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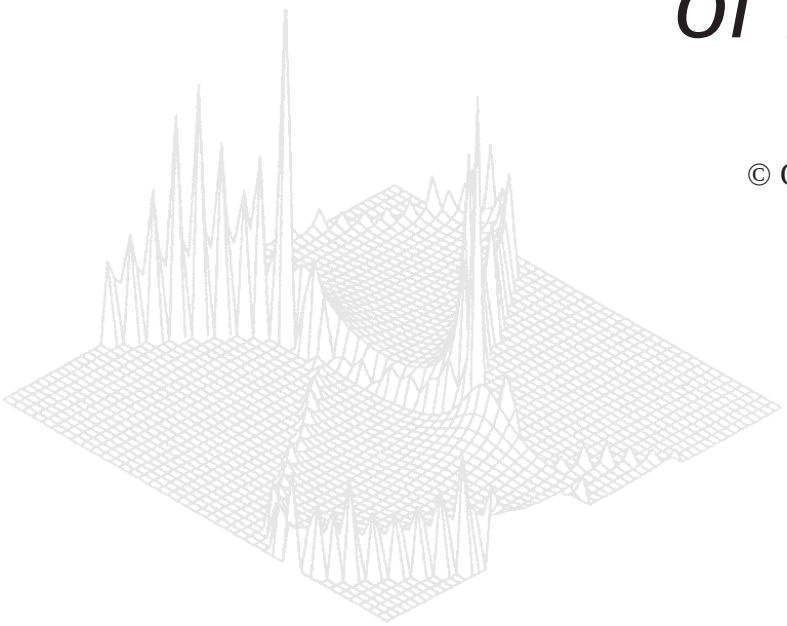
Australian Journal of Physics

Volume 53, 2000



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Aust. J. Phys., 2000, **53**, 453–9

Phase Transition Properties of a Finite Ferroelectric Superlattice from the Transverse Ising Model

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Abstract

We consider a finite ferroelectric superlattice in which the elementary unit cell is made up of l atomic layers of type A and n atomic layers of type B. Based on the transverse Ising model we examine the phase transition properties of the ferroelectric superlattice. Using the transfer matrix method we derive the equation for the Curie temperature of the superlattice. Numerical results are given for the dependence of the Curie temperature on the thickness and exchange constants of the superlattice.

1. Introduction

Possibly because of the great difficulty of growing well characterised samples, experimental studies of ferroelectric superlattices have been published only in recent years (Iijima *et al.* 1992; Tsurumi *et al.* 1994; Wiener-Avnear 1994; Tabata *et al.* 1994; Kanno *et al.* 1996; Zhao *et al.* 1999). Some exploratory theoretical work on ferroelectric superlattices has appeared (Tilley 1988; Schwenk *et al.* 1988, 1990). Their starting point is the Ginzburg–Laudau phenomenological theory.

On the microscopic level, the transverse Ising model (TIM) (de Gennes 1963; Binder 1987; Tilley and Zeks 1984; Cottam *et al.* 1984) was used to study infinite ferroelectric superlattices under mean field theory (Qu *et al.* 1994, 1995; Zhong and Smith 1998) or effective field theory (Zhou and Yang 1997). From the experimental point of view the TIM is a valuable model because of its possible applications, for example, in studies of hydrogen bonded ferroelectrics (de Gennes 1963), cooperative Jahn–Teller systems (Elliot *et al.* 1971) and strongly anisotropic magnetic materials in a transverse field (Wang and Cooper 1968). The reviews of Blinc and Zeks (1972) and Stinchcombe (1973) give more details about possible applications of the TIM.

In the present paper, we consider a finite ferroelectric superlattice in which the elementary unit cell is made up of l atomic layers of type A and n atomic layers of type B. The mean-field approximation is employed and the equation for the Curie temperature is obtained by use of the transfer matrix method. We study two models of the superlattice which alternate as $ABAB...AB$ (Model I) or

 10.1071/PH99080 0004-9506/00/030453$05.00

ABABA...BA (Model II). Numerical results are given for the dependence of the Curie temperature on the thickness and exchange constants of the superlattice.

2. The Curie Temperature

We start with the TIM (de Gennes 1963; Sy 1993; Qu *et al.* 1994, 1995; Zhong and Smith 1998; Bouziane *et al.* 1999)

$$H = -\tfrac{1}{2} \sum_{(i,j)} \sum_{(r,r')} J_{ij} S^z_{ir} S^z_{jr'} - \sum_{ir} \Omega_i S^x_{ir} \,, \tag{1}$$

where S^x_{ir}, S^z_{ir} are the x and z components of the pseudo-spin, (i,j) are plane indices and (r,r') are different sites of the planes, and J_{ij} denote the exchange constants. We assume that the transverse field Ω_i is dependent only on the layer index and consider the interaction between nearest neighbouring sites. For simplicity, we take Ω to be the same in the superlattice because the important qualitative features result from the difference of J_{ij}.

The spin average $\langle \vec{S}_i \rangle$ is obtained from the mean field theory

$$\langle \vec{S}_i \rangle = \frac{\vec{H}_i}{2|\vec{H}_i|} \tanh\left(\frac{|\vec{H}_i|}{2k_B T}\right), \tag{2}$$

where $\vec{H}_i(\Omega, 0, \sum_j J_{ij} \langle S^z_j \rangle)$ is the mean field acting on the ith spin, k_B is the Boltzmann constant and T is the temperature.

At a temperature close to and below the Curie temperature, $\langle S^x_i \rangle$ and $\langle S^z_i \rangle$ are small, $|\vec{H}_i| \approx \Omega$, and equation (2) can be approximated as

$$\langle S^x_i \rangle = \tfrac{1}{2} \tanh\left(\frac{\Omega}{2k_B T}\right), \tag{3}$$

$$\langle S^z_i \rangle = \frac{1}{2\Omega} \tanh\left(\frac{\Omega}{2k_B T}\right) [z_0 J_{ii} \langle S^z_i \rangle + z(J_{i,i+1} \langle S^z_{i+1} \rangle + J_{i,i-1} \langle S^z_{i-1} \rangle)] \,. \tag{4}$$

Here z_0 and z are the numbers of nearest neighbours in a certain plane and between successive planes respectively.

Let us rewrite equation (4) in matrix form in analogy with Barnas (1992):

$$\begin{pmatrix} m_{i+1} \\ m_i \end{pmatrix} = M_i \begin{pmatrix} m_i \\ m_{i-1} \end{pmatrix}, \tag{5}$$

with M_i as the transfer matrix defined by

$$M_i = \begin{pmatrix} (\tau - z_0 J_{ii})/z J_{i,i+1} & -J_{i,i-1}/J_{i,i+1} \\ 1 & 0 \end{pmatrix}, \tag{6}$$

where $m_i = \langle S^z_i \rangle$ and $\tau = 2\Omega / z J_{i,i+1} \coth[\Omega/(2k_B T)]$.

We consider a ferroelectric superlattice which alternates as $ABAB...AB$. In each elementary unit AB, there are l atomic layers of type A and n atomic layers of type B. The intralayer exchange constants are given by J_A and J_B, whereas the exchange constant between different layers is described by J_{AB}. We assume that there are N elementary units and the layer index is from 0 to $N(l+n)-1$. In this case, the transfer matrix M_i reduces to two types:

$$M_A = \begin{pmatrix} X_A & -1 \\ 1 & 0 \end{pmatrix}, \qquad M_B = \begin{pmatrix} X_B & -1 \\ 1 & 0 \end{pmatrix}, \tag{7}$$

where $X_A = \tau - j_A$, $X_B = \tau - j_B$, $j_A = z_0 J_A / z J_{AB}$, $j_B = z_0 J_B / z J_{AB}$, and $\tau = 2\Omega / z J_{AB} \coth[\Omega / 2 k_B T]$.

From equation (5), we get

$$\begin{pmatrix} m_{N(l+n)-1} \\ m_{N(l+n)-2} \end{pmatrix} = R \begin{pmatrix} m_1 \\ m_0 \end{pmatrix}, \tag{8}$$

where

$$R = M_B^{n-1} (M_A^l M_B^n)^{N-1} M_A^{l-1} \tag{9}$$

is the total transfer matrix.

From the above equation and the following,

$$m_1 = X_A m_0 \,, \qquad m_{N(l+n)-2} = X_B m_{N(l+n)-1} \,, \tag{10}$$

we obtain the equation for the Curie temperature of the superlattice as

$$R_{11} X_A X_B + R_{12} X_B - R_{21} X_A - R_{22} = 0 \,. \tag{11}$$

Next we consider Model II, the superlattice which alternates as $ABA...BA$, and assume that the superlattice has $N(l+n)+l$ layers. The total transfer matrix becomes

$$S = M_A^{l-1} M_B R \tag{12}$$

and the equation for the Curie temperature is obtained as

$$S_{11} X_A^2 + (S_{12} - S_{21}) X_A - S_{22} = 0 \,. \tag{13}$$

For a unimodular matrix M, the nth power of M can be linearised as (Yariv and Yeh 1992; Wang *et al.* 1999*a*, 1999*b*)

$$M^n = U_n M - U_{n-1} I \,, \tag{14}$$

where I is the unit matrix, $U_n = (\lambda_+^n - \lambda_-^n)/(\lambda_+ - \lambda_-)$, and $\lambda_\pm$ are the two eigenvalues of the matrix M.

Using equation (14), we obtain

$$M_A^l = E_l M_A - E_{l-1} I \,, \tag{15}$$

$$M_B^n = F_n M_B - F_{n-1} I \,, \tag{16}$$

where $E_l = (\alpha_+^l - \alpha_-^l)/(\alpha_+ - \alpha_-)$, $F_n = (\beta_+^n - \beta_-^n)/(\beta_+ - \beta_-)$, $\alpha_\pm = (X_A \pm \sqrt{X_A^2 - 4})/2$ and $\beta_\pm = (X_B \pm \sqrt{X_B^2 - 4})/2$. Then from equations (15) and (16), the matrix $M_A^l M_B^n$ in equation (9) can be written explicitly as

$$M_{AB} = M_A^l M_B^n \tag{17}$$

$$= \begin{pmatrix} (E_l X_a - E_{l-1})(F_n X_b - E_{n-1}) - E_l F_n & -(E_l X_a - E_{l-1}) F_n + E_l E_{n-1} \\ E_l (F_n X_b - E_{n-1}) - E_{l-1} F_n & -E_l F_n + E_{l-1} E_{n-1} \end{pmatrix} .$$

The trace of the matrix M_{AB} is

$$\mathrm{tr} = (E_l X_a - E_{l-1})(F_n X_b - E_{n-1}) - 2E_l F_n + E_{l-1} E_{n-1} \,. \tag{18}$$

Since $\det(M_{AB}) = 1$, the eigenvalues of the matrix M_{AB} are $\gamma_\pm = (\mathrm{tr} \pm \sqrt{\mathrm{tr}^2 - 4})/2$. Then using equation (14), we get

$$M_{AB}^{N-1} = G_{N-1} M_{AB} - G_{N-2} I \,, \tag{19}$$

where $G_N = (\gamma_+^N - \gamma_-^N)/(\gamma_+ - \gamma_-)$.

Using equations (15)–(19), we express the total transfer matrix R and S in terms of X_A, X_B, E_1, F_n and G_N. We can get an explicit expression for equations (11) and (13) for the Curie temperature by substituting the matrix

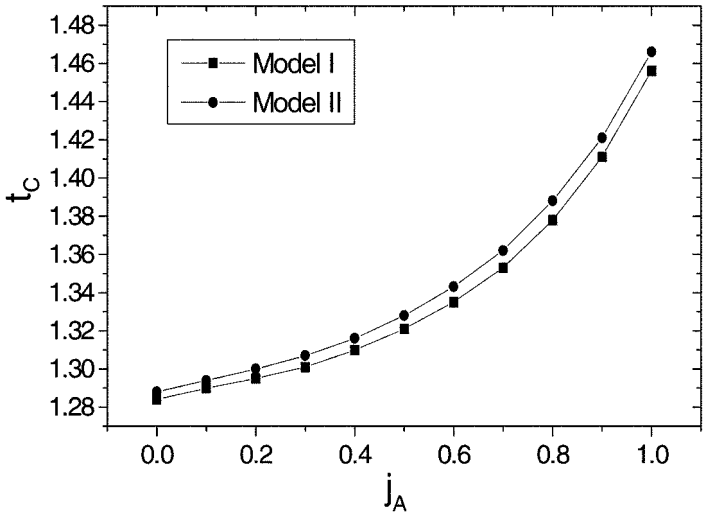

Fig. 1. Dependence of the reduced Curie temperature t_C on the reduced exchange constant j_A in Models I and II. The parameters are $j_B = 1$, $l = n = N = 2$ and $\omega = 0{\cdot}5$.

elements of R and S into equations (11) and (13), respectively. The results are tedious, and we only give the numerical results.

Fig. 1 gives the dependence of the reduced Curie temperature t_C against the reduced exchange constant j_A in Models I and II. The Curie temperature increases with an increase in j_A. It is clear that the Curie temperatures in Model II are larger than those in Model I. The reason is that the superlattice in Model II is thicker than that in Model I. The fact that the Curie temperature increases with an increase in j_A can also be seen in Fig. 2. This shows the dependence

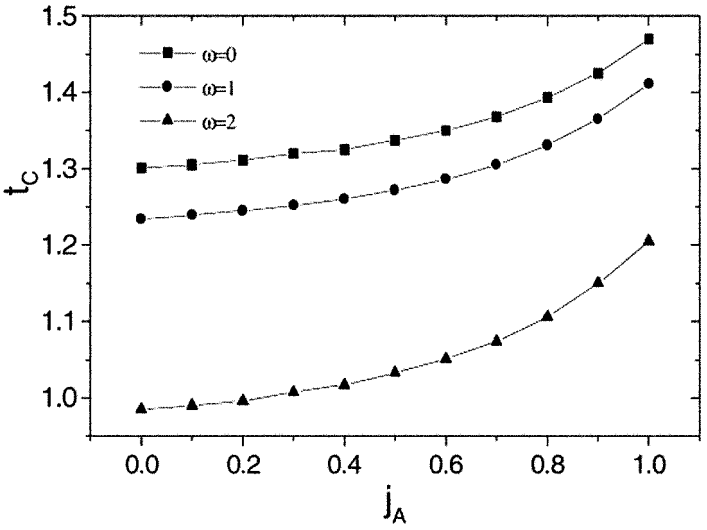

Fig. 2. Dependence of the reduced Curie temperature t_C on the reduced exchange constant j_A for different ω in Model I. The parameters are $j_B = 1$ and $l = n = N = 2$.

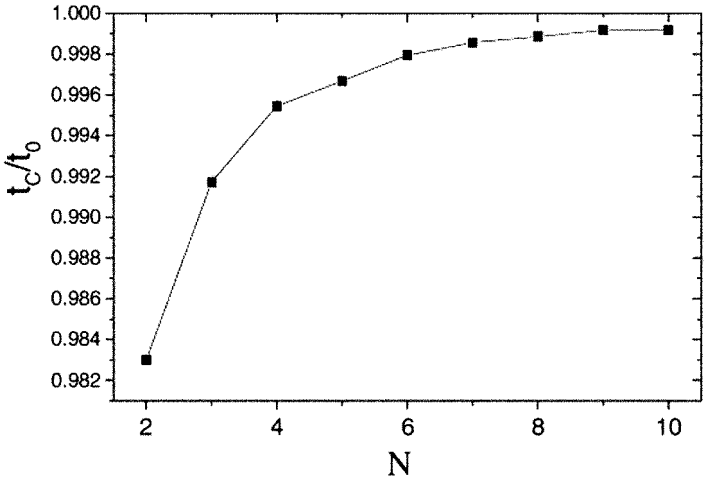

Fig. 3. Dependence of the Curie temperature on the number of elementary units N in Model I. The parameters are $j_A = 1 \cdot 2$, $j_B = 1$, $l = n = 2$ and $\omega = 0 \cdot 5$.

of the reduced Curie temperature t_C against the reduced exchange constant j_A for different ω in Model I. The transverse field causes a reduction in the Curie temperature. In other words, the Curie temperature decreases with an increase in ω.

Fig. 3 shows the dependence of the Curie temperature on the number of elementary units N in Model I. Here t_0 is the Curie temperature of the corresponding infinite superlattice, which can be determined from the equation (Wang *et al.* 1999*a*, 1999*b*)

$$\text{trace}\,(M_A^l M_B^n) = 2\,. \tag{20}$$

The Curie temperature of a finite superlattice is always less than that of a corresponding infinite superlattice, and it increases with an increase in the number of elementary units N which approach t_0 asymptotically, for large values of N.

3. Conclusion

In conclusion, we have studied the phase transition properties of a finite ferroelectric superlattice in which the elementary unit cell is made up of l atomic layers of type A and n atomic layers of type B. Using the transfer matrix method we derived the equation for the Curie temperature of the superlattice. Numerical results are given for the dependence of the Curie temperature on the thickness and exchange constants of the superlattice. The method proposed here can be applied to the finite superlattice in which each elementary unit cell is made up of many types of materials and the atomic layers of each type can be arbitrary. Experimentally, the finite superlattice is more realistic than the infinite superlattice. We hope that the present work will have relevance to future experiments.

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Manuscript received 23 September 1999, accepted 5 January 2000