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# A study of the critical behaviour of a normal ferrimagnetic spinel by high-temperature series expansions

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## Abstract

The critical properties of a ferrimagnetic spinel system ( $AB_2X_4$ , A and B are magnetic ions) are studied by the method of exact high-temperature series expansions. Terms up to seventh order were computed for the magnetic susceptibility  $\chi = \sum_{n=0}^7 a_n (\frac{1}{k_B T})^n$ . The calculations are given for the three nearest neighbours' exchange integrals  $J_{AA}$ ,  $J_{AB}$  and  $J_{BB}$ . The Padé approximants method is used to estimate the critical exponent  $\gamma$  associated with the magnetic susceptibility. A net variation of  $\gamma$  with exchange couplings has been observed. This variation presents some unusual characteristics. The magnetic asymmetric interactions and the competition between the exchange interactions are important for the magnetic phase transition in ferrimagnetic spinels.

We make comparisons with experiment by studying real Heisenberg spinel ferrite systems  $ACr_2S_4$  (A = Fe, Co). The results of  $\gamma$  and  $T_c$  obtained by the present approach are in agreement with the experimental values.

## 1. Introduction

The magnetic phase transitions and critical phenomena of frustrated magnetic systems have been extensively studied during the last decade (for reviews see [1]). Due to the extra degree of freedom arising from the degeneracy of the ground state of such a system, the nature of magnetic phase transitions can be entirely different from that of a non-frustrated compound. Among frustrated materials which display nonconventional magnetic properties are those with the spinel structure [2–9].

The magnetic spinel systems of the form  $AB_2X_4$  (space group  $Fd\bar{3}m$ ) constitute a class of materials which are very suitable to study new types of magnetic behaviours induced by various degrees and types of disorder and frustration. These systems have received considerable attention for their interesting electrical and magnetic properties [10–12].

The case of ferrimagnetic spinel compounds with two magnetic sublattices A and B are particularly interesting because they may exhibit particular disordered magnetic ions in different sites. The ordered structure may be considered to consist of six interpenetrating face-centred cubic lattices, two consisting of A-sites and four of B-sites. The magnetic structures depend on the types of magnetic ions residing in the tetrahedral (A), the octahedral (B) sites and the relative strengths of the inter- ( $J_{AB}$ ) and intra-sublattice interactions ( $J_{AA}$ ,  $J_{BB}$ ). In general, all the three exchange interactions  $J_{AA}$ ,  $J_{BB}$  and  $J_{AB}$  are negative. Furthermore, when all the metal ions (cations) are magnetic, usually the inter-sublattice interaction  $J_{AB}$  is the strongest with  $|J_{AB}| \gg |J_{BB}| > |J_{AA}|$ . Thus,  $J_{AB}$  renders the spinel system as ferrimagnetic with A-site moments aligned antiparallel to the B-site moments keeping the A–A and B–B couplings inherently frustrated [13].

The determination of critical exponents is an important aspect of the theoretical description and experimental characterization of magnetic systems [14]. Competition

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between interactions causes various intriguing phenomena in some magnetic systems. When the spin has a continuous degree of freedom, as in the case of the Heisenberg model, the competition often gives rise to noncollinear spin orderings. Such noncollinear spin orderings are characterized by new types of symmetry which differ from those characterizing the collinear or unfrustrated magnets [15].

High-temperature series expansions (HTSEs) are used to examine the dependence of the critical exponent  $\gamma$  associated with the magnetic susceptibility upon relative strengths of the inter- ( $J_{AB}$ ) and intra-sublattice interactions ( $J_{AA}$ ,  $J_{BB}$ ) in ferrimagnetic spinels.

The HTSE method considered here has been widely developed [16–20] because it is one of the most powerful and rigorous ways to study physical systems. It provides valid estimations of critical temperatures for real magnetic systems. Hoping to obtain more information on the critical properties of Heisenberg ferrimagnetic spinels, we have computed the seventh-order term in a high-temperature series expansion for two sublattices with arbitrary spins. We derived the series of magnetic susceptibilities in powers of  $\beta = \frac{1}{k_B T}$  with nearest neighbour exchange couplings ( $J_{AA}$ ,  $J_{BB}$ ,  $J_{AB}$ ).

The Padé approximants (PA) [21] analysis of the exact HTSEs of the magnetic susceptibility has been used for computing the value of critical exponent  $\gamma$  and the critical temperature  $T_c$ .

We make comparison with experiment by studying real Heisenberg spinel ferrite systems  $ACr_2S_4$  ( $A = Fe, Co$ ) which has recently attracted considerable attention due to their colossal magnetoresistance effect [22]. The obtained results of  $\gamma$  and  $T_c$  by the present approach are in agreement with the experimental ones

## 2. High-temperature series expansions

In order to deduce the expression for the magnetic susceptibility of the ferrimagnetic spinel with two sublattices, the Hamiltonian of the semi-classical Heisenberg spin model is given as:

$$H = -2J_{AA} \sum_{(i,i')} \vec{S}_i \vec{S}_{i'} - 2J_{BB} \sum_{(j,j')} \vec{\sigma}_j \vec{\sigma}_{j'} - 2J_{AB} \sum_{(i,j)} \vec{S}_i \vec{\sigma}_j - \mu_B h_{ex} \left( g_A \sum_i S_i^z - g_B \sum_j \sigma_j^z \right), \quad (1)$$

where  $\vec{S}$  and  $\vec{\sigma}$  are spin vectors of magnitudes  $\vec{S}^2 = S(S+1)$  and  $\vec{\sigma}^2 = \sigma(\sigma+1)$ , in sublattice A and B respectively.  $g_A$  and  $g_B$  are the corresponding gyromagnetic factors and  $\mu_B$  is the Bohr magneton.  $h_{ex}$  is an external magnetic field ( $z$  direction) introduced in order to provide an easy determination of the magnetic susceptibility. The first summation is over all spin pair nearest-neighbours in sublattice A, the second is over all spin pair nearest-neighbours in sublattice B and the third is between all spin pair nearest-neighbours in A and B.  $J_{AA}$ ,  $J_{BB}$  and  $J_{AB}$  are the intra- and inter-sublattice exchange interactions between neighbouring spins.

The magnetization of the ferrimagnetic system is given by:

$$M = \mu_B \left( g_A \sum_i \langle S_i^z \rangle + g_B \sum_j \langle \sigma_j^z \rangle \right) \quad (2)$$

$\langle \dots \rangle$  denotes an equilibrium thermal average. After computing the first derivative of magnetization  $\chi = \left( \frac{\partial M}{\partial h_{ex}} \right)_{h_{ex} \rightarrow 0}$ , we have obtained the general expression of magnetic susceptibility for the collinear normal ferrimagnetic spinel as follows:

$$\chi = \left( \frac{\mu_B^2}{3k_B T} \right) \left( N_A g_A^2 \vec{S}^2 + N_B g_B^2 \vec{\sigma}^2 + g_A^2 \sum_{i \neq i'} \langle \vec{S}_i \vec{S}_{i'} \rangle + g_B^2 \sum_{j \neq j'} \langle \vec{\sigma}_j \vec{\sigma}_{j'} \rangle - 2g_A g_B \sum_{i,j} \langle \vec{S}_i \vec{\sigma}_j \rangle \right), \quad (3)$$

where  $N_A$  and  $N_B$  are the number of magnetic ions in sublattice A and B, respectively.

Finally, a simple form of the magnetic susceptibility is obtained:

$$\chi = \left( \frac{\mu_B^2}{3k_B T} \right) (N_A g_A^2 \vec{S}^2 + N_B g_B^2 \vec{\sigma}^2 + N_A g_A^2 \gamma_{AA} + N_B g_B^2 \gamma_{BB} - 2N_B g_A g_B \gamma_{BA}). \quad (4)$$

Following the procedure given in [12, 25], we compute the expressions of spin correlation functions  $\gamma_{AA}$ ,  $\gamma_{BB}$  and  $\gamma_{AB}$  in terms of powers of  $\beta$  and mixed powers of  $J_1 = 2J_{BB} \vec{\sigma}^2$ ,  $J_2 = 2J_{AB} \vec{S} \vec{\sigma}$  and  $J_3 = 2J_{AA} \vec{S}^2$ .

The correlation functions may be expressed as the following:

$$\begin{aligned} \gamma_{AA} &= \vec{S}^2 \sum_{q=1}^7 \sum_{m=0}^q \sum_{n=0}^{q-m} \sum_{p=0}^{q-m-n} a(m, n, p, q) J_1^m J_2^n J_3^p \beta^q \\ \gamma_{BB} &= \vec{\sigma}^2 \sum_{q=1}^7 \sum_{m=0}^q \sum_{n=0}^{q-m} \sum_{p=0}^{q-m-n} b(m, n, p, q) J_1^m J_2^n J_3^p \beta^q \\ \gamma_{AB} &= \vec{S} \vec{\sigma} \sum_{q=1}^7 \sum_{m=0}^q \sum_{n=0}^{q-m} \sum_{p=0}^{q-m-n} c(m, n, p, q) J_1^m J_2^n J_3^p \beta^q. \end{aligned} \quad (5)$$

Nonzero coefficients  $a(m, n, p, q)$ ,  $b(m, n, p, q)$  and  $c(m, n, p, q)$ , up to order 7 on  $\beta$  are listed in tables 1, 2 and 3 respectively.

## 3. Series analysis and results

The simplest assumption that one can make concerning the nature of the singularity of the magnetic susceptibility  $\chi$  is that in the neighbourhood of the critical point the function exhibits an asymptotic behaviour.

$$\chi(T) \propto (T_c - T)^{-\gamma}. \quad (6)$$

There are two standard methods for the analysis of series expansions, namely:

- (i) the ratio method and variation;
- (ii) the Padé approximants method.

**Table 1.** Nonzero coefficients  $c(m, n, p, q)$  of the correlation function  $\gamma_{BA}$ .

$(m, n, p, q)$	$c(m, n, p, q)$	$(m, n, p, q)$	$c(m, n, p, q)$	$(m, n, p, q)$	$c(m, n, p, q)$
(1, 0, 0, 1)	2	(0, 1, 4, 5)	104/45	(1, 3, 3, 7)	872 398/3645
(0, 1, 1, 2)	8/3	(0, 3, 2, 5)	87 622/2025	(3, 1, 3, 7)	226 472/18 225
(1, 1, 0, 2)	34/9	(2, 1, 2, 5)	80/9	(0, 1, 6, 7)	81 584/42 525
(1, 1, 1, 3)	16/3	(0, 3, 3, 6)	1019 216/18 225	(0, 5, 2, 7)	171 526 118/382 725
(2, 1, 0, 3)	172/27	(2, 3, 1, 6)	221 953/729	(0, 6, 1, 7)	16/567
(0, 1, 2, 3)	8/3	(3, 1, 2, 6)	1792/135	(1, 6, 0, 7)	68/2835
(0, 3, 0, 3)	494/45	(2, 1, 3, 6)	10 112/1215	(5, 2, 0, 7)	8/315
(0, 3, 1, 4)	746/27	(1, 5, 0, 6)	2149 376/6075	(0, 3, 4, 7)	126 088 366/1913 625
(3, 1, 0, 4)	3868/405	(0, 5, 1, 6)	3860 318/18 225	(2, 5, 0, 7)	483 802 024/382 725
(2, 1, 1, 4)	80/9	(3, 3, 0, 6)	4452 341/18 225	(6, 1, 0, 7)	2038 348/76 545
(1, 1, 2, 4)	16/3	(4, 1, 1, 6)	848/45	(3, 3, 1, 7)	35 844 608/54 675
(0, 1, 3, 4)	112/45	(0, 1, 5, 6)	2032/945	(4, 1, 2, 7)	848/45
(1, 3, 0, 4)	1195/27	(5, 1, 0, 6)	808 316/42 525	(2, 1, 4, 7)	9424/1215
(2, 3, 0, 5)	701 276/6075	(1, 3, 2, 6)	3332 276/18 225	(6, 0, 1, 7)	16/2835
(3, 1, 1, 5)	1792/135	(1, 1, 4, 6)	1888/405	(1, 1, 5, 7)	1744/405
(4, 1, 0, 5)	5492/405	(4, 3, 0, 7)	41 712 593/91 125	(0, 7, 0, 7)	192 541 946/637 875
(0, 5, 0, 5)	33 316/567	(5, 1, 1, 7)	482 752/18 225	(0, 2, 5, 7)	8/567
(1, 3, 1, 5)	27 803/243	(2, 3, 2, 7)	135 323 753/273 375		
(1, 1, 3, 5)	2024/405	(1, 5, 1, 7)	2506 351 409/1913 625		

**Table 2.** Nonzero coefficients  $b(m, n, p, q)$  of the correlation function  $\gamma_{BB}$ .

$(m, n, p, q)$	$b(m, n, p, q)$	$(m, n, p, q)$	$b(m, n, p, q)$	$(m, n, p, q)$	$b(m, n, p, q)$
(1, 0, 0, 1)	2	(1, 4, 0, 5)	1416 524/6075	(1, 6, 0, 7)	630 901 223/382 725
(2, 0, 0, 2)	10/3	(0, 2, 3, 5)	4024/405	(0, 2, 5, 7)	218 516/25 515
(0, 2, 0, 2)	22/3	(5, 0, 0, 5)	15 616/1575	(0, 4, 3, 7)	11 784 542/54 675
(3, 0, 0, 3)	224/45	(1, 2, 2, 5)	9856/243	(0, 6, 1, 7)	99 633 071/127 575
(1, 2, 0, 3)	739/27	(6, 0, 0, 6)	592 664/42 525	(4, 2, 1, 7)	148 310 524/382 725
(0, 2, 1, 3)	92/9	(0, 2, 4, 6)	11 216/1215	(2, 2, 3, 7)	1849 297/18 225
(0, 4, 0, 4)	5528/135	(2, 4, 0, 6)	325 121/405	(1, 4, 2, 7)	268 887 851/273 375
(0, 2, 2, 4)	284/27	(1, 2, 3, 6)	47 072/1215	(1, 2, 4, 7)	131 708/3645
(4, 0, 0, 4)	106/15	(1, 4, 1, 6)	11 051 807/18 225	(3, 4, 0, 7)	4108 034 257/1913 625
(1, 2, 1, 4)	1049/27	(2, 2, 2, 6)	127 876/1215	(5, 2, 0, 7)	24 520 799/54 675
(2, 2, 0, 4)	3097/45	(3, 2, 1, 6)	758 651/3645	(7, 0, 0, 7)	12 441 284/637 875
(2, 2, 1, 5)	40 231/405	(0, 6, 0, 6)	1097 084/5103	(3, 2, 2, 7)	489 181/2187
(3, 2, 0, 5)	172 586/1215	(0, 4, 2, 6)	2998 372/18 225	(2, 4, 1, 7)	4669 868/2187
(0, 4, 1, 5)	125 788/1215	(4, 2, 0, 6)	3702 346/14 175		

**Table 3.** Nonzero coefficients  $a(m, n, p, q)$  of the correlation function  $\gamma_{AA}$ .

$(m, n, p, q)$	$a(m, n, p, q)$	$(m, n, p, q)$	$a(m, n, p, q)$	$(m, n, p, q)$	$a(m, n, p, q)$
(0, 0, 1, 1)	4/3	(0, 4, 1, 5)	706 946/6075	(3, 2, 3, 7)	915 829/6075
(0, 2, 0, 2)	53/9	(1, 4, 0, 5)	167 294/1215	(5, 2, 0, 7)	6620 456/382 725
(0, 0, 2, 2)	4/3	(0, 0, 5, 5)	1016/945	(2, 4, 1, 7)	386 566 502/273 375
(1, 2, 0, 3)	116/9	(0, 2, 3, 5)	88/3	(1, 6, 0, 7)	675 001 052/637 875
(0, 2, 1, 3)	404/27	(1, 2, 3, 6)	9434/135	(0, 2, 5, 7)	4803 382/127 575
(0, 0, 3, 3)	56/45	(0, 2, 4, 6)	867 712/25 515	(0, 4, 3, 7)	84 221 632/212 625
(1, 2, 1, 4)	2788/81	(2, 4, 0, 6)	2254 517/6075	(0, 6, 1, 7)	301 410 296/382 725
(2, 2, 0, 4)	1802/81	(0, 0, 0, 6)	41 152/42 525	(4, 2, 1, 7)	1496 656/10 935
(0, 2, 2, 4)	1028/45	(3, 2, 1, 6)	340 796/3645	(2, 2, 3, 7)	50 792/405
(0, 4, 0, 4)	4364/135	(1, 4, 1, 6)	9330 116/18 225	(1, 4, 2, 7)	59 782 103/54 675
(0, 0, 4, 4)	52/45	(2, 2, 2, 6)	7750/81	(1, 2, 4, 7)	6224 368/76 545
(2, 2, 1, 5)	14 620/243	(0, 6, 0, 6)	860 921/5103	(3, 4, 0, 7)	44 023 634/54 675
(3, 2, 0, 5)	41 386/1215	(0, 4, 2, 6)	885 472/3645	(0, 0, 7, 7)	21 856/25 515
(1, 2, 2, 5)	7267/135	(4, 2, 0, 6)	179 194/3645		

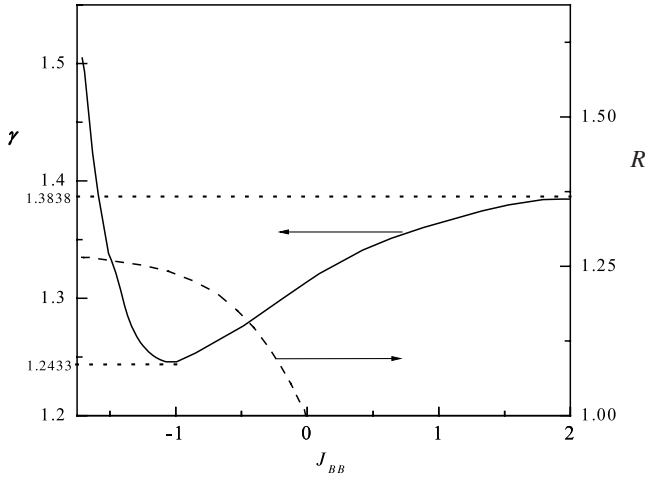
We shall use the second method to obtain estimates for the critical parameters  $T_c$  and  $\gamma$  for the ferrimagnetic spinel. Excellent reviews of these methods are available [24, 25].

The Padé approximants [23] method attempts to represent the magnetic susceptibility as a quotient of two finite polynomials with degrees  $M$  and  $N$ . The singularities of the

function are then estimated by computing the zeros of the denominator polynomial.

The usual approach is to compute the series for the logarithmic derivative of  $\chi(T)$ ,

$$\frac{d}{dT} \log [\chi(T)] \approx \frac{-\gamma}{T - T_c} \quad (7)$$



**Figure 1.** The critical exponent  $\gamma$  (solid line) and  $R$ , representing the ratio of inter-to intraplanar correlations, (dashed line) as a function of the intra-sublattice  $J_{BB}$  for  $J_{AA} = 0$ .

as this function has a simple pole  $T_c$  and should be well represented by Padé approximants  $[M, N]$ . The exponent  $\gamma$  is then re-estimated from the approximants to

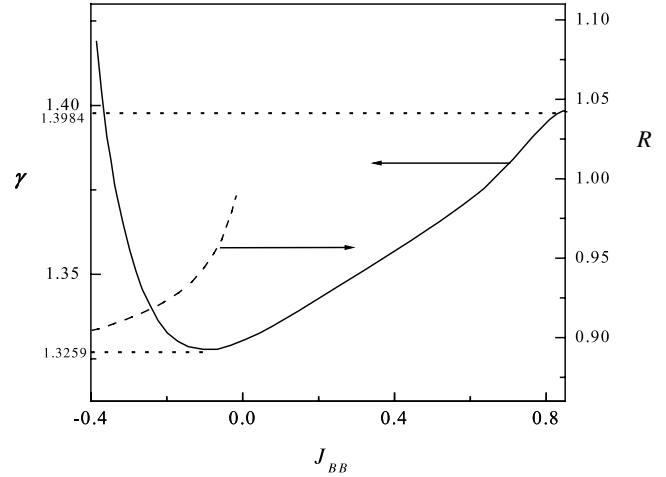
$$(T - T_c) \frac{d}{dT} \log[\chi(T)] \quad (8)$$

evaluated at  $T = T_c$ .

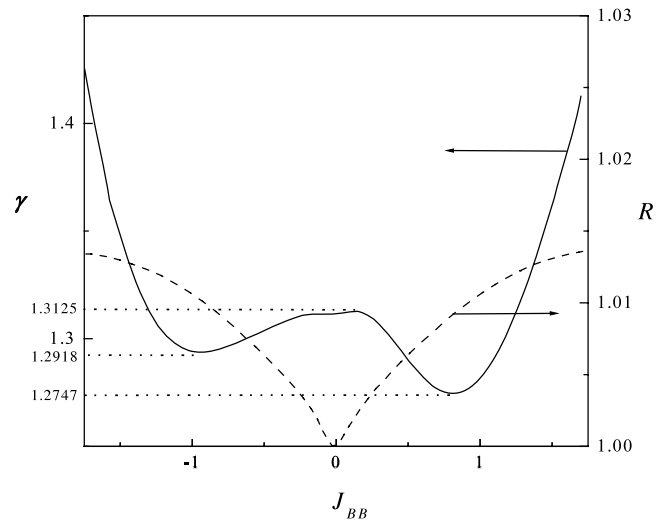
We have calculated the values of the critical exponent  $\gamma$  as a function of intra-sublattices exchanges integrals  $J_{AA}$  and  $J_{BB}$ , and for arbitrary values of spins for the series up to order 7. This procedure was repeated for series up to order 6 and 5. Between the order 6 and 7, the analysis of the series is not affected significantly. The approximants are well converged and estimates are accurate to high precision  $\sim 1\%$ . When the number of terms decreases from 6 to 5, the analysis of the series shows that estimates of  $\gamma$  increase by 10%. For short series  $n \leq 4$  the accuracy in the calculation is not expected to be high.

The gyromagnetic factors  $g_A$  and  $g_B$  were assumed to be equal to 2. For all the cases,  $J_{AB} = -1$  K. It is necessary to point out here that we will not take into account the stability of the spin configuration.

First, we analyse the case where  $J_{AA}$  is weaker (i.e.  $J_{AA} = 0$ ). The behaviour of  $\gamma$  with  $J_{BB}$  is reported in figure 1. From this figure, we can see that the critical exponent (i) decreases rapidly with increasing antiferromagnetic  $J_{BB}$  value until a minimum ( $\gamma = 1.2433$ ), (ii) increases and tends to be constant ( $\gamma = 1.3838$ ) for large ferromagnetic value of  $J_{BB}$ . To examine this variation, we display in the same figure the behaviour of the ratio  $R$  of inter-sublattice correlations to the intra-sublattice correlations. We remark that for large ferromagnetic values of  $J_{BB}$  and weak values of  $R$ , the value of  $\gamma$  is close to that of the 3D Heisenberg model [26, 27]. There is no interaction between the spins in sublattices A and B. The system can be considered as a B-spinel lattice (only the B-sites are occupied by magnetic ions) with consistent critical exponent. When the effect of inter-sublattice correlation is



**Figure 2.** As in figure 1 but with  $J_{AA} = J_{BB}$ .



**Figure 3.** As in figure 1 but with  $J_{AA} = -J_{BB}$ .

more pronounced,  $R$  increases and  $\gamma$  takes the value of Ising-like system [26]. For the large values of  $R$  ( $J_{BB} < -1$  K), the frustration becomes very important and will be responsible for the net divergence of  $\gamma$ .

In figure 2, we present the case where  $J_{AA}$  is equal to  $J_{BB}$ . The dependence of the critical exponent  $\gamma$  on the intra-sublattice exchange coupling is similar to that of figure 1. The minimum of  $\gamma$  is 1.3259. This value is somewhat similar to that of the known XY model [28].

From the plot of the ratio  $R$ , we note that the intra-sublattice correlations are important in the antiferromagnetic exchange integral  $J_{BB}$  region and as consequence  $\gamma$  takes the value of the planar model. For  $J_{BB} < -0.25$ ,  $\gamma$  diverges as a consequence of the strong frustration between different interactions in sublattices A and B.

We have also examined the case where  $J_{AA} = -J_{BB}$ . Figure 3 illustrates the critical exponent versus the intra-sublattice exchange integral  $J_{BB}$ . It can be seen that the curve has two minima ( $\gamma = 1.2747$  and  $1.2938$ ) and diverges for large values of  $J_{BB}$  when the frustration is strong. The major

**Table 4.** The critical temperature and critical exponent  $\gamma$  for the magnetic susceptibility of  $\text{FeCr}_2\text{S}_4$ .

$[M, N]$	[3, 2]	[4, 2]	[3, 3]	[4, 3]	[1, 4]	[2, 4]	[1, 5]	[2, 5]
$T_c$	176.788	176.824	178.556	178.797	176.725	178.587	176.889	178.730
$\gamma$	1.307	1.309	1.321	1.307	1.329	1.326	1.322	1.322

**Table 5.** The critical temperature and critical exponent  $\gamma$  for the magnetic susceptibility of  $\text{CoCr}_2\text{S}_4$ .

$[M, N]$	[3, 2]	[4, 2]	[3, 3]	[4, 3]	[1, 4]	[2, 4]	[1, 5]	[2, 5]
$T_c$	239.393	241.349	240.303	239.725	241.421	240.267	240.447	239.429
$\gamma$	1.300	1.312	1.326	1.300	1.330	1.327	1.326	1.330

frustrations among spins arise from the competition between the ferromagnetic and antiferromagnetic interactions within and between spins in sublattices A and B. The ratio  $R$  governs the behaviour of  $\gamma$  versus exchange interactions.

Finally, we apply this model to magnetic spinel semiconductors  $\text{FeCr}_2\text{S}_4$  and  $\text{CoCr}_2\text{S}_4$ . Both systems are normal spinel ferrites with collinear configuration. The physical parameters are taken from reference [29] and are: for  $\text{FeCr}_2\text{S}_4$ , the exchange couplings are  $J_{\text{Fe-Cr}} = -10$  K and  $J_{\text{Cr-Cr}} = -0.95$  K, the gyromagnetic factors are  $g_{\text{Cr}} = 1.98$  and  $g_{\text{Fe}} = 2.1$ . The system presents ferrimagnetic order below the critical temperature  $T_c = 177$  K. For  $\text{CoCr}_2\text{S}_4$ ,  $J_{\text{Co-Cr}} = -17.5$  K,  $J_{\text{Cr-Cr}} = -2.3$  K and  $g_{\text{Co}} = 2.3$ . The system presents ferrimagnetic order below the critical temperature  $T_c = 240$  K. In the two systems the interaction between Fe-Fe and Co-Co is negligible.

The sequences of  $[M, N]$  Padé approximants to the series have been evaluated and are presented in tables 4 and 5. The estimated critical temperatures are in good agreement with the experimental values found in  $\text{FeCr}_2\text{S}_4$  and  $\text{CoCr}_2\text{S}_4$ .

#### 4. Conclusion

In this paper, high-temperature series expansion (HTSE) of the spin correlation functions of a normal ferrimagnetic spinel lattice is computed to order 7 in  $\beta = \frac{1}{k_B T}$  for the Heisenberg model. For the sake of convenience, we have taken only the first inter- ( $J_{\text{AB}}$ ) and intra-sublattice interactions ( $J_{\text{AA}}, J_{\text{BB}}$ ). HTSEs extrapolated with the Padé approximants (PA) method are shown to be convenient to provide a 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 ferrimagnetic spinel lattice. A net variation of critical exponent  $\gamma$ , associated with magnetic susceptibility, with exchange coupling has been observed. This variation presents some unusual characteristics. We cannot claim to understand what causes  $\gamma$  to behave in this fashion. Nevertheless, an instructive phenomenological picture in view of the magnetic symmetry of the interactions in the system may be given. The magnetic asymmetric interactions and the competition between the exchange couplings are important for the magnetic phase transition in a ferrimagnetic spinel. In particular, we have three distinct regions: for the symmetric region (weak values of  $R$ ),  $\gamma$  tends to the value predicted by the Heisenberg

model. In the asymmetric region,  $\gamma$  approaches the values predicted by the Ising model. For the highly frustrated system,  $\gamma$  is large. It was argued that the critical properties of a variety of frustrated magnets are often different from those of conventional unfrustrated magnets [28].

The application of the present theory to particular chalcogenide spinels  $\text{FeCr}_2\text{S}_4$  and  $\text{CoCr}_2\text{S}_4$  gives the estimates values of critical temperature  $T_c$  and critical exponent  $\gamma$ .

The sequences of  $[M, N]$  PA to the series have been evaluated. 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%. Estimates of the critical exponents associated with magnetic susceptibility are found to be  $\gamma = 1.317$  for  $\text{FeCr}_2\text{S}_4$  and  $\gamma = 1.328$  for  $\text{CoCr}_2\text{S}_4$ . The central values of the obtained critical temperatures are  $T_c = 177.74$  K for  $\text{FeCr}_2\text{S}_4$  and  $T_c = 240.42$  K for  $\text{CoCr}_2\text{S}_4$ . These values are in good agreement with the experimental ones given in [29].

Finally, the study of the critical properties of systems with ferrimagnetic spinel structure in the framework of HTSEs that takes into account magnetic correlations is very significant and may bring an important correction in a relatively simple way.

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#### References

- [1] Diep H T (ed) 1994 *Magnetic Systems with Competing Interactions (Frustrated Spin Systems)* (Singapore: World Scientific)
- [2] Freeman S and Wojtowicz P J 1969 *Phys. Rev.* **177** 882
- [3] Wojtowicz P J 1967 *Phys. Rev.* **155** 492
- [4] Brown J R, Jenkins R C L, Price S, Proykova Y O, Salt D W, Tinsley C J and Hunter G J A 1992 *J. Magn. Magn. Mater.* **104–107** 207
- [5] Hunter G J A, Jenkins R C L and Tinsley C J 1990 *J. Phys. A: Math. Gen.* **23** 4547
- [6] Hunter G J A, Evans C W, Jenkins R C L, Tinsley C J and Wynn E W 1995 *J. Magn. Magn. Mater.* **140–144** 1513
- [7] Cherriet Y, Hamedoun M and Chatwiti A 2002 *J. Magn. Magn. Mater.* **247** 242
- [8] Haug M, Fahnle M, Kronmüller H and Haberey F 1987 *Phys. Status Solidi b* **144** 411
- [9] Dormann J L and Nogues M 1990 *J. Phys.: Condens. Matter* **2** 1223



- [10] Somasundaram P, Honig J M, Pekarek T M and Crooker P C 1996 *J. Appl. Phys.* **79** 5401
- [11] Pouget S and Alba M 1995 *J. Phys.: Condens. Matter* **7** 4739
- [12] Hamedoun M, Housa M, Benzakour N and Hourmatallah A 1998 *J. Phys.: Condens. Matter* **10** 3611
- [13] Singh D H, Gupta M and Gupta R 2002 *Phys. Rev. B* **65** 064432
- [14] Godenfeld N 1992 *Lectures in Phase Transitions and the Renormalisation Group* (Reading, MA: Addison-Wesley)
- [15] Kawamura H 1988 *J. Appl. Phys.* **63** 3086
- [16] Hamedoun M, Bakrim H, Filali A, Hourmatallah A, Benzakour N and Sagredo V 2004 *J. Alloys Compounds* **369** 70
- [17] Hamedoun M, Bakrim H, Hourmatallah A and Benzakour N 2003 *Surf. Sci.* **539** 159
- [18] Hamedoun M, Bakrim H, Hourmatallah A and Benzakour N 2003 *Superlatt. Microstruct.* **33** 131
- [19] Bakrim H, Bouslykhane K, Hamedoun M, Hourmatallah A and Benzakour N 2005 *J. Magn. Magn. Mater.* **285** 327
- [20] Moron M C 1996 *J. Phys.: Condens. Matter* **8** 11141
- [21] Baker G A and Graves-Morris P 1996 *Padé Approximants* (Cambridge: Cambridge University Press)
- [22] Ramirez A P, Cava R J and Krajewski J 1997 *Nature* **386** 156
- [23] Stanly H E 1967 *Phys. Rev.* **158** 537
- [24] Hunter D L and Baker G A 1973 *Phys. Rev. B* **7** 3346
- [25] Gaunt D S and Guttman A J 1974 *Phase Transitions and Critical Phenomena* vol 3 ed C Domb and M S Green (New York: Academic) pp 181–243
- [26] Guillou J C LE and Zinn-Justin J 1977 *Phys. Rev. Lett.* **2** 95
- [27] For a review, see: Collins M F 1989 *Magnetic Critical Scattering* (Oxford: Oxford University Press)
- [28] Afif K, Benyoussef A and Hamedoun M 2002 *Chin. Phys. Lett.* **19** 1187 and references therein
- [29] Gibart P, Dormann J L and Pellerin Y 1969 *Phys. Status Solidi* **36** 187