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A pseudo-two-dimensional Ising model of ferroelectrics

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Abstract. In this paper we consider an Ising bilayer as a chain of simple ladders, characterized by intra-ladder and inter-ladder couplings, to study in detail the para- and ferroelectric behaviours (also considering the electrostrictive corrections in the intra-ladder direction of the plane) for different ratios of interaction constants and different values of electric field, by using a transfer matrix method for the intra-ladder interactions and the MF approximation for the inter-ladder interactions. In the particular case in which the interaction between two layers (planes) is taken to be zero, our results reproduce entirely those obtained previously for a pseudo-one-dimensional Ising model, which explains the experimental data well for the thermodynamic properties of 1D ferroelectrics such as CsD_2PO_4 (d-CDP).

1. Introduction

In [1, 2] we considered Ising spin ladders to study in detail the behaviour of different thermodynamic properties related to different values of temperature, the ratio of interaction constants and the magnetic field. These systems, like the other intermediate systems, can help to provide further insight into the physics of 1D spin chains and 2D high-T_c spin systems, both of which have shown interesting and unusual magnetic and superconducting properties (see [3–17] and references therein). Thus, the orthorhombic compound vanadyl pyrophosphate $(VO)_2P_2O_7$ clearly shows a ladder configuration of spin-$\frac{1}{2}$ V^{4+} ions in its crystal lattice. Also, some years ago, many theories based upon the pseudo-one-dimensional (1D) Ising model were applied to explain the thermodynamic properties of the 1D ferroelectrics such as CsH_2PO_4 (CDP), $PbHPO_4$ (PMP) and their deuterated analogues d-CDP and d-PMP ([18–24] and references therein).

In the present work we use our previous results for a simple ladder (two coupled Ising spin chains) to consider a more complicated model and concretely an Ising bilayer (a linear chain of simple ladders), characterized by intra-ladder and inter-ladder couplings. Through this model, which could be applied to biological lipid bilayers [25–28], we study the para- and ferroelectric behaviours of different thermodynamic properties for different ratios of interaction constants and different values of electric field, by using a transfer matrix method for the intra-ladder interactions and the MF approximation for the inter-ladder interactions. In a particular case of our model, in which the interactions between two layers (planes) are taken to be zero, our results reproduce entirely those obtained previously for a pseudo-one-dimensional Ising model, which explains well the experimental data for the thermodynamic properties of 1D ferroelectrics such as CsD_2PO_4 (d-CDP) [24].

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2. The model and its Hamiltonian

The related low-dimensional system investigated in this work is an Ising bilayer (two x–y planes) as a chain of simple ladders, characterized by intra-ladder and inter-ladder couplings. We denote by J_x the nearest-neighbour interaction along the long axis of the ladder, by J_z the nearest-neighbour interaction across the rungs of the ladder (J_x and J_z are the intra-ladder couplings) and by J_y the inter-ladder interaction between two nearest-neighbour spins (or dipoles) (figure 1). The Hamiltonian of this model for nearest-neighbour interactions is

$$H = -\sum_{i,j=1}^{N,N}\{J_x[S^{(1)}(i,j)S^{(1)}(i+1,j) + S^{(2)}(i,j)S^{(2)}(i+1,j)] + J_y[S^{(1)}(i,j)S^{(1)}(i,j+1) + S^{(2)}(i,j)S^{(2)}(i,j+1)]\} - J_z\sum_{i,j=1}^{N,N} S^{(1)}(i,j)S^{(2)}(i,j) - E\sum_{i,j=1}^{N,N}[S^{(1)}(i,j) + S^{(2)}(i,j)] \quad (1)$$

where E is the electric field and $S^{(1)}(i,j)$ or $S^{(2)}(i,j) = \pm 1$ at site (i,j) of plane (1) or (2), respectively. We will also consider periodic boundary conditions in the x and y axis directions.

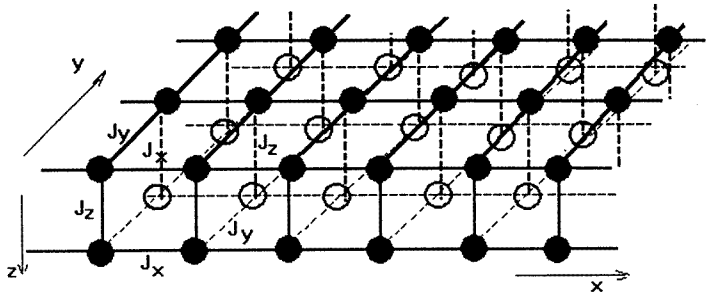

Figure 1. A schematic presentation of an Ising spin bilayer.

Through this model we study the para- and ferroelectric behaviours of different thermodynamic properties for different ratios of interaction constants and different values of electric field, by using a transfer matrix method for the intra-ladder interactions and the MF approximation for the inter-ladder interactions.

3. A ladder with two coupled Ising spin chains

In this section, following [1, 2] we consider an Ising 'ladder' with N columns or rungs and two coupled spin chains, referred to as 1 and 2. The ladder Hamiltonian with a strength-J_x interaction along the long (chain) axis of the ladder (or the x axis of the plane) and a J_z interaction across the rungs is given by

$$H_L = -J_x\sum_{i=1}^{N}[S^{(1)}(i,j)S^{(1)}(i+1,j) + S^{(2)}(i,j)S^{(2)}(i+1,j)] - J_z\sum_{i=1}^{N} S^{(1)}(i,j)S^{(2)}(i,j) - E\sum_{i=1}^{N}[S^{(1)}(i,j) + S^{(2)}(i,j)] \qquad \forall j \quad (2)$$

where $S^{(1)}(i, j)$ or $S^{(2)}(i, j) = \pm 1$ at site (i, j) (for fixed j) for the first or second chain of the ladder, respectively. We will also consider ladders with periodic boundary conditions in the long-axis direction.

By developing this Hamiltonian in 'elementary segments' (corresponding to modified squares) of the type

$$\begin{aligned}-\{J_x[S^{(1)}(i, j)S^{(1)}(i+1, j) + S^{(2)}(i, j)S^{(2)}(i+1, j)] + \frac{J_z}{2}[S^{(2)}(i, j)S^{(1)}(i, j) \\ + S^{(1)}(i+1, j)S^{(2)}(i+1, j)] \\ + \frac{E}{2}[S^{(2)}(i, j) + S^{(1)}(i, j) + S^{(1)}(i+1, j) + S^{(2)}(i+1, j)]\}\end{aligned} \tag{3}$$

the partition function can be written as

$$Z_L = \sum \exp[-H_L/(k_B T)] = \mathrm{Tr}(\mathbf{T}^N) \tag{4}$$

where k_B is the Boltzmann constant, T is the absolute temperature and $\mathbf{T}$ is the transfer matrix.

Denoting $k_x = k = J_x/(k_B T) = 1/t$ (with $t = k_B T/J_x$, the reduced temperature) $k_z = J_z/(k_B T) = rk$ (with $r = J_z/J_x$, the ratio of interaction constants), $e_0 = E/(k_B T) = ke$ (with $e = E/J_z$, the reduced field) and $x_L = \exp k$, $y_L = \exp(rk)$ and $z_L = \exp(ke)$, the transfer matrix, $\mathbf{T}$, with $2^2 \times 2^2$ elements and symmetrically related to the main diagonal, constructed following the decreasing order of bit numbers from the configuration (1111) (the four spins $S^{(1)}(i, j)$, $S^{(1)}(i+1, j)$, $S^{(2)}(i, j)$ and $S^{(2)}(i+1, j)$ are 'up') to the configuration (0000) (the four spins point 'down') is given by

$$\mathbf{T} = \begin{pmatrix} x_L^2 y_L z_L^2 & z_L & z_L & x_L^{-2} y_L \\ z_L & x_L^{-2} y_L^{-1} & x_L^2 y_L^{-1} & z_L^{-1} \\ z_L & x_L^2 y_L^{-1} & x_L^{-2} y_L^{-1} & z_L^{-1} \\ x_L^{-2} y_L & z_L^{-1} & z_L^{-1} & x_L^2 y_L z_L^{-2} \end{pmatrix} \tag{5}$$

By the usual procedure for the diagonalization of the matrix (5) we can find the largest eigenvalue $\lambda_{max}(e, k) = \lambda(e, k) = \lambda$. Since this maximum eigenvalue is equal to the grand partition function per 'elementary segment' (or two spins), in the thermodynamic limit ($N \to \infty$), the thermodynamic potential or the free energy per spin, in units of J_x, is derived as

$$g_L = g_0(e, k) = -\frac{k_B T}{J_x} \lim_{N \to \infty} [(2N)^{-1} \ln[\lambda(e, k)^N] = \frac{1}{2k} \ln(\lambda)\,. \tag{6}$$

Concretely, on diagonalizing this matrix, we find for the eigenvalues $\lambda_1 = -2\exp(-kr) \times \sinh(2k)$ and the others are the roots of the equation

$$\lambda^3 + A\lambda^2 + B\lambda + C = 0 \tag{7}$$

with

$$\begin{aligned} A &= -2\{\exp[k(2+r)]\cosh(2ke) + \exp(-kr)\cosh(2k)\} \\ B &= 4\sinh(2k)[\exp(2kr)\cosh(2k) + \exp(2k)\cosh(2ke)] \\ C &= -8\exp(rk)\sinh^3(2k)\,. \end{aligned} \tag{8}$$

Following the usual route to resolve the equation of the third order, choosing the largest eigenvalue λ among these solutions ($\lambda_1 < 0$ is omitted), after the first and second

differentiation of (7) with respect to k (or e) we find

$$\lambda' = \frac{A'\lambda^2 + B'\lambda + C'}{3\lambda^2 + 2A\lambda + B}$$
$$\lambda'' = -\frac{2\lambda'^2(3\lambda + A) + 2\lambda'(2A'\lambda + B') + (A''\lambda^2 + B''\lambda + C'')}{3\lambda^2 + 2A\lambda + B} \tag{9}$$

where A', B' and C' and A'', B'' and C'' are the first and second derivatives of A, B and C, given by (8), versus k (or e), respectively.

From the usual thermodynamic relations the specific heat is given by

$$c = c_0(e, k) = \frac{k^2}{2}\frac{\partial^2}{\partial k^2}\ln\lambda = \frac{k^2}{2}\left[\frac{\lambda''}{\lambda} - \left(\frac{\lambda'}{\lambda}\right)^2\right] \tag{10}$$

where λ' and λ'' are the first and second derivatives of λ with respect to k, respectively. For the polarization per spin p (in reduced units) and the susceptibility χ we can write

$$p = -\frac{\partial g_0}{\partial e} = \frac{1}{2k}\frac{\partial}{\partial e}\ln\lambda \qquad \chi = \frac{\partial p}{\partial k^2} = \frac{1}{2k}\frac{\partial^2}{\partial e^2}\ln\lambda\,. \tag{11}$$

4. The free energy of the bilayer

Following [24] performing a Legendre transformation on $g_0(k, \xi)$, we obtain the Helmholtz free energy for the simple Ising ladder as $f_0(k, p)$. If the inter-ladder interactions are treated by an MF approximation, then the internal energy term of the average polarization, $-J_y p^2$ is added to the free energy. Furthermore, if the bilayer is deformable or compressible along the y axis, then the elastic term $\sum \frac{1}{2}\epsilon_{ij}\xi_i\xi_j$ (which is abbreviated as $\frac{1}{2}\epsilon\xi^2$ in units of J_x) should be added to the free energy. Thereby, we have

$$f(p, k) = f_0(p, k) - \frac{J_y}{J_x}p^2 + \frac{v}{2}\epsilon\xi^2 \tag{12}$$

where ϵ is the elastic constant, ξ is the strain and v is the volume of a molecular unit. Based on the assumption above, the coupling between the polarization and the strain is taken into the inter-ladder interaction constant as the following expansion [24]:

$$J_y(\xi) \approx J_y^{(0)} + \frac{\partial J_y}{\partial_y}\xi = J_y^{(0)} + J_y^{(1)}\xi \tag{13}$$

where the higher-order terms are neglected. Equilibrium values of ξ and p are determined by two conditions:

$$\frac{\partial f}{\partial \xi} = 0 \qquad \frac{\partial f}{\partial p} = 0\,. \tag{14}$$

Considering these conditions, we can rewrite (12) as

$$f(p, k) = f_0(p, k) - \frac{J_y^{(0)}}{J_x}p^2 - \frac{(J_y^{(1)}/J_x)^2}{2v\epsilon}p^4\,. \tag{15}$$

5. The main thermodynamic quantities in the absence of an external field

5.1. The Curie temperature

By means of the usual calculations we obtain various thermodynamic quantities from the last equation (15). The molecular field, in the mean-field approximation, in reduced units (of J_x) is found to be

$$e_m = 2(J_y^{(0)}/J_x)(p + \delta p^3) = 2qp(1 + \delta p^2) \tag{16}$$

where $q = J_y^{(0)}/J_x$ and $\delta = [(J_y^{(1)}/J_x)^2](\epsilon v q)$ is a new constant related to electrostrictive corrections. In the absence of an external electric field the internal molecular field e_m will play the role of e in relations (5)–(11). So, by substituting (16) into (9), in the limit $p \to 0$, we find the equation

$$1 = 4k_c q \frac{2\exp(2k_c)[\exp(k_c r)\lambda_c - 2\sinh(2k_c)]}{3\lambda_c^2 + 2A_c\lambda_c + B_c} \tag{17}$$

with $\lambda_c = \lambda(e = 0, k_c)$, $A_c = A(e = 0, k_c)$ and $B_c = B(e = 0, k_c)$, to determine the reduced Curie temperature $t_c = 1/k_c$ as its solution. The electrostrictive correction does not appear in this limit equation, namely it does not influence the values of the critical temperature. Resolving this equation numerically, we studied the variation of the critical temperature t_c with respect to different values of inter-plane interactions r intra-plane interactions q (all interaction constants are considered positive). As an example, two of these curves, giving the value of critical temperatures for $r = 1$ and different q or for $q = 1$ and different r, are represented in figure 2. It is clearly visible from the curve with $r = 1$ that, when $q = 0$, the critical temperature is $t_c = 0$; that is, there is no longer a finite phase transition from the paraelectric phase to the ferroelectric one. In this case, for $r \neq 0$ and $q = 0$, the bilayer becomes a ladder. However, the problem of Ising ladders is straightforward with a cross-over between a 2D high-temperature region and a 1D low-temperature region when the 2D correlation length equals the width. Since there is no ordering at finite temperature in the 1D Ising model the absence of finite polarization is quite normal for $q = 0$.

In the special case in which $r = 0$ (in the case of a plane) the equation (17) is reduced to the simple equation $\exp(-2k_c) = 2k_c q$, obtained before in [24].

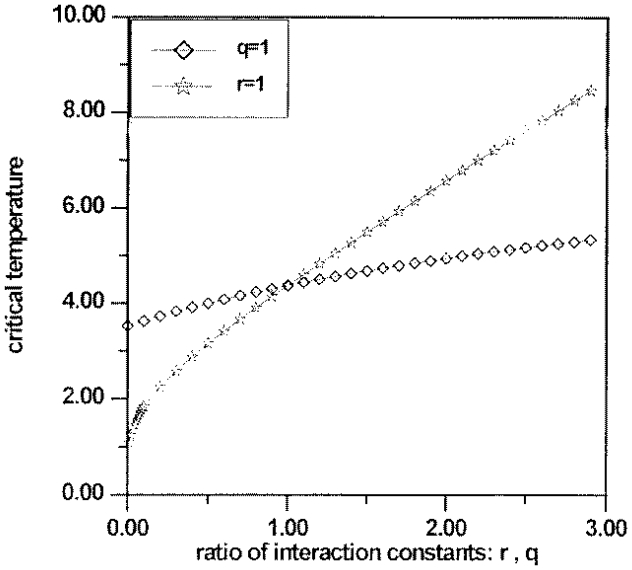

Figure 2. The variation of the critical temperature t_c for $r = 1$ (or $q = 1$) versus the ratio of interaction constants q (or r).

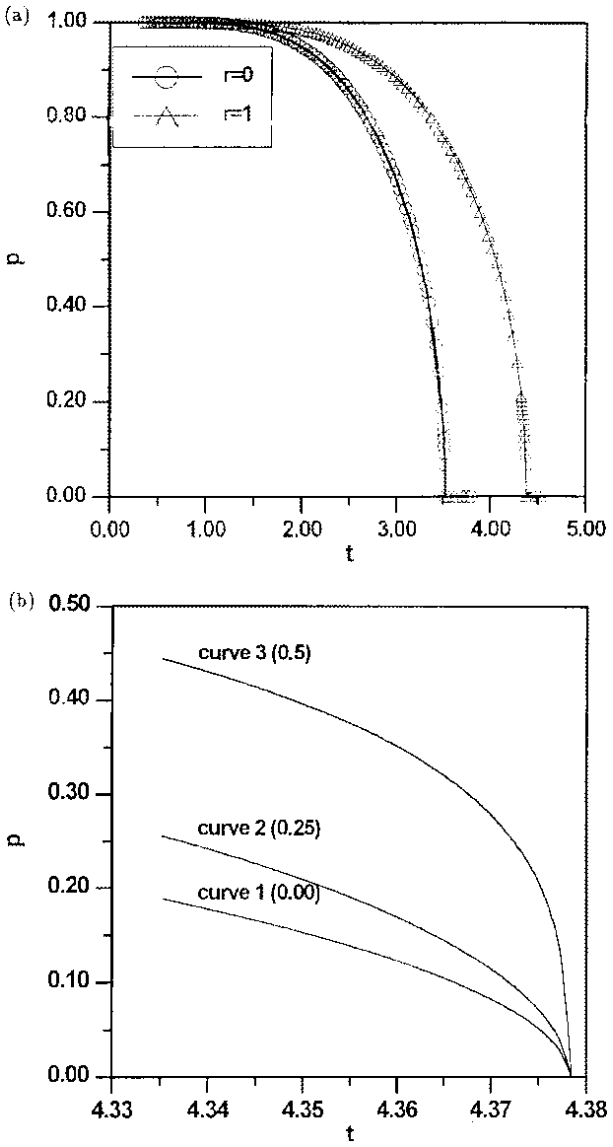

Figure 3. (*a*) The variation of the spontaneous polarization p versus the reduced temperature t for two different cases: $r = 0$, $q = 1$ and $r = 1$, $q = 1$, when the electrostrictive corrections are not taken into account ($\delta = 0$) and an external electric field is absent ($e_{ext} = 0$). (*b*) The p–t diagrams, near the critical temperature t_c, in the presence of the electromechanical coupling δ ($\delta = 0$ (curve 1), 0.25 (curve 2) and 0.5 (curve 3)), for $r = 1$, $q = 1$ and $e_{ext} = 0$.

5.2. The spontaneous polarization

From the combination of equation (8), (9) and (11), where the field e is replaced by e_m, given by (16), we obtain the self-consistent equation for the spontaneous polarization:

$$p = \frac{2\exp(2k)[\exp(kr)\lambda(e,k) - 2\sinh(2k)]}{[3\lambda^2(e,k) + 2A(e,k)\lambda(e,k) + B(e,k)]}\sinh(2ke) \tag{18}$$

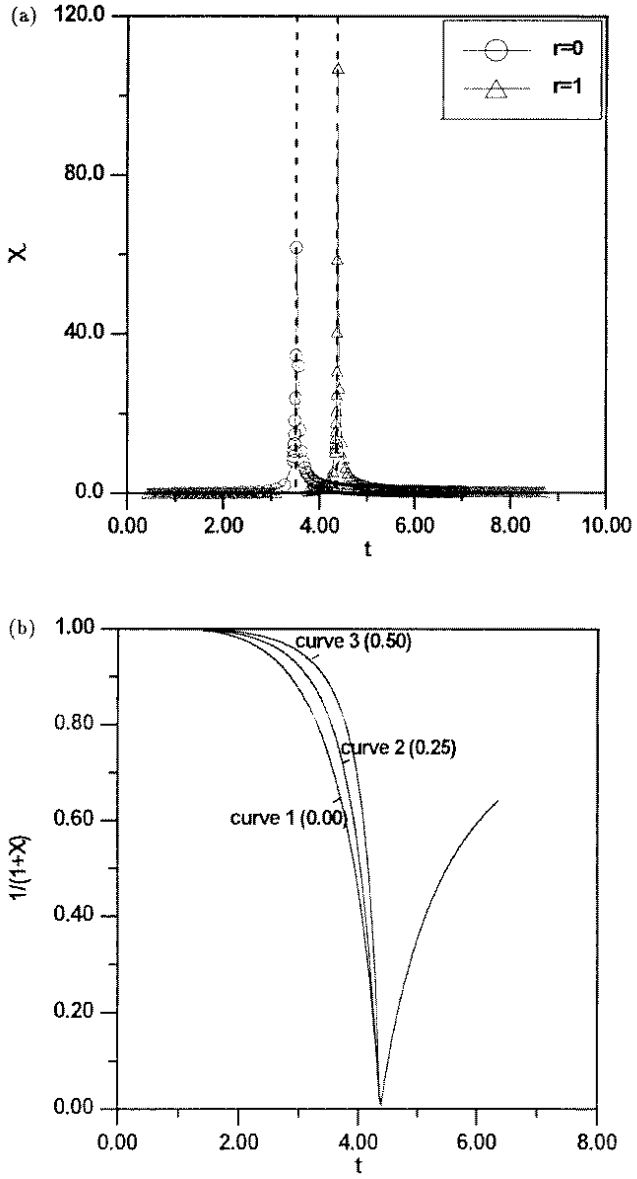

Figure 4. (*a*) The variation of the susceptibility χ versus the temperature t in two cases: $r = 0$ and $r = 1$, for $q = 1$, $\delta = 0$ and $e_{ext} = 0$. (*b*) The $[1/(1 + \chi)]$–t diagrams for three different values of δ ($\delta = 0$ (curve 1), 0.25 (curve 2) and 0.5 (curve 3)), in the case of isotropic interactions ($r = q = 1$) and $e_{ext} = 0$.

with $\lambda(e, k) = \lambda(e_m, k)$, the largest root of the third-order equation of the type (7).

The variation of the spontaneous polarization p versus the reduced temperature t for two different cases, the simple isotropic plane ($r = 0,\ q = 1$) and the isotropic bilayer ($r = 1,\ q = 1$) both with $\delta = 0$ is represented in figure 3(*a*). In figure 3(*b*) we show the influence of δ on the spontaneous polarization near the critical point. It is evident

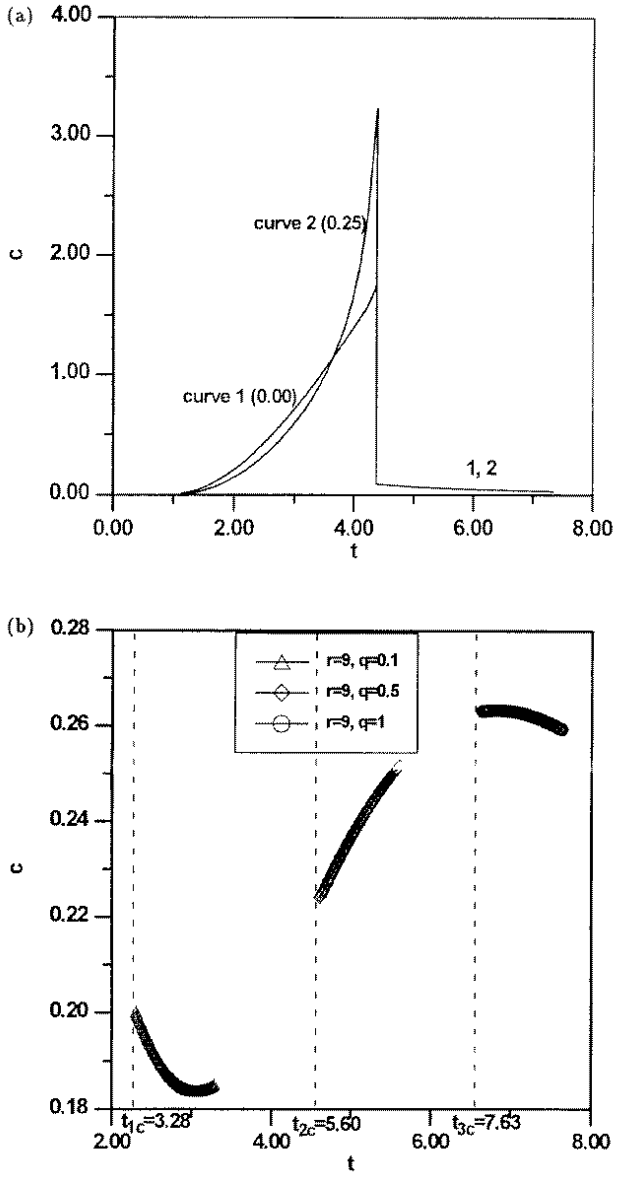

Figure 5. (*a*) The c–t diagrams for two values of the electrostrictive correction parameter δ ($\delta = 0$ (curve 1) and 0.25 (curve 2)), in the case of the isotropic interactions ($r = 1$ and $q = 1$) and $e_{ext} = 0$. (*b*) The c–t diagrams for an extreme case $r = 9$, $\delta = 0$, $e_{ext} = 0$ and $q = 0.1$, 0.5 and 1. The different shapes of the tail in these curves (for $t > t_c$) are related to the value of t_c.

from figure 3(*b*) that the slope of the polarization curves near the critical point depends clearly on the values of the electrostrictive correction δ. In the special case in which $r = 0$ equation (18) is reduced to the simple equation $p(1 - p^2)^{-1/2} \exp(-2k) = \sinh(ke_m)$ [24].

5.3. The susceptibility

Following the usual calculations for the susceptibility χ when the external field e_{ext} is taken zero, we find

$$\chi = \frac{\partial p/\partial e}{1 - 2q(1 + 3\delta p^2)(\partial p/\partial e)} \tag{19}$$

where $e = e_m + e_{ext}$ with $e_{ext} = 0$ after differentiation. For the derivative $(\partial p/\partial e)|_{e_{ext}} = 0$, after long analytical calculations, we obtain two branches, the first on one side of the critical temperature and the second on its other side (one for $t < t_c$ and the other for $t > t_c$). Their analytical expressions are not represented here. Based on these expressions and equation (19) we have studied different variations of the susceptibility χ. In figure 4(a) we have represented the variation of χ versus t, for the two different cases considered in figure 3(a). To better illustrate the influence of δ on this quantity, in the region $t < t_c$, instead of χ we have also studied the quantity $1/(1 + \chi)$ (equal to the inverse of the dielectric constant ϵ for a linear dependence of p on e) (figure 4(b)). In the special case in which $r = 0$, from (19) and the concrete expressions for $\partial p/\partial e$ we reobtain the following simple expressions: $\chi = k[\exp(-2k) - 2kq]^{-1}$ for $t > t_c$ and $\chi = k\{(1 - p^2)^{-1}[p^2 + \exp(-4k)(1 - p^2)]^{-1/2} - 2kq(1 + 3\delta p^2)\}^{-1}$ for $t < t_c$ [24].

5.4. The specific heat

The specific heat at constant field and constant stress is given by

$$c = c_0(k, e) + 2qk^3(1 + 3\delta p^2)[1 + 2q(1 + 3\delta p^2)\chi]\left(\frac{\partial p}{\partial k}\right)^2 \tag{20}$$

with $e = 2qp(1 + \delta p^2)$.

For illustration, in figure 5(a) the c–t diagrams for two values of the electrostrictive correction parameter δ ($\delta = 0$ and 0.25) are represented, in the case of an isotropic interaction model ($r = 1$ and $q = 1$). Among the different cases considered for this Ising spin bilayer, we have also studied the extreme cases in which the inter-plane interactions are quite strong. In figure 5(b) the case in which $r = 9$ is represented (while $\delta = 0$ and $q = 0.1$, 0.5 and 1). The different shapes of the tail in these curves (for $t > t_c$) can be easily explained by considering the position of the t_c related to the double peak of the specific heat for the simple Ising spin ladders [1]. In the special case in which $r = 0$, for $t > t_c$, the expression given by (20) is reduced to the simple formula $c = [k/\cosh(k)]^2$ [24].

6. The influence of an external field

In the presence of an external field e_{ext} we substitute $e = 2qp(1 + \delta p^2)$ by $e = 2qp(1 + \delta p^2) + e_{ext}$. Some of the results obtained in this case are illustrated in figure 6(a)–(c). In figure 6(a) the variation of p with respect to t is represented, in the presence of an external field, for three different cases (with $r = 1$ and $q = 1$). Curve 1 with $\delta = 0.1$ and curve 2 with $\delta = 0.5$ illustrate the influence of the electrostrictive correction δ for $e_{ext} = 0.1$, while the comparison between curve 1 with $e_{ext} = 0.1$ and $\delta = 0.1$ and curve 3 with $e_{ext} = 0.3$ and $\delta = 0.1$ illustrates the tendency of the external field to broaden the p–t diagram. In figure 6(b) and (c) we have represented the influence of the electrostrictive

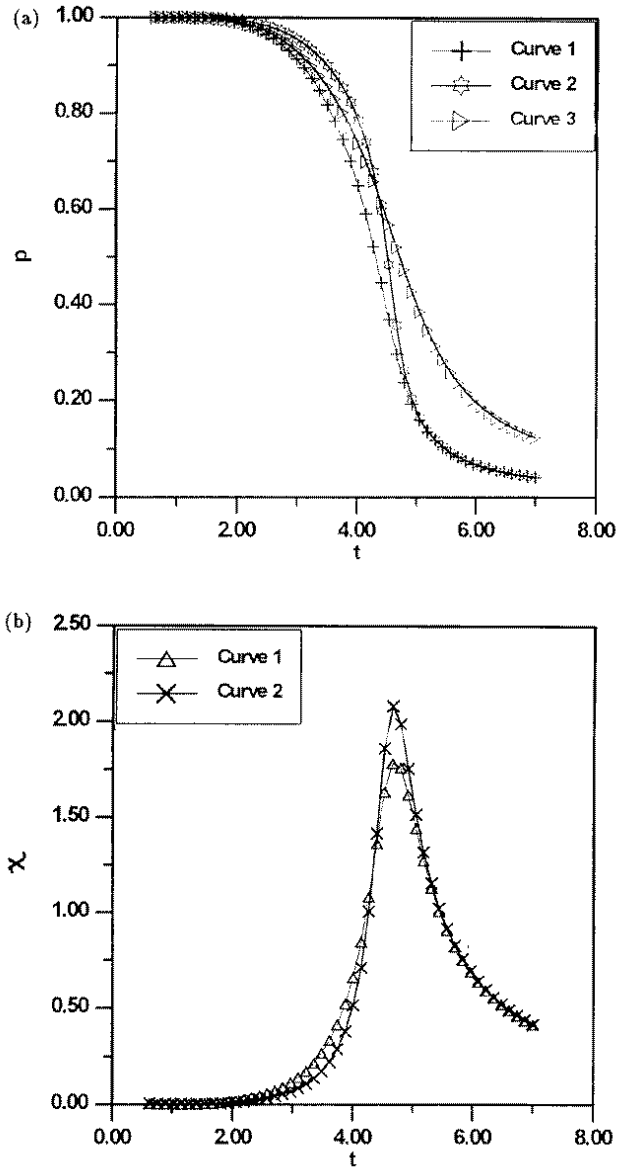

Figure 6. (*a*) The variation of p with respect to t, in the presence of an external field, for three different cases with $r = 1$, $q = 1$ and $\delta = 0.1$, $e_{ext} = 0.1$ (curve 1); $\delta = 0.5$, $e_{ext} = 0.1$ (curve 2) and $\delta = 0.1$, $e_{ext} = 0.3$ (curve 3). (*b*) The χ–t diagrams for two cases (both with $r = 1$, $q = 1$ and $e_{ext} = 0.1$) with $\delta = 0$ (curve 1) and $\delta = 0.25$ (curve 2). (*c*) The c–t diagrams for two cases (both with $r = 1$, $q = 1$ and $e_{ext} = 0.1$) with $\delta = 0$ (curve 1) and $\delta = 0.25$ (curve 2).

corrections on the χ–t and c–t diagrams, respectively, for $e_{ext} = 0.1$ ($r = 1$ and $q = 1$), comparing the case without the electrostrictive corrections, $\delta = 0$ (curve 1), with the case in which $\delta = 0.25$ (curve 2).

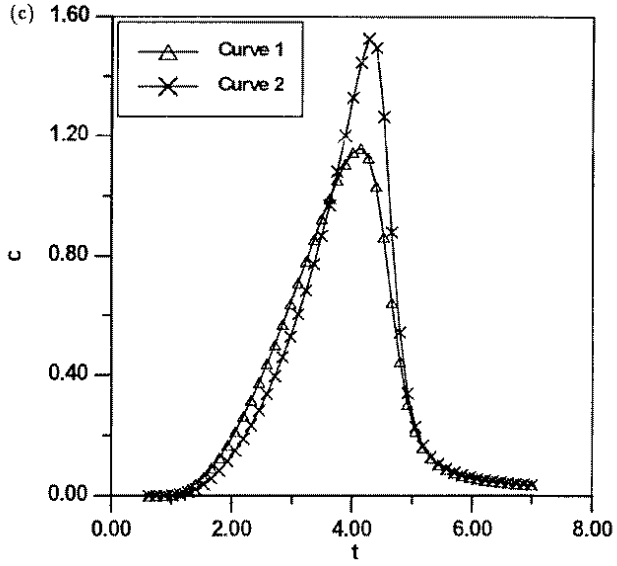

Figure 6. Continued.

7. Conclusions

In this paper we have considered the Ising spin bilayer in order to study in detail the behaviour of different thermodynamic quantities such as the polarization, the susceptibility and the specific heat, related to different values of the temperature, the ratio of interaction constants, the electromechanical coupling and the electric field. Our results are intermediate between the results obtained entirely analytically and those obtained by using the MF approximation. In the special case in which inter-plane or inter-layer interactions are absent, we again find the same results as those obtained previously to explain the experimental data for the static thermodynamic properties of CDP crystals in the para- and ferroelectric phases. Considering the fact that there are spin pairings in the spin-ladder systems when inter-chain couplings of the antiferromagnetic type are present, we intend to study carefully interactions of this kind for inter-layer and intra-layer interactions.

Acknowledgments

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