

# Critical Properties and Exchange Interactions of Antiferromagnetic A-spinel Lattice: A Study Through High-temperature Series Expansions.

M. Hamedoun<sup>a,\*</sup>, H. Bakrim<sup>a</sup>, A. Hourmatallah<sup>a,b</sup> and N. Benzakour<sup>a</sup>

a- *Laboratoire de Physique du Solide, Université Sidi Mohammed Ben Abdellah, Faculté des sciences Dhar Mahraz, BP 1796, Fes, Morocco.*

b- *Equipe de Physique du Solide, Ecole Normale Supérieure, B. P 5206, Bensouda, Fes, Morocco.*

We derive high- temperature series expansions for the spin correlation functions of the A-spinel lattice. The development is extended to the order 6 in  $\beta = \frac{1}{k_B T}$  with nearest-neighbour and next-nearest neighbour interactions. The

results are given for various neighbour correlation functions (up to the third). The behaviour with the temperature is presented. The critical region is studied by applying the Padé approximants method to the corresponding high- temperature series expansion of the magnetic susceptibility and the correlation length. The approach is applied to the experimental results of the particular system A-spinel  $\text{CoCo}_2\text{O}_4$ . The following estimates are obtained for the familiar critical exponents:  $\gamma = 1.382 \pm 0.010$  and  $\nu = 0.701 \pm 0.012$ .

**Keywords:** high-temperature series expansions, spin correlation functions, critical exponents, exchange interactions.

## I. Introduction

Materials with spinel structures are of continuing interest because of their wide variety of physical properties which is of major importance in most fields of pure and applied solid state physics. This is essentially related to the existence of two types of crystallographic sublattice, the tetrahedral (A) and the octahedral (B), available for the metal ion.

Much previous works, using series expansion methods for the spinel lattice, have restricted themselves to B-spinel lattice (only B ion is magnetic) [1, 2]. Nevertheless, the A-spinel (only A ion is magnetic) is of considerable interest in the theory of the magnetism. This interest arises because of the A-spinel lattice exhibits weak magnetism which coexists with the superconductivity behaviour [3]. In this latter, the A magnetic ions are located in tetrahedral sites of the spinel lattice.

In this work, it is intended to extend the development of high-temperature series expansion (HTSE) of the spin correlation functions to the order 6 in  $\beta = \frac{1}{k_B T}$  with nearest neighbour

(n n) and next-nearest neighbour (n n n) interactions,  $J_1$  and  $J_2$  respectively, for the A-spinel lattice  $\text{AB}_2\text{X}_4$ .

To deduce the spin correlation function:

$$\gamma_{i,j} = \frac{\langle \vec{S}_i \vec{S}_j \rangle}{S(S+1)}$$

between spins at sites i and j, we have used the diagrammatic representation performed by Stanley and Kaplan (SK) [4]. Their

method is general and can be applied to any lattice. We have calculated the spin correlation functions between first, second and third n n spins  $\gamma_1$ ,  $\gamma_2$  and  $\gamma_3$  respectively. The magnetic susceptibility  $\chi$  and the correlation length  $\xi$  have been derived. The Padé approximants analysis of the exact HTSE of the magnetic susceptibility has been shown to be a useful method to study the critical properties of real magnetic systems[5, 6]. We shall use this method to analyse the critical region of the A-spinel lattice.

The study of critical temperature versus exchange constants has been usually restricted in the literature to the case of  $J_1$ . On the other hand the application of the final results of some available theories to analysis of experimental data is not as straight forward as desired. In this work, we have estimated the critical temperature  $T_N$  as a function of the ratio  $\frac{J_2}{J_1}$  in the case of A-spinel lattice. The

approximation reported in this work can be used to calculate, without ambiguity, the  $J_2$  by using the data of  $T_N$  and  $J_1$  reported in the literature and obtained from experimental measurements.

The approximation we present in this work is applied to the particular A-spinel lattice  $\text{CoCo}_2\text{O}_4$ . This system is ordered antiferromagnetically with  $T_N = 34$  K and  $J_1 = -0.169$  K ( $0.05 \text{ cm}^{-1}$ ) [3]. The obtained  $J_2$  is expected to be 1.321 K. The critical exponents  $\gamma$  for the magnetic susceptibility and  $\nu$  for the correlation length are found to be equal to 1.382 and 0.701 respectively. These values are in good agreement with those of 3D Heisenberg

model namely,  $1.3866 \pm 0.0012$  and  $0.7054 \pm 0.0011$  [7, 13].

## II. Theory and results

Stanley and Kaplan (SK) [4, 8, 9] have performed a diagrammatic representation of the terms of the series, which permits the calculation of the zero-field spin correlation function:  $\gamma_{i,j} = \frac{\langle \vec{S}_i \vec{S}_j \rangle}{S(S+1)}$

between spins at sites  $i$  and  $j$ . Their method is general and can be applied to any lattice. This semiclassical treatment is a simplification of the more complex procedure of Rushbrooke and Wood [10] used for calculation of the susceptibility in the quantum-mechanical case.

In this work, we have calculated the spin correlation function between first, second and third nearest-neighbour spins  $\gamma_1$ ,  $\gamma_2$  and  $\gamma_3$  respectively to order 6 in  $\beta = \frac{1}{k_B T}$  with nearest-neighbour (n.n) and next nearest-neighbour (n.n.n) constant couplings. Starting with the zero-field Heisenberg Hamiltonien:

$$H = -2J_1 \sum_{ij} \vec{S}_i \vec{S}_j - 2J_2 \sum_{ik} \vec{S}_i \vec{S}_k \quad (1)$$

where the first and second summation runs over all pair of nearest and next nearest neighbours respectively.

The expansion of the spin correlation function in power of  $\beta$  is obtained as follows [8]:

$$\langle \vec{S}_i \vec{S}_j \rangle = \frac{\text{Tr} \vec{S}_i \vec{S}_j e^{-\beta H}}{\text{Tr} e^{-\beta H}} = \sum_{l=0}^{\infty} \frac{(-1)^l \alpha_l \beta^l}{l!} \quad (2)$$

with

$$\alpha_l = v_l - \sum_{k=0}^{l-1} C_k^l \alpha_k \mu_{l-k}, \quad v_m = \langle \vec{S}_i \vec{S}_j H^m \rangle_{T=0}$$

$$\text{and } \mu_m = \langle H^m \rangle_{T=0}. \quad (3)$$

The calculation of the coefficients of the  $\gamma_i$  according to the diagrammatic methods involves two distinct phases:

- the first step is to find and to catalogue all diagrams or graphs which can be constructed from dashed line connecting the site  $i$  and  $j$  and  $l$  straight lines, and to determine a diagrams whose contribution is nonvanishing.
- The second one consists of computing the number of times that a diagram occurs in the lattice.

The step (a) has already been accomplished in SK work.

For each topological form of a diagram, a full line can either represent  $J_1$  or  $J_2$ . We must, then derive from each topological from a class of

diagrams. Each of them represents a term of the series  $J_1^m$  and  $J_2^n$  ( $n, m=0, 1, \dots, 1, m+n=1$ ). it is especially the limiting factor in how far one can carry the expansion.

Equation (2) combined with the results of appendix A<sub>1</sub> given in Ref. [2] and the results of appendix of this work permits the computation of spin correlation functions  $\gamma_i$  ( $i=1, 2, 3$ ) in terms of powers of  $\beta$  and mixed powers of  $J_1$  and  $J_2$ .

The wavelength-dependent susceptibility  $\chi(\vec{q})$  and the correlation function  $S(\vec{q})$  may be expressed as follows:

$$\chi(\vec{q}) = g \mu_B^2 \beta \sum_{i,j} \langle \vec{S}_i \vec{S}_j \rangle e^{-i\vec{q} \vec{R}_{ij}} \quad (4)$$

$$S(\vec{q}) = \sum_{i,j} \langle \vec{S}_i \vec{S}_j \rangle e^{-i\vec{q} \vec{R}_{ij}} \quad (5)$$

where  $\mu_B$  is the Bohr magneton,  $g$  the gyromagnetic ratio,  $\vec{R}_{ij}$  the separation vector between spin  $i$  and  $j$  and  $\vec{q}$  the wave vector.

In order to obtain a qualitative measure of the correlation length  $\xi(T)$  for a given temperature  $T$ , we expand the correlation function  $S(\vec{q})$  in a Taylor expansion about the magnetic reciprocal lattice  $\vec{q}_0$  of the given system [11]:

$$S(\vec{q}) = S(\vec{q}_0) \left[ 1 - \xi^2(T) (\vec{q} - \vec{q}_0)^2 + 0(\vec{q} - \vec{q}_0)^4 \right] \quad (6)$$

Recasting this in the Ornstein- Zernike from, the following asymptotic form is obtained:

$$S(\vec{q}) = \frac{S(\vec{q}_0) \kappa^2(T)}{[\kappa^2(T) + (\vec{q} - \vec{q}_0)^2]} \quad (7)$$

where  $\kappa(T) = \xi^{-1}(T)$ .

For an ideal antiferromagnetic A-spinel structure, the spins are ordered along the wave vector  $\vec{q} = (0,0,q)$ , and one can obtain:

$$S(q) = 2 \left[ 1 + 4\gamma_1 \cos\left(\frac{q\pi}{2}\right) + 2\gamma_2 (2 + 4\cos(q\pi)) + \gamma_3 \left( 4\cos\left(\frac{3q\pi}{2}\right) + 8\cos\left(\frac{q\pi}{2}\right) \right) \right] \quad (8)$$

Expanding the cosines of this equation in the Taylor expansion about  $\vec{q} = \vec{q}_0$  and using equation (6), we obtain:

$$\left( \frac{\xi}{a} \right)^2 = \frac{1}{S(q_0)} \left\{ \frac{1}{2} \gamma_1 - 4\gamma_2 + \frac{11}{2} \gamma_3 \right\} \quad (9)$$

where  $a$  is lattice parameter. The formulas of  $\left( \frac{\xi}{a} \right)$

and  $S(q)$  are qualitatively similar to those obtained in the case of B-spinel lattice [2].

The simplest assumption that one can make concerning the nature of the singularity of the magnetic susceptibility  $\chi(q)$  and the correlation

length  $\xi(T)$  is that in the neighbourhood of the critical points the above two functions exhibit an asymptotic behaviour.

$$\chi(q)\alpha(T_N - T)^{-\gamma} \quad (10)$$

and

$$\xi^2(T)\alpha(T_N - T)^{-2\nu} \quad (11)$$

$T_N$  represents the critical temperature, deduced by the dlog Padé- approximate (PA) methods,  $\gamma$  and  $\nu$  are the critical exponents.

The [M, N] PA to the series  $\chi(\beta)$  is a rational  $P_M/Q_N$  with  $P_M$  and  $Q_N$  polynomials of degrees M and N in  $\beta$  satisfying

$$\frac{d}{d\beta} \log \chi(\beta) = \frac{P_M}{Q_N} + O(\beta^{M+N+1}).$$

This enables us to develop the expression of susceptibility and the correlation length as power of the ratio  $\alpha = \frac{J_2}{J_1}$  and of the term  $\tau = \frac{2S(S+1)J_1}{k_B T}$  in

the case of antiferromagnetic A-spinel lattice:

$$\chi = g\mu_B^2 \beta \sum_{m=0}^n \sum_{n=1}^6 a(m, n) \alpha^m \tau^n \quad (12)$$

$$\xi^2(T) = \sum_{m=1}^n \sum_{n=1}^6 b(m, n) \alpha^m \tau^n \quad (13)$$

The coefficients  $a(m, n)$  and  $b(m, n)$  are presented in Tables 1 and 2.

We have followed this procedure for several values of first- and second- neighbour exchange constants. As results of this calculation, we have estimated for an antiferromagnetic A-spinel system ( $J_1 < 0$ ), the dependence of the  $\tau_N = \frac{2S(S+1)J_1}{T_N}$  as function of the

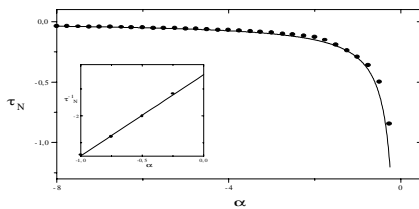


Fig.1 : The dependence of  $\tau_N = \frac{2S(S+1)J_1}{T_N}$  on the ratio  $\alpha = \frac{J_2}{J_1}$  for a general spin S Heisenberg model with ideal antiferromagnetic ordering of the A-spinel lattice. Inset

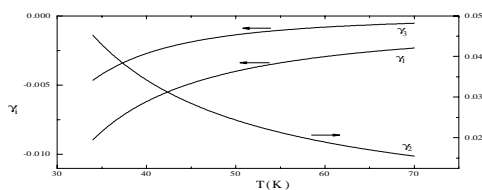


Fig.2 : The spin correlation functions  $\gamma_i$  ( $i=1, 2, 3$ ) versus temperature for  $\text{CoCo}_2\text{O}_4$  compound.

shows  $\frac{1}{\tau_N}$  against  $\alpha$  (The solid lines are guides to the eye).

ratio  $\alpha = \frac{J_2}{J_1}$ . The results have been calculated for

32 values of  $\alpha$  and are plotted in Fig.1. This figure beautifully illustrates the fact that as  $J_2$  ( $J_2 > 0$ ) decreases with respect to a reference constant value  $J_1$ , the system becomes less stable. No acceptable convergence was found for the case where  $J_2$  was antiferromagnetic ( $J_2 < 0$ ). This is because several poles were significant and none could be estimated with sufficient accuracy to be able to extract  $T_N$  successfully. The variation of  $T_N$  with  $\alpha$  can be well represented by

$$T_N(\alpha) = T_N(0)[1 + m\alpha] \quad (14)$$

in the range  $-1 \leq \alpha \leq 0$  with  $m$  is estimated to be equal to -6.477.  $T_N(0)$  is the Néel temperature of the lattice when only nearest-neighbour interactions are present. This result is similar to that obtained in Ref. [6].

The obtained results are applied to the particular A-spinel insulator compound  $\text{CoCo}_2\text{O}_4$ . This system ordered antiferromagnetically below  $T_N = 34$  K with a Curie-Weiss constant  $\theta$  equal to -108 K. Only  $\text{Co}^{2+}$  ion on tetrahedral sites were magnetic with spin value  $S = \frac{3}{2}$ .

Because of the existence of the transferred hyperfine coupling between the nuclear spin of the  $\text{Co}^{3+}$  ion at the octahedral B-site and the electron spins of the  $\text{Co}^{2+}$  ions, the mean field approximation cannot be applicable for this structure. Fukai and al [3] have estimated the first exchange interaction constant to be  $J_1 = -0.169$  K ( $0.05 \text{ cm}^{-1}$ ). Using the result presented in Fig.1, we have obtained  $J_2$  equal to 1.321 K.

The values of  $J_1$  and  $J_2$  are used to compute the evolution of the spin correlation's functions with the temperature  $\gamma_i$  ( $i=1, 2, 3$ ). The Fig. 2 shows

this evolution. The main feature of these curves is the decrease with T, i.e the order is destroyed by thermal disorder. For all temperatures, the  $\gamma_1$  and  $\gamma_3$  are negative and the  $\gamma_2$  is positive. We remark that all the three correlation functions still not vanish at high temperature. This may suggest the existence of short range order in the  $\text{CoCo}_2\text{O}_4$  insulator for temperature above  $T_N$ .

The thermal variation of  $\xi(T)$  is presented in Fig. 3. The result exhibits an unexpected feature: the onset of long range order at  $T_N$  is not associated with a maximum in  $\xi(T)$ . Indeed  $\xi(T)$  increases on further cooling. There is possibly a break in the slope of the curve of  $\xi(T)$  against temperature at

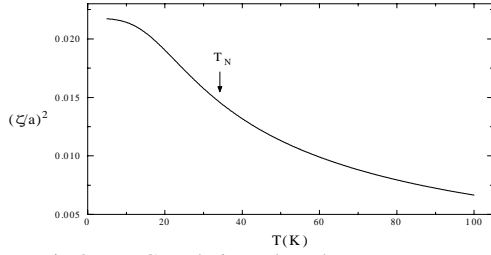


Fig.3 : Correlation length versus temperature in  $\text{CoCo}_2\text{O}_4$ . The arrows indicate the critical temperature  $T_N$ .

$T_N$ , a similar behaviour is observed in the system  $\text{KMn}_x\text{Zn}_{1-x}\text{F}_3$  near the percolation threshold [12]. An explanation based on the cluster topology of spins in the backbone of the infinite cluster is proposed [12].

We have obtained estimates of  $\gamma$  and  $\nu$  from the residues of the singularities in the sequences of  $[M, N]$  Padé approximants to the logarithmic derivatives of susceptibility and the correlation length series. Estimates of  $\gamma$  and  $\nu$  obtained in this way are close to  $1.382 \pm 0.010$  and  $0.701 \pm 0.012$  respectively.

### III- Conclusion

High temperature series expansion ( HTSE ) of the spin correlation functions of the A-spinel lattice are computed to order 6 in  $\beta = \frac{1}{k_B T}$  for

Heisenberg model having both nearest- and next nearest- neighbour exchange integrals,  $J_1$  and  $J_2$  respectively. HTSE extrapolated with Padé approximation (PA) method are shown to be convenient to provide valid estimation of parameters associated with the critical region. The theoretical considerations provide a useful tool for a straightforward interpretation and understanding of experimental data of any A-spinel lattice. The PA method is used to study the dependence of the critical temperature on the ratio of the relative strength of first- and second neighbour interactions

$\alpha = \frac{J_2}{J_1}$ . It is found that the variation is well

represented by  $T(\alpha) = T(0)[1 - 6.477\alpha]$  for values of  $\alpha$  in the range  $-1 \leq \alpha \leq 0$ .  $T(0)$  is the critical temperature of the nearest-neighbour model. The application of the present theory to particular A-spinel system  $\text{CoCo}_2\text{O}_4$  gives  $J_2 = 1.321$  K when considering the experimental values of  $T_N = 34$  K and  $J_1 = -0.169$  K given in Ref. [3].

The variation of the three first correlation functions  $\gamma_i$  ( $i=1, 2, 3$ ) and the correlation length  $\xi$  with the temperature are presented. The sequence of  $[M, N]$

PA to the series have been evaluated for the considered  $\alpha$ . By examining the behaviour of these PA, the convergence was found to be quite rapid and we expect the result to be accurate to within 1% except in the range  $-1 \leq \alpha \leq 0$ . Estimates of the critical exponents associated with susceptibility and correlation length are found to be  $\gamma = 1.382$  and  $\nu = 0.701$ . These values may be compared with those of 3D Heisenberg model [7, 13], namely,  $1.3866 \pm 0.0012$ , and  $0.7054 \pm 0.0011$ . The agreement is excellent. Similar studies for diluted A-spinel lattice are under consideration.

### Appendix:

As the  $n^{(p, q, \dots)}$  are coefficients of  $J_p, J_q, \dots$ , and the latter product is invariant under a change in the order of  $p, q, \dots$ , we can sum up the  $n^{(p, q, \dots)}$  over all combinations of the

$(p, q, \dots)$ 's. We note  $(p, q, \dots) = \sum_{C(p, q, \dots)} n^{(p, q, \dots)}$ .











For instance, we note  $(122) = n^{(1, 2, 2)} + n^{(2, 1, 2)} + n^{(2, 2, 1)}$  and so on. The  $(p, q, \dots)$  are listed in the table 1 with their number of occurrence in the A-spinel lattice for each neighbour up to order  $l=6$ , and which are necessary for the computation of the  $\gamma_i$ 's. The number of occurrence of a diagram in the lattice depends only on the number of sites in the diagram.






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| Order l       | Diagram type<br>( $J_p, J_q, J_r, \dots$ )                                          | First n. n                                                                                                          | Second n. n                              | Third n. n                                 |
|---------------|-------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------|------------------------------------------|--------------------------------------------|
| $\tau_1$      |    | (1)<br>(2)                                                                                                          | 0<br>1                                   | 0<br>0                                     |
| $\tau_2$      |    | (1 1)<br>(1 2)<br>(2 2)                                                                                             | 0<br>6<br>4                              | 0<br>4<br>0                                |
| $\tau_3^2$    |    | (1 1 1)<br>(1 1 2)<br>(1 2 2)<br>(2 2 2)                                                                            | 0<br>16<br>0<br>22                       | 2<br>0<br>42<br>0                          |
| $\tau_4^3$    |    | (1 1 1 1)<br>(1 1 1 2)<br>(1 1 2 2)<br>(1 2 2 2)<br>(2 2 2 2)                                                       | 0<br>24<br>0<br>426<br>0                 | 2<br>0<br>213<br>0<br>140                  |
| $\tau_5^1$    |    | (1 1 1 1 1)<br>(1 1 1 1 2)<br>(1 1 1 2 2)<br>(1 1 2 2 2)<br>(1 2 2 2 2)<br>(2 2 2 2 2)                              | 6<br>0<br>576<br>0<br>3876<br>0          | 0<br>96<br>0<br>2584<br>0<br>970           |
| $\tau_5^6$    |   | (1 1 1 1)<br>(1 1 1 2)<br>(1 1 2 2)<br>(1 2 2 2)<br>(2 2 2 2)                                                       | 0<br>0<br>15<br>0<br>0                   | 0<br>0<br>4<br>0<br>6                      |
| $\tau_6^1$    |  | (1 1 1 1 1 1)<br>(1 1 1 1 1 2)<br>(1 1 1 1 2 2)<br>(1 1 1 2 2 2)<br>(1 1 2 2 2 2)<br>(1 2 2 2 2 2)<br>(2 2 2 2 2 2) | 0<br>216<br>0<br>9966<br>0<br>35766<br>0 | 12<br>0<br>2496<br>0<br>29805<br>0<br>7196 |
| $\tau_6^5$    |  | (1 1 1 1 1 1)<br>(1 1 1 1 1 2)<br>(1 1 1 1 2 2)<br>(1 1 1 2 2 2)<br>(1 1 2 2 2 2)<br>(1 2 2 2 2 2)<br>(2 2 2 2 2 2) | 0<br>0<br>0<br>40<br>0<br>0<br>0         | 0<br>0<br>0<br>0<br>12<br>0<br>8           |
| $\tau_6^{10}$ |  | (1 1 1 1 1)<br>(1 1 1 1 2)<br>(1 1 1 2 2)<br>(1 1 2 2 2)<br>(1 2 2 2 2)<br>(2 2 2 2 2)                              | 0<br>0<br>0<br>234<br>0<br>0             | 0<br>6<br>0<br>78<br>0<br>64               |
| $\tau_6^{13}$ |  | (1 1 1 1 1 1)<br>(1 1 1 1 1 2)<br>(1 1 1 1 2 2)<br>(1 1 1 2 2 2)<br>(1 1 2 2 2 2)<br>(1 2 2 2 2 2)<br>(2 2 2 2 2 2) | 0<br>0<br>0<br>0<br>0<br>30<br>18        | 0<br>0<br>0<br>10<br>0<br>0<br>12          |

| Order l                                                                                              | Diagram type<br>$(J_p, J_q, J_r, \dots)$ |                                                               | First n. n               | Second n. n              | Third n. n               |
|------------------------------------------------------------------------------------------------------|------------------------------------------|---------------------------------------------------------------|--------------------------|--------------------------|--------------------------|
|                                                                                                      | $(J_p^2)$ or $(J_p^2, J_q^2)$            | $(J_r, J_o, \dots)$                                           |                          |                          |                          |
| $\tau_6^4$<br>      | (1)                                      | (1 1 1 1)<br>(1 1 1 2)<br>(1 1 2 2)<br>(1 2 2 2)<br>(2 2 2 2) | 0<br>0<br>0<br>117<br>0  | 0<br>0<br>39<br>0<br>0   | 0<br>0<br>0<br>90<br>0   |
|                                                                                                      | (2)                                      | (1 1 1 1)<br>(1 1 1 2)<br>(1 1 2 2)<br>(1 2 2 2)<br>(2 2 2 2) | 0<br>18<br>0<br>66<br>0  | 0<br>0<br>56<br>0<br>64  | 0<br>26<br>0<br>44<br>0  |
| $\tau_6^{11}$<br>   | (1)                                      | (1 1 1 1)<br>(1 1 1 2)<br>(1 1 2 2)<br>(1 2 2 2)<br>(2 2 2 2) | 0<br>0<br>0<br>54<br>0   | 0<br>0<br>060<br>0<br>0  | 0<br>10<br>0<br>78<br>0  |
|                                                                                                      | (2)                                      | (1 1 1 1)<br>(1 1 1 2)<br>(1 1 2 2)<br>(1 2 2 2)<br>(2 2 2 2) | 0<br>12<br>0<br>132<br>0 | 2<br>0<br>34<br>0<br>64  | 0<br>22<br>0<br>104<br>0 |
| $\tau_6^{12}$<br>   | (1)                                      | (1 1 1 1)<br>(1 1 1 2)<br>(1 1 2 2)<br>(1 2 2 2)<br>(2 2 2 2) | 0<br>0<br>12<br>54<br>0  | 0<br>2<br>18<br>22<br>0  | 0<br>4<br>16<br>30<br>0  |
|                                                                                                      | (2)                                      | (1 1 1 1)<br>(1 1 1 2)<br>(1 1 2 2)<br>(1 2 2 2)<br>(2 2 2 2) | 0<br>0<br>56<br>72<br>0  | 0<br>18<br>32<br>0<br>64 | 4<br>0<br>31<br>96<br>0  |
| $\tau_6^{15}$<br> | (1 1)                                    | (1 1)<br>(1 2)<br>(2 2)                                       | 0<br>0<br>0              | 0<br>0<br>4              | 0<br>2<br>0              |
|                                                                                                      | (1 2)                                    | (1 1)<br>(1 2)<br>(2 2)                                       | 0<br>15<br>0             | 5<br>0<br>0              | 0<br>12<br>0             |
|                                                                                                      | (2 2)                                    | (1 1)<br>(1 2)<br>(2 2)                                       | 0<br>12<br>0             | 0<br>0<br>12             | 0<br>8<br>0              |
| $\tau_6^{14}$<br> | (1)                                      | (1 1 1)<br>(1 1 2)<br>(1 2 2)<br>(2 2 2)                      | 0<br>0<br>0<br>12        | 0<br>5<br>0<br>0         | 0<br>2<br>0<br>8         |
|                                                                                                      | (2)                                      | (1 1 1)<br>(1 1 2)<br>(1 2 2)<br>(2 2 2)                      | 0<br>15<br>0<br>0        | 0<br>4<br>0<br>12        | 0<br>12<br>0<br>0        |

|        |                    |        |                         |        |                            |
|--------|--------------------|--------|-------------------------|--------|----------------------------|
| (m, n) | a(m, n)            | (m, n) | a(m, n)                 | (m, n) | a(m, n)                    |
| (0, 0) | 1                  | (0, 4) | $\frac{112}{2025}$      | (5, 5) | $\frac{2386144}{212625}$   |
| (0, 1) | $\frac{-16}{45}$   | (1, 4) | $\frac{-3728}{2025}$    | (0, 6) | $\frac{9776}{212625}$      |
| (1, 1) | $\frac{16}{15}$    | (2, 4) | $\frac{5632}{675}$      | (1, 6) | $\frac{-734176}{637875}$   |
| (0, 2) | $\frac{16}{45}$    | (3, 4) | $\frac{-42256}{2025}$   | (2, 6) | $\frac{11248}{1215}$       |
| (1, 2) | $\frac{-32}{15}$   | (4, 4) | $\frac{10432}{2025}$    | (3, 6) | $\frac{-5915984}{91125}$   |
| (2, 2) | $\frac{64}{45}$    | (0, 5) | $\frac{-704}{14175}$    | (4, 6) | $\frac{5379056}{42525}$    |
| (0, 3) | $\frac{-16}{75}$   | (1, 5) | $\frac{3232}{3375}$     | (5, 6) | $\frac{-41697584}{212625}$ |
| (1, 3) | $\frac{256}{135}$  | (2, 5) | $\frac{-355376}{30375}$ | (6, 6) | $\frac{3478912}{1275575}$  |
| (2, 3) | $\frac{-928}{135}$ | (3, 5) | $\frac{202112}{6075}$   |        |                            |
| (3, 3) | $\frac{1712}{675}$ | (4, 5) | $\frac{-385984}{6075}$  |        |                            |

**Table.1:** Nonzero coefficients  $a(m, n)$  of the magnetic susceptibility, eq (12) in the text.

|        |                               |        |                                      |        |                                           |
|--------|-------------------------------|--------|--------------------------------------|--------|-------------------------------------------|
| (m, n) | b(m, n)                       | (m, n) | b(m, n)                              | (m, n) | b(m, n)                                   |
| (0, 1) | $\frac{1}{210}$               | (3, 4) | $\frac{-17916953}{243101250}$        | (4, 6) | $\frac{-858172626628}{268019128125}$      |
| (1, 1) | $\frac{-4}{105}$              | (4, 4) | $\frac{-11962192}{121550625}$        | (5, 6) | $\frac{-2081566923263}{2680191281250}$    |
| (0, 2) | $\frac{-44}{3675}$            | (0, 5) | $\frac{-5243482}{12762815625}$       | (6, 6) | $\frac{-667883884798}{1340095640625}$     |
| (1, 2) | $\frac{17}{11025}$            | (1, 5) | $\frac{-45542536}{2552563125}$       | (0, 7) | $\frac{-66065411632}{140710042265625}$    |
| (2, 2) | $\frac{-368}{11025}$          | (2, 5) | $\frac{-665249063}{5105126250}$      | (1, 7) | $\frac{912779579272}{140710042265625}$    |
| (0, 3) | $\frac{-2213}{771750}$        | (3, 5) | $\frac{-2177796064}{2552563125}$     | (2, 7) | $\frac{-21797492555528}{140710042265625}$ |
| (1, 3) | $\frac{-8576}{165375}$        | (4, 5) | $\frac{-89203154}{364651875}$        | (3, 7) | $\frac{2107216697288}{28142008453125}$    |
| (2, 3) | $\frac{-20984}{1157625}$      | (5, 5) | $\frac{-2599272904}{12762815625}$    | (4, 7) | $\frac{-104062365799304}{28142008453125}$ |
| (3, 3) | $\frac{-60316}{1157625}$      | (0, 6) | $\frac{-2088467932}{1340095640625}$  | (5, 7) | $\frac{696863846615696}{140710042265625}$ |
| (0, 4) | $\frac{-18532}{121550625}$    | (1, 6) | $\frac{-28002872983}{2680191281250}$ | (6, 7) | $\frac{-64269320718296}{140710042265625}$ |
| (1, 4) | $\frac{-5619337}{243101250}$  | (2, 6) | $\frac{-8183740084}{38288446875}$    | (7, 7) | $\frac{55333352151008}{46903347421875}$   |
| (2, 4) | $\frac{-26529968}{121550625}$ | (3, 6) | $\frac{-337316720399}{536038256250}$ |        |                                           |

**Table.2:** Nonzero coefficients of the Correlation length, eq (13) in the text.