given in this paper. Thus only the small wave-vector properties are considered in this section. de Gennes[7] has shown that at small wave vectors

$$\langle \omega_X^n \rangle = d_n k^2 ,$$

where n is a positive even integer. For cubic symmetry, d_n is independent of the direction of $\vec{k}$.

The coefficients d_n can be expressed in terms of the high-temperature series for $\langle \omega^2 \rangle$ and $\langle \omega^4 \rangle$ by expanding the cosine functions defining the terms σ in Appendix A. Table I lists the results for simple-cubic and body-centered-cubic lattices with spins of $\frac{1}{2}$, 1, and $\frac{5}{2}$. The series for the reciprocal of d_2 is also given. The series are normalized in all cases to give a leading temperature-independent term of unity.

It is to be expected that there will be a critical "slowing down"[10,16] so that the coefficients d_n will tend to zero as the critical temperature is approached from above. The coefficients given in Table I do show such a tendency. This is perhaps illustrated more clearly in the series for $\langle \omega^2 \rangle^{-1}$ which should diverge at T_c. For spin $\frac{5}{2}$, with the simple-cubic and the body-centered-cubic lattices, the ratios of the successive terms in the expansion are 35.5, 26.1, and 26.9 and 47.1, 32.8, and 38.0, respectively. One might expect rather generally that these ratios will tend to values of J/kT_c as one goes to higher terms in the expansion[15] if the quantity $\langle \omega^2 \rangle^{-1}$ does indeed diverge at T_c. For the two lattices the critical ratios are 24.8 and 34.7, respectively,[19] so that the series are certainly consistent with the expected divergences. Unfortunately we do not have a sufficient number of terms for any reliance to be placed on critical indices obtained from these ratios.

The treatment is believed to be the first high-temperature series for dynamic critical properties. It would be desirable to connect this work to the hydrodynamic approach to critical properties of Kawasaki[10] and of Halperin and Hohenberg.[12] Unfortunately this is not straightforward since hydrodynamic theory gives a Lorentzian frequency dependence at small wave vectors and frequencies. The width of the Lorentzian function is Dk^2, where D is the diffusion constant. The difficulty that arises is that the moments of a Lorentzian function are infinite while the actual moments are known to be finite. There must be some kind of truncation of the Lorentzian form at high frequencies. Models to describe this truncation have been proposed in Refs. 1–3. The results show that the relationship between the diffusion constant and the moments is model dependent. Thus it may not be possible to relate simply the critical behavior of the diffusion constant to that of the moments.

B. Antiferromagnets

A similar treatment to that of Sec. III A can be carried out for the Heisenberg antiferromagnet. Here it is appropriate to calculate the moments at a wave vector $\vec{\tau}$ corresponding to the antiferromagnetic reciprocal lattice. Table II lists the results for the simple-cubic and body-centered-cubic lattices with spins of $\frac{1}{2}$, 1, and $\frac{5}{2}$. The series for the reciprocal of the second moment is also given. As with the ferromagnet, the series are normalized in all cases to give a temperature-independent term of unity.

TABLE I. The first four expansion coefficients of the second moment and its inverse for a simple-cubic and a body-centered-cubic ferromagnetic lattice with spin values S of $\frac{1}{2}$, 1, and $\frac{5}{2}$. The expansion is in powers of $\theta^{-1} = \beta J$ at long wavelengths and has been normalized to give a leading temperature-independent term of unity.

Spin S	Coefficient	Second moment sc lattice	Second moment bcc lattice	Inverse second moment sc lattice	Inverse second moment bcc lattice	Fourth moment sc lattice	Fourth moment bcc lattice
$\frac{1}{2}$	0	1	1	1	1	1	1
	1	−3.5	−4.5	3.5	4.5	−4.389	−5.654
	2	3.75	6.583	8.5	13.667		
	3	−1.583	−4.5	15.04	36.375		
1	0	1	1	1	1	1	1
	1	−8.5	−11.167	8.5	11.167	−10.62	−13.91
	2	20.0	38.67	52.25	86.03		
	3	−21.43	−171.0	295.6	699.8		
$2\frac{1}{2}$	0	1	1	1	1	1	1
	1	−35.500	−47.167	35.500	47.167	−44.03	−58.35
	2	333.35	677.5	926.9	1547.2		
	3	−3897.7	−17726	24969	58745		

As with the ferromagnet, it might be expected that there will be a critical "slowing down" so that the moments tend to zero as the Néel temperature is approached from above. The coefficients in Table II do reveal such a slowing down tendency. We can adopt the same procedure as with the ferromagnetic case and write down the ratio of successive terms in the series expansion for $\langle \omega^2 \rangle^{-1}$. With spin $\frac{5}{2}$ and simple-cubic and body-centered-cubic lattices these ratios are 34.5, 26.2, and 27.7 and 46.2, 32.8, and 38.8, respectively. For these two lattices the critical ratios are 25.1 and 35.0, respectively,[20] so that the series are consistent with the expected divergences. There is not, however, a sufficient number of terms for any reliance to be placed on critical indices obtained from these ratios.

For the Heisenberg antiferromagnet, dynamic scaling[12] predicts that the frequency function will have a characteristic width at $q=\vec{\tau}$ that scales as the reduced temperature to a power of $3\nu/2$ (≈ 1.04). It seems reasonable to take this characteristic width as proportional to the square root of the second moment. This will be correct so long as the dominant contributions to the second moment come from sufficiently low frequencies that the critical transition dominates the properties.

IV. NUMERICAL VALUES

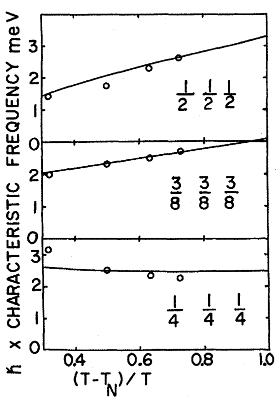

FIG. 1. Characteristic frequencies in rubidium manganese fluoride as a function of temperature at the points $\frac{1}{2}\frac{1}{2}\frac{1}{2}$, $\frac{3}{8}\frac{3}{8}\frac{3}{8}$, and $\frac{1}{4}\frac{1}{4}\frac{1}{4}$. Open circles are the experimental results of Tucciarone *et al.* (Ref. 23), and the lines the results of the calculations given in the present paper. No disposable parameters are used in the calculations.

Much work in evaluating numerical results has already been carried out by McFadden and Tahir-Kheli.[5] A lot of their emphasis was on limitingly high temperatures, however, and in this section we look at lower temperatures. Particular emphasis is placed on comparison to the recent work of Tucciarone, Corliss, and Hasting[23] on rubidium manganese fluoride. They parametrize their frequency spectra by frequencies ω_T such that half the area under a spectrum lies between frequencies $-\omega_T$ and $+\omega_T$. If the spectra were Gaussian in shape this would correspond to

$$\omega_T = 0.676\,\langle \omega^2 \rangle^{1/2} .$$

Figure 1 shows the calculated values of ω_T using this equation and also the data of Tucciarone *et al.* The agreement is reasonably good seeing that no disposable parameters have been used in the calculation.

It is also of interest to examine the temperature variation of the line shape. This is conveniently done[4] in terms of the parameter α given by

$$\alpha = \frac{\langle \omega^4 \rangle}{3\langle \omega^2 \rangle^2} - 1 .$$

α is zero for a Gaussian line shape, infinite for a

TABLE II. The first four expansion coefficients of the second moment and its reciprocal for a simple-cubic and a body-centered-cubic antiferromagnetic lattice with spin values S of $\frac{1}{2}$, 1, and $\frac{5}{2}$. The moments are taken at the antiferromagnetic reciprocal lattice point ($\frac{1}{2}\frac{1}{2}\frac{1}{2}$ for the simple-cubic lattice, 100 for the body-centered cubic lattice) and are expressed in powers of $\theta^{-1} = \beta |J|$. The expansions have been normalized to give a leading temperature-independent term of unity.

		Second moment		Inverse second moment		Fourth moment	
Spin S	Coefficient	sc lattice	bcc lattice	sc lattice	bcc lattice	sc lattice	bcc lattice
$\frac{1}{2}$	0	1	1	1	1	1	1
	1	−2.5	−3.5	2.5	3.5	−3.648	−5.311
	2	−0.25	1.25	6.5	11.0		
	3	0.417	−3.5	16.46	37.625		
1	0	1	1	1	1	1	1
	1	−7.5	−10.167	7.5	10.167	−11.43	−15.87
	2	9.333	24.444	46.917	78.917		
	3	−19.907	−149.611	301.782	703.412		
$2\frac{1}{2}$	0	1	1	1	1	1	1
	1	−34.5	−46.167	34.5	46.167	−53.50	−72.92
	2	286.68	615.29	903.56	1516.07		
	3	−3769.66	−17178.6	25052.1	58764.3		

Lorentzian, and $-\frac{2}{3}$ for two δ-function peaks at $\pm\omega_c$.

Since just the first temperature coefficient of $\langle\omega^4\rangle$ is known, only the first temperature coefficient of α can be determined. When this term is numerically determined from the expressions given in the Appendix, it is found to be small throughout the zone in both body-centered-cubic and simple-cubic lattices. This is because the temperature dependence of $\langle\omega^4\rangle$ tends to be of the same sign as that of $\langle\omega^2\rangle$ and of somewhat larger magnitude. It follows that at high temperatures, where the first term dominates, the main effects of temperature changes are to be observed in changes of width and area of a frequency distribution rather than in changes of shape.

It is of particular interest to examine the value of α at the points in the zone where it has its lowest value. With $S=\frac{5}{2}$, this occurs at the point $\frac{1}{2}\frac{1}{2}\frac{1}{2}$ for the simple-cubic lattice where $\alpha=-0.225$ at $T=\infty$ and at the point 100 for the body-centered-cubic lattice where $\alpha=-0.211$ at $T=\infty$. These points give spectra that are more "square topped" than a Gaussian shape, though the value of α is not sufficiently negative that it is necessary to have maxima at frequencies other than zero. We believe that a value of α less than -0.4 is required for it to be formally necessary for a peak to have at least one maximum at a nonzero frequency, though we have no proof of this.

For smoothly behaved distributions, values of α in the region -0.25 to -0.3 would probably ensure the existence of peaks at nonzero frequencies. It is apparent that not-very-large changes in α might be required at the particular points in the zone mentioned above in order to give such peaks. However, calculations show that the first temperature coefficient of α at both the points mentioned above is positive. Thus the effect of decreasing the temperature is to make the peaks more nearly Gaussian in shape. This conclusion is qualitatively confirmed by the experiments of Tucciarone *et al.*[23] for rubidium manganese fluoride and of Schulhof[24] for manganese fluoride (in this case the lattice is body-centered tetragonal not body-centered cubic as assumed in the present calculations).

The calculations show that there will be no separation of the scattering into discernible "paramagnon" peaks at high temperatures. With only the first temperature coefficient of the fourth moment, we are not able to make any predictions about the behavior near to the critical temperature.

In contrast to this result the analogous calculation for the ferromagnet shows a negative value of the first temperature coefficient of α in a ferromagnet at $\frac{1}{2}\frac{1}{2}\frac{1}{2}$ for the simple-cubic lattice and 100 for the body-centered-cubic lattice. Thus at a temperature of ten times the critical temperature α is -0.280 in the simple-cubic lattice and -0.263 in the body-centered-cubic lattice. It is likely that paramagnon peaks will be present in both lattices at relatively high temperatures (of order $5T_c$) at the particular points in reciprocal space mentioned above. Experimentally, this effect has not been looked for owing to the dearth of good Heisenberg ferromagnets.

APPENDIX

In this appendix formal expressions are given for the high-temperature expansions of the moments. Following Rushbrooke and Wood,[19,20] 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

$$\langle\omega_s^2\rangle=\frac{X^2J^2}{S(\vec{\mathrm{k}})}\sum_{r=0}^{\infty}a_r\Theta^{-r}\ .$$

It is found that

$$a_0=\tfrac{8}{9}(z-\sigma_1)\ ,$$

$$a_1=-\tfrac{4}{9}(z-\sigma_1)\ ,$$

$$a_2=\tfrac{4}{405}(z-\sigma_1)[8(5p_2-3)X^2+4(15z-8)X+9]-\tfrac{8}{27}X[z(z-1)-\sigma_2]\ ,$$

$$\begin{aligned}a_3=-\tfrac{4}{1215}\{&(z-\sigma_1)[18(X^2+6X-1)\\&+60(z-1)(X^2+3X)-210X^2(2z-1)]\\&+40X^2\sigma_1(z-1)^2+20X^2\sigma_1p_2\\&+120(X^2-X)(z^2-z-\sigma_2)-80X^2\sigma_2(z-1)\\&-60X^2\sigma_2'+40X^2\sigma_3+100X^2zp_2\}\ .\end{aligned}$$

The notation is the same as that of Refs. 14, 19, and 20 except that the symbol Σ of Ref. 14 has been replaced by σ. z is the number of nearest neighbors to any given atom, p_2 is the number of nonintersecting circuits of four neighboring atoms involving both a given atom and a particular one of its nearest neighbors. For the simple-cubic and body-centered-cubic lattices, z equals 6 and 8 and p_2 equals 4 and 12, respectively.

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

Formally,

$$\sigma_1=\sum_{\rho}\cos(\vec{\mathrm{k}}\cdot\vec{\rho})\ ,$$

$$\sigma_2=\sum_{\rho}\sum_{\rho'}\cos[\vec{\mathrm{k}}\cdot(\vec{\rho}+\vec{\rho}\,')](1-\delta_{-\vec{\rho},\vec{\rho}\,'})\ ,$$

$$\sigma_3=\sum_{\rho}\sum_{\rho'}\sum_{\rho''}\cos[\vec{\mathrm{k}}\cdot(\vec{\rho}+\vec{\rho}\,'+\vec{\rho}\,'')](1-\delta_{-\vec{\rho},\vec{\rho}\,'})\times(1-\delta_{-\vec{\rho}\,',\vec{\rho}\,''})(1-\delta_{-\vec{\rho},\vec{\rho}\,'+\vec{\rho}\,''})\ .$$

The summation giving σ_2' is similar to that giving

σ_2 with the added restriction that $(\vec{\rho}+\vec{\rho}\,')$ must be an atom on a closed nonintersecting circuit of four neighboring atoms including the atom at the origin. Formally,

$$\sigma_2' = \sum_{\vec{\rho}} \sum_{\vec{\rho}\,'} \cos[\vec{k}\cdot(\vec{\rho}+\vec{\rho}\,')]\,(1-\delta_{-\vec{\rho},\vec{\rho}\,'})$$

$$\times \sum_{\vec{\rho}\,''} \sum_{\vec{\rho}\,'''} \delta_{-\vec{\rho},\vec{\rho}\,'+\vec{\rho}\,''+\vec{\rho}\,'''}\,(1-\delta_{-\vec{\rho}\,',\vec{\rho}\,''})\ .$$

At $\vec{k}=0$, $\sigma_2' = 2zp_2$.

An analogous series can be formed for $\langle \omega_X^2 \rangle$ by the equation

$$\langle \omega_X^2 \rangle = \frac{X^2 N g^2 \mu_B^2 J}{\Theta \chi(\vec{k})} \sum_{r=0}^{\infty} b_r \Theta^{-r}\ .$$

It is found that

$$b_0 = a_0\ ,$$

$$b_1 = a_1\ ,$$

$$b_2 = \tfrac{32}{405}(z-\sigma_1)[(5p_2-3)X^2-(5z-2)X+3]\ ,$$

$$b_3 = \tfrac{4}{243}(z-\sigma_1)(24zX^2-20p_2X^2+30zX-12X-9).$$

The expressions for the b coefficients are simpler than those for the a coefficients.

The fourth moment can be expanded in an analogous manner, giving

$$\langle \omega^4 \rangle = \frac{X^2 J^4}{s(\vec{k})} \sum_{r=0}^{\infty} c_r \Theta^{-r}\ .$$

We should define series for both the correlation function and the susceptibility, as we did for the second moments, but for the terms with r equal to 0 or 1 the two sets of coefficients are identical. Thus only the one series is written down here with the coefficients given by

$$c_0 = \tfrac{32}{27} X[2(5z-2)(z-\sigma_1)-3(z^2-z-\sigma_2)] - \tfrac{16}{9}(z-\sigma_1)\ ,$$

$$\begin{aligned} c_1 = -\tfrac{16}{405}[&9(z-\sigma_1)(-8X^2+22X-7) \\ &+30(z-\sigma_1)(z-1)(-8X^2+11X) \\ &+40\sigma_1(z-1)^2X^2+120(z^2-z-\sigma_2)(X^2-X) \\ &\quad -80\sigma_2(z-1)X^2+40\sigma_3X^2+60X^2(2p_2\sigma_1-\sigma_2')]\ . \end{aligned}$$

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