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Critical phenomena in ferromagnetic superlattices

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Abstract. Within the framework of the high-temperature series expansions technique, we examine the phase transition and the critical phenomena of a two-component superlattice with simple cubic structure, through three models: Ising, XY and Heisenberg. The reduced critical temperature of the system is studied as a function of the thickness of the constituents and the exchange interactions in each material, and within the interface. We show the existence of a critical thickness of the unit cell at which the reduced critical temperature of the binary superlattice remains insensitive to the exchange coupling within the interfaces. The values of the effective critical exponent γ_{eff} associated with the magnetic susceptibility agreed with the universal classes in the limit cases where the superlattice is still comparable to an infinite simple cubic lattice. We attribute the breakdown in the universality hypothesis to the crossover effects.

PACS. 75.70.Cn Magnetic properties of interfaces – 75.70.-i Magnetic properties of thin films, surfaces, and interfaces

1 Introduction

In recent years, there has been major growth in the body of experimental and theoretical knowledge regarding the origin and behavior of the phase transition in magnetic layered structures and superlattices. Layered composite materials may be distinctly different from those of their bulk counterparts [1,2]. In particular, research has been focused on systems such as magnetic superlattices [3,4] and multilayers [5,6].

Magnetic superlattices are defined as periodic layered structures with alternating layers having different magnetic properties. Phase transitions in the superlattices and multilayers have their own behaviours, which are different from those in the bulk materials. They have been investigated by use of various theoretical methods. Fishman et al. [7] investigated a periodic multilayer consisting of two ferromagnetic materials in a theory based on general Ginzburg-Landau formulation. They obtained the transition temperature and spin wave spectra. The Landau formalism of Camley and Tilley [8] has been applied to calculate the critical temperature in this system [9]. For more complicated superlattices with an arbitrary number of different layers in an elementary unit, Barnas [10] has derived some general dispersion equations for the bulk and surface polaritons. These equations are then applied to magnetostatic modes and retarded wave propagation in the Voigt geometry [11]. Recently, Sy and Ow [12], using the mean field approximation, and Seidov and Shaulov [13], using the effective field theory with the differential operator technique, studied the phase transition in an alternating magnetic superlattice. Saber et al. [14] using the effective field theory with a probability distribution technique that accounts for the self-spin correlation functions, studied the critical properties in a magnetic superlattice consisting of two ferromagnetic materials with different bulk transition temperatures. The critical temperatures were obtained as functions of the site dilution and thickness of the superlattice with various exchange interactions in the same material and across the interface.

In this contribution we present a theoretical study of the critical properties of an infinite, alternating superlattice using the high-temperature series expansions (HTSE) extrapolated with the Padé Approximants method [15]. We have considered three kinds of models, namely Ising, XY and Heisenberg types. The series expansions for the magnetic susceptibility have been derived to order six in the reciprocal temperature, including nearest neighbour exchange couplings in the two superlattice constituents and across the interface (see Fig. 1). Our intention is to study the effects of different exchange couplings on the critical temperature of the superlattice and on the effective critical exponent associated with the magnetic susceptibility. The semiclassical method of HTSE given by Stanley and Kaplan [16] has the following advantages: firstly, one can exactly deal with any symmetry of the magnetic structure; secondly, we can easily treat the Ising, XY and Heisenberg models in a unified way; thirdly, this method is more fundamental than usual molecular field approaches as it takes into consideration spin-spin correlation. The

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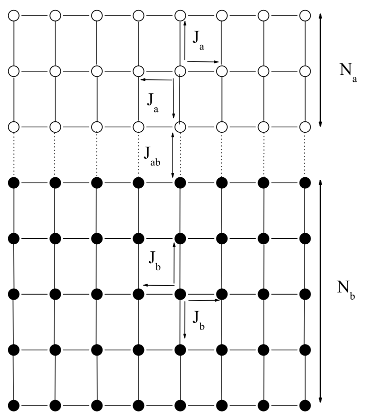

Fig. 1. Two-dimensional cross section of a unit cell of an infinite superlattice composed of two ferromagnetic materials A and B, where $N = N_a + N_b$ is the thickness of the cell.

method considered here has been widely developed [17,18] because it is one of the most powerful and rigorous ways to study physical systems. It provides valid estimations of the critical temperatures for real magnetic systems [19–21].

2 Formalism

We consider an infinite simple cubic superlattice in which N_a layers of material A alternate with N_b layers of material B, which is the $ABAB \cdots$ structure. The periodic condition suggests that we only consider one unit cell which interacts with its nearest-neighbours via the interface coupling. A schematic diagram of the unit cell under consideration is given in Figure 1. The coupling strength between nearest-neighbouring spins in A (B) is denoted by $J_a (J_b)$ while J_{ab} stands for the exchange coupling between nearest-neighbouring spins across the interface. Starting with the zero-field Heisenberg Hamiltonian:

$$H = -2 \sum_{ij} J_{ij}\, \mathbf{S}_i \cdot \mathbf{S}_j. \tag{1}$$

The summation runs over all the pairs of nearest-neighbour pair interactions in the unit cell. The sign of the J_{ij} is assumed to be positive (ferromagnetic). $\mathbf{S}_i$ is the spin operator at site i, of length $\bar{S}^2 = S(S+1)$ for the two materials. The statistics of our spin system are studied using the HTSE method whose starting point is the expansion of the correlation function $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle = \mathrm{Tr}\, \mathbf{S}_i \cdot \mathbf{S}_j e^{-\beta H} / \mathrm{Tr} e^{-\beta H}$ between spins at sites i and j in powers of β [16]:

$$\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle = \sum_{l=0}^{\infty} \frac{(-1)^l}{l!} \alpha_l \beta^l, \tag{2}$$

where $\beta = 1/k_B\, T$ with k_B being the Boltzmann constant. The calculation of the coefficients α_l leads to a diagrammatic representation [16], which involves two separate phases:

- The finding and cataloging of all diagrams or graphs which can be constructed from one dashed line connecting sites i and j, and l straight lines, and the determination of diagrams whose contribution is nonvanishing.
- Counting the number of times that each diagram can occur in the magnetic system.

In our case, we have to deal with nearest-neighbour coupling J_{ij}. The coefficient α_l may be expressed for each topological graph as [16]:

$$\alpha_l = \bar{S}^2 \left(-2\bar{S}^2\right)^l \left(J_{i\,k_1}^{m_1} J_{k_2\,k_3}^{m_2} \cdots J_{k_w\,j}^{m_v}\right) [\alpha_l], \tag{3}$$

with the condition $\sum_{r=1}^{v} m_r = l$ for $m_r = 0, 1, \cdots, l$. The weight $[\alpha_l]$ of each graph is tabulated and given in reference [16] and k_1, $k_2, \cdots$, k_w represent the sites surrounding the sites i and j. The relationship between the magnetic susceptibility per spins $\chi(T)$ and the correlation function may be expressed as follows:

$$\chi(T) = \frac{\beta}{N\overline{N}} \sum_{ij} \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle. \tag{4}$$

N is the number of the layers in the unit cell and $\overline{N}$ is the number of magnetic ions in each layer. The HTSE technique is developed for the magnetic susceptibility $\chi(T)$ with arbitrary exchange couplings J_a, J_b and J_{ab} up to order 6 in β. We obtain the following function:

$$\chi(T) = g\mu_B^2 \beta \bar{S}^2 \sum_{n=0}^{6} \sum_{p=0}^{n} \sum_{q=0}^{n} a_\nu(p,q,n) \left(\frac{J_b}{J_a}\right)^p \left(\frac{J_{ab}}{J_a}\right)^q \tau^{-n}, \tag{5}$$

with $\tau = k_B\, T/2\bar{S}^2 J_a$ and the condition $p + q \leq n$. g is the gyromagnetic ratio and μ_B is the Bohr magneton. For the Ising and XY models, we considered the new values of $[\alpha_l]$ by using the transformation depending only on the dimension ν of the spin (i.e.: $\nu = 1$ for the Ising type, $\nu = 2$ for XY type and $\nu = 3$ for the Heisenberg type), given in Table 1 of reference [22]. The coefficients $a_\nu(p,q,n)$ are computed for some unit cell thicknesses ($N_a = 3, \cdots, 32; N_b = 5$) and are available on request. We use the well-known Padé Approximants method [15] to estimate the critical parameters $\tau_c(\nu) = k_B T_c / 2\, \bar{S}^2 J_a$ and the effective critical exponent $\gamma_{\mathit{eff}}(\nu)$ associated with the magnetic susceptibility $\chi(T)$.

3 Results and discussions

The calculations were carried out in order to investigate the effects of the superlattice thickness and different exchange couplings on the reduced critical temperature $\tau_c(\nu) = k_B T_c / 2\, \bar{S}^2 J_a$ and on the effective critical exponent $\gamma_{\mathit{eff}}(\nu)$ associated with the magnetic susceptibility χ. Throughout this paper, we take J_a as the unit of energy ($J_a = 1$) and N_b will be equal to 5.

We first consider the symmetric case with $J_b = J_a$. Figure 2a shows the variation of $\tau_c(\nu)$ with J_{ab} for three

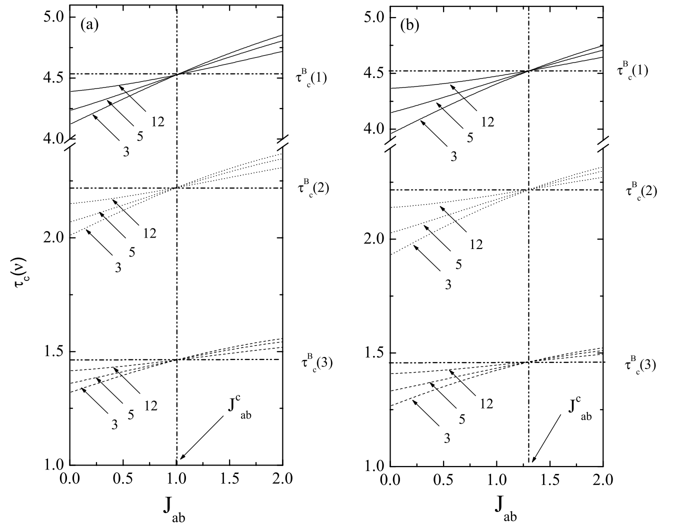

Fig. 2. The reduced critical temperature $\tau_c(\nu)$ versus the parameter J_{ab} for case where $N_a = 5$ and for the three models: Heisenberg (dashed lines), XY (doted lines) and Ising (solid lines). The number accompanying each curve denotes the value of thickness N_a of the constituent A. The doted-dashed horizontal lines correspond to the bulk critical temperature of the material: (a) $J_b = 1.0$; (b) $J_b = 0.95$.

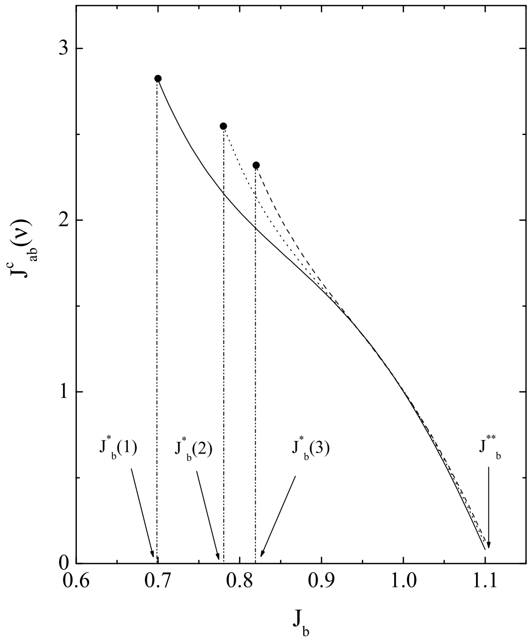

Fig. 3. Phase diagram in terms of the coupling J^c_{ab} versus J_b, for the three models: Heisenberg (dashed lines), XY (doted lines) and Ising (solid lines).

values of the thickness ($N_a = 3,\ 5,\ 12$). We see that all the curves intersect at the same abscissa, $J^c_{ab} = 1$, and ordinate points $\tau^B_c(\nu)$, with $\tau^B_c(1) = 4.526$, $\tau^B_c(2) = 2.2202$ and $\tau^B_c(3) = 1.4651$. At these points, the superlattice becomes similar to a three-dimensional infinite bulk system, where the ordering temperature remains insensitive to the thickness of the unit cell. As we enlarge the value of J_{ab} the superlattice ordering temperature is raised, and when J_{ab} is greater than J^c_{ab}, $\tau_c(\nu)$ becomes higher than that of the bulk one, $\tau^B_c(\nu)$. Another characteristic of the curves, as indicated in Figure 2a, is the increasing reduced Curie temperature when the thickness of the superlattice increases for $J_{ab} < J^c_{ab}$. The opposite tendency is seen for $J_{ab} > J^c_{ab}$, which may be explained by the appearance of magnetic order in the interfaces above J^c_{ab} [20]. In Figure 2b, we display the case when J_b is equal to 0.95; this figure is qualitatively similar to Figure 2a. We remark that the strength of J_{ab}, acting on the interfaces, causes the critical value J^c_{ab} to move to a higher value ($J^c_{ab} = 1.29$) in the same way for the three models. According to this result, it should be necessary to compute the phase diagram in the $(J^c_{ab}(\nu),\ J_b)$ plane.

In Figure 3 we investigate this phase diagram by plotting the dependence of $J^c_{ab}(\nu)$ on J_b; for Ising, XY and Heisenberg models. It can be seen that $J^c_{ab}(\nu)$ increases when J_b decreases and vanishes below the critical value $J^*_b(\nu)$ and above the critical value J^{**}_b: $J^*_b(1) = 0.7$, $J^*_b(2) = 0.78$, $J^*_b(3) = 0.82$ and $J^{**}_b(\nu = 1,\ 2,\ 3) = 1.11$. When the magnetic order of slab A (B) dominates i.e. $J_b > J^{**}_b$ ($J_b < J^*_b$), the superlattice becomes equivalent to a succession of relatively isolated ferromagnetic films and will not be comparable to an infinite bulk structure. Then the reduced critical temperature of the system depends on the thickness of the unit cell, and on the strength of the exchange couplings in the bulk (slabs A and B). We note that the three curves coincide in the range $0.95 \leq J_b \leq 1.10$ and take distinct values for $J_b < 0.95$ and $J_b > 1.05$. In the latter ranges and for a fixed value of J_b, $J^c_{ab}(\nu)$ increases with the dimensionality of the spin. This is ascribed to the degree of freedom of the spin. A comparable situation has been obtained by Saber et al. [24] when they studied the effect of a transverse field on the 1/2-Ising superlattice. They found that the transverse field increases the critical values $J^c_{ab}(1)$ for lower values of the parameter J_b (see Fig. 6 of Ref. [24]).

To study the effect of the interface interaction parameter on the reduced critical temperature of the superlattice, $\tau_c(\nu)$ is calculated as a function of the thickness N_a of slab A and for four values of J_{ab} (Figs. 4a to d). Figure 4a corresponds to the symmetric ($J_b = J_a$). For $J_{ab} < 1.0$, $\tau_c(\nu)$ increases with N_a and approaches asymptotically $\tau^B_c(\nu)$ as the number of layers becomes larger. For $J_{ab} > 1.0$, $\tau_c(\nu)$ decreases as N_a increases and tends to a constant limit. One may explain this feature as follows: for $J_{ab} > 1.0$, the system may be ordered in the interfaces before it is in the two slabs, i.e. the interface magnetic order dominates and when the number of layers is very large, the system can be considered practically as a two-constituent superlattice with a temperature depending on magnetic coupling at the interface J_{ab}. For $J_{ab} < 1.0$, the system may be ordered in the two materials A and B before it orders in the interfaces and when the number of layers increases, the ordering temperature of the system tends to the bulk one. For the symmetric case, the magnetic order of the two constituents A and B compete with the magnetic order of the interface. In

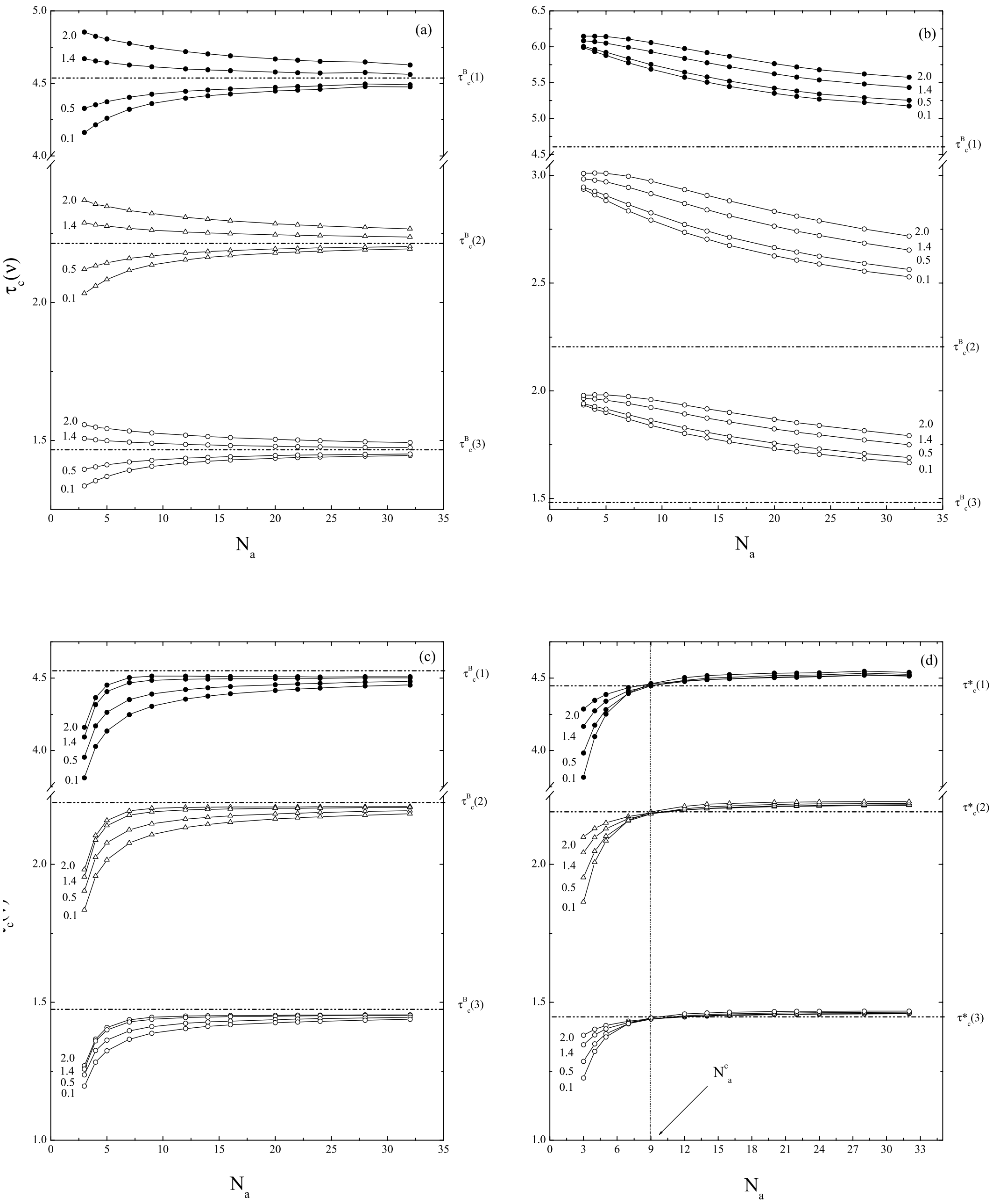

Fig. 4. The variation of $\tau_c(\nu)$ as a function of thickness N_a, of slab A (N_b is fixed to 5) for given values of J_b and for the Heisenberg (open circles), XY (open triangles) and Ising (solid circles). The number accompanying each curve denotes the value of J_{ab}: (a) $J_b = 1$; (b) $J_b = 1.4$; (c) $J_b = 0.1$; (d) $J_b = 0.696$.

Figures 4b to d, we investigate the asymmetric case ($J_b \neq J_a$). Several characteristics appear when J_b takes some values outside the range $[J_b^*(\nu), J_b^{**}]$, where J_{ab}^c vanishes (see Fig. 3). In Figure 4b, we display the variation of $\tau_c(\nu)$ with the thickness N_a, for $J_b = 1.4$. This figure shows that the value of the reduced critical temperature $\tau_c(\nu)$ decreases with N_a and remains higher than the temperature of the bulk, for all values of J_{ab}. The opposite tendency is shown in Figure 4c when J_b is weak ($J_b = 0.1$); the reduced critical temperature $\tau_c(\nu)$ increases with N_a but remains less than the temperature of the bulk for all values of J_{ab}. These behaviours may be understood as follows: for $J_b \notin [J_b^*(\nu), J_b^{**}]$ the system may be assimilated to a succession of isolated thin films where the slab A constitutes the bulk of the thin film (because of the variation of N_a) bordered by two pseudo-surfaces formed from the slab B. When the magnetic order of the pseudo-surfaces dominates ($J_b = 1.4$), the reduced critical temperature of the superlattice will be higher than the bulk one. The opposite situation is obtained if the magnetic order of the bulk (slab A) is greater ($J_b = 0.1$), independent of the strength of the interface exchange coupling J_{ab}. The variation of the $\tau_c(\nu)$ is then governed by competition of the magnetic order of the constituent A and the constituent

Table 1. The adjusted values of J_b and the corresponding critical temperatures τ_c^* (ν = 1,2,3) for each critical thickness N_a^c.

N_a^c	3	4	5	7	9	12	14	16	20	22	24	28
J_b	0.421	0.512	0.579	0.648	0.696	0.750	0.771	0.778	0.801	0.811	0.813	0.817
$\tau_c^*(1)$	4.3039	4.3915	4.4092	4.4375	4.4435	-	-	-	-	-	-	-
$\tau_c^*(2)$	2.1062	2.1554	2.1652	2.1793	2.1819	2.1819	2.1845	2.1896	-	-	-	-
$\tau_c^*(3)$	1.3843	1.4202	1.4273	1.4367	1.438	1.4383	1.4395	1.4429	1.4452	1.446	1.4476	1.45

B. In the intermediate case i.e. $J_b = 0.696$, Figure 4d shows that for a fixed value of J_{ab} and for $J_b < 1$, $\tau_c(\nu)$ increases with N_a gradually to reach a constant for large values of the thickness. In this case, an interesting result appears: all the curves corresponding to the different values of J_{ab}, coincide in a critical length $N_a^c = 9$ in which the reduced temperature $\tau_c^*(\nu)$ of the superlattice does not depend on the interface exchange parameter J_{ab}. N_a^c is the same for the three models. The system presents a bulk behaviour with a reduced critical temperature less than the bulk one: $\tau_c^*(\nu) < \tau_c^B(\nu)$. The magnetic orders of the constituents A and B compete between each other and lead to a critical value of N_a^c at which this competition may be cancelled. The value of N_a^c remains sensitive to the choice of the value of J_b because the exchange coupling of each slab acts indirectly onto the correlation function between two sites of the two slabs (Eqs. (2) and (3)). In Table 1, we give the values of N_a^c and the corresponding critical temperatures $\tau_c^*(\nu)$ for each adjusted value of J_b.

On the other hand, according to the universality hypothesis, critical phenomena can be described by quantities that do not depend on the microscopic details of a system, but only on global properties such as the dimensionality and the symmetry of the order parameter. It has been a point of interest to see the influence of exchange couplings on the behaviour of the effective critical exponent $\gamma_{eff}(\nu)$, associated with the magnetic susceptibility of the superlattice. The $\gamma_{eff}(\nu)$ dependence of the value of J_{ab}, for the three models and for some lengths of the unit cell, is depicted in Figure 5.

For the symmetric case, $\gamma_{eff}(\nu)$ vary continuously with the enhancement of the exchange coupling within the interface and remain independent of J_{ab} in the neighbourhood of $J_{ab} = 1$; namely in the range $0.868 \leq J_{ab} \leq 1.101$. The corresponding values of the effective critical exponent were: $\gamma_{eff}(1) = 1.2456$, $\gamma_{eff}(2) = 1.2916$ and $\gamma_{eff}(3) = 1.3819$ which may be compared with the values of the universality classes. However, the deviation from the universal value ($\gamma = 4/3$) for the Heisenberg case is not negligible. It is worth noting that the present value almost coincides with that obtained by Stanley [23] and Chakraborty [25] for the simple cubic lattice. For $J_{ab} > 1.101$ and $J_{ab} < 0.868$, the estimate of $\gamma_{eff}(\nu)$, regarded as a function of J_{ab} and the unit cell thickness, shows rapid variation. To understand what causes $\gamma_{eff}(\nu)$ to behave in this fashion, an instructive phenomenological picture in view of the symmetry of the system may be given. For $0.868 \leq J_{ab} \leq 1.101$, the system is similar to a simple cubic lattice with consistent critical exponents. The parameter J_{ab} introduces some magnetic asymmetry into the system and thus causes a noticeable inhomogeneity on the distribution of the exchange couplings because those variation effects were not equivalent in the three crystallographic directions. The latest characteristic (asymmetry) may be viewed as a defect of the system. In other words, the variation of $\gamma_{eff}(\nu)$ is essentially due to the breakdown of the cyclic condition (the translation invariance) along the normal direction. A change in the exponent is expected when the effective dimensionality of the superlattice is altered by the *lattice anisotropy*. The effective critical exponent describes the singular behavior of the phase transition when the system *crosses over* from one universality class (three dimension) to another, i.e. as the system approaches the thin film-like limit (succession of isolated thin films) for which the real dimensionality is not well established.

4 Conclusion

The high-temperature series expansions method is applied to study the magnetic properties of two-component superlattice having simple cubic structure, through Ising, XY and Heisenberg models. We have discussed the effects of the interface exchange coupling J_{ab} the bulk exchange couplings J_a and J_b, and the thickness of the unit cell on the magnetic properties of the superlattice.

A number of characteristic behaviours have been reported. The main result is that there exists a critical J_{ab}^c of J_{ab}, below which the reduced critical temperature of the system $\tau_c(\nu) = k_B T_c / 2\bar{S}^2 J_a$ is less than the bulk one, and then the system may ordered in the bulk form before in the interfaces. The opposite tendency is seen for $J_{ab} > J_{ab}^c$ and the magnetic order of the interfaces becomes prominent. The values of J_{ab}^c for the three models (Ising, XY, Heisenberg) coincide in the range of J_b, where the system is still comparable to the infinite simple cubic lattice. For higher ($J_b = 1.4$) and weaker ($J_b = 0.1$) values of J_b, the system is similar to a succession of isolated films and then the critical parameter J_{ab}^c vanishes. In this case and when we investigated the effect of the thickness of the unit cell, a competition of the magnetic order between the constituent A and the constituent B appears and leads to a $\tau_c(\nu)$ behaviour which may be compared to that of thin film. The balance of the magnetic order gives a special characteristic: for $J_b < J_a$ and $N_b > N_a$, there exists a critical length

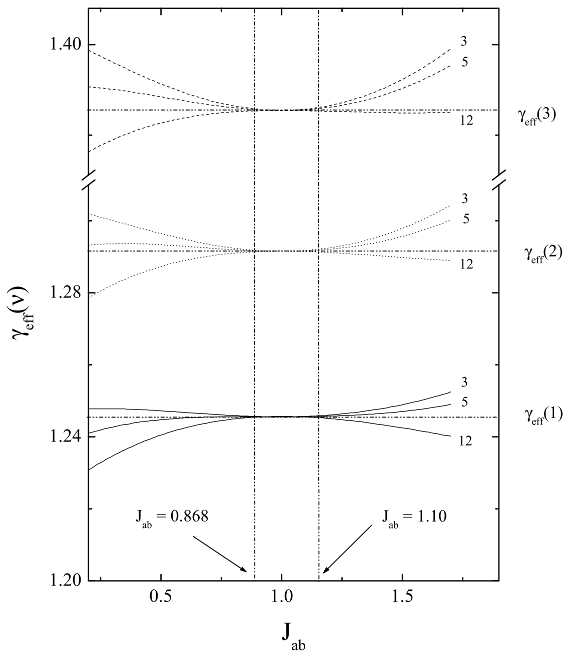

Fig. 5. The variation of the effective critical exponent $\gamma_{eff}\ (\nu)$ as a function of J_{ab}, for $J_b = 1$ and for the models: Heisenberg (dashed lines), XY (dotted lines) and Ising (solid lines). The number accompanying each curve denotes the value of thickness N_a of the constituent A.

$N^c = N_b + N_a^c$ of the unit cell at which $\tau_c\ (\nu)$ remains insensitive to the exchange coupling, J_{ab}, within the interfaces. We have also made a study of the dependence of the behaviours of the effective critical exponent $\gamma_{eff}\ (\nu)$, associated with the magnetic susceptibility χ, with J_{ab} for the symmetric case ($J_b = J_a$). Some discussions are given concerning the variation of $\gamma_{eff}\ (\nu)$ with J_{ab}, in view of the symmetry of the superlattice. The values of the effective critical exponent $\gamma_{eff}\ (\nu)$ corresponding to the three models (Ising, XY, Heisenberg) are close to the universal classes, in the limit cases when the system is still comparable to an infinite simple cubic lattice. The crossover effects from three-dimensional superlattice to a succession of isolated thin films, are the origin of the variation of the effective critical exponent.

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