Phase transition properties of a finite ferroelectric superlattice from the transverse Ising model

Xiao-Guang Wang¹,² * Ning-Ning Liu² Shao-Hua Pan¹,² and Guo-Zhen Yang¹,²

1.China Center of Advanced Science and Technology (World Laboratory), P.O.Box 8730, Beijing 100080, People’s Republic of China
2.Laboratory of Optical Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100080, People’s Republic of China

(December 31, 2021)

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.

*email:xyw@aphy.iphy.ac.cn, Ref.number:PH99080.
I. INTRODUCTION

Possibly because of the great difficulty of growing well characterized 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, Tanaka and Kawai 1994; Kanno et al. 1996; Zhao T. et al. 1999). Some exploratory theoretical work on ferroelectric superlattices has appeared (Tilley 1988; Schwenk, Fishman and Schwabl 1988; Schwenk, Fishman and Schwabl 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, Tilley and Zeks 1984) was used to study infinite ferroelectric superlattices under mean field theory (Qu, Zhong and Zhang 1994; Qu, Zhong and Zhang 1995; Zhong and Smith 1998) or effective field theory (Zhou and Yang). 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 ABABA...BA (Model II). Numerical results are given for the dependence of the Curie temperature on the thickness and exchange constants of the superlattice.
II. THE CURIE TEMPERATURE

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

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

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, \( J_{ij} \) denote the exchange constants. We assume that the transverse field \( \Omega_i \) is dependent only on layer index and consider the interaction between neighboring sites. For simplicity, we take \( \Omega \) the same in the superlattice because the main qualitative important features result from the difference of \( J_{ij} \).

The spin average \( \langle \vec{S}_i \rangle \), obtained from the mean field theory

\[ \langle \vec{S}_i \rangle = \frac{\vec{H}_i}{2|\vec{H}_i|} \tanh(\frac{|\vec{H}_i|}{2k_BT}) \]  

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

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

\[ \langle S^x_i \rangle = \frac{1}{2} \tanh(\frac{\Omega}{2k_BT}) \]  

\[ \langle S^z_i \rangle = \frac{1}{2\Omega} \tanh(\frac{\Omega}{2k_BT})[z_0 J_{ii} \langle S^z_i \rangle + z(J_{i,i+1} \langle S^z_{i+1} \rangle + J_{i,i-1} \langle S^z_{i-1} \rangle)] \]  

Here \( z_0 \) and \( z \) are the numbers of nearest neighbors in a certain plane and between successive planes respectively.

Let us rewrite Eq.(4) in matrix form in analogy with the reference (Barnas 1992)

\[ \begin{pmatrix} m_{i+1} \\ m_i \end{pmatrix} = M_i \begin{pmatrix} m_i \\ m_{i-1} \end{pmatrix} \]  

with \( M_i \) as the transfer matrix defined by

\[ M_i = \begin{pmatrix} (\tau - z_0 J_{ii})/(z J_{i,i+1}) & -J_{i,i-1}/J_{i,i+1} \\ 1 & 0 \end{pmatrix}. \]  

where \( m_i = \langle S^z_i \rangle \) and \( \tau = 2\Omega/(zJ_{i,i+1}) \coth[\Omega/(2k_BT)] \).
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 constants between different layers is described by \( J_{AB} \). We assume 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:

\[
M_A = \begin{pmatrix} X_A & -1 \\ 1 & 0 \end{pmatrix}, M_B = \begin{pmatrix} X_B & -1 \\ 1 & 0 \end{pmatrix},
\]

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 \( \tau = 2\Omega/(z J_{AB}) \coth[\Omega/(2k_BT)] \).

From Eq. (5), we get

\[
\begin{pmatrix} m_{N(l+n)-1} \\ m_{N(l+n)-2} \end{pmatrix} = R \begin{pmatrix} m_1 \\ m_0 \end{pmatrix}
\]

where

\[
R = M_B^{-1}(M_A^l M_B^n)^{N-1} M_A^{-1}
\]

is the total transfer matrix.

From the above equation and the following equations

\[
m_1 = X_A m_0, m_{N(l+n)-2} = X_B m_{N(l+n)-1},
\]

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

\[
R_{11} X_A X_B + R_{12} X_B - R_{21} X_A - R_{22} = 0.
\]

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

\[
S = M_A^{l-1} M_B R
\]

and the equation for the Curie temperature is obtained as

\[
S_{11} X_A^2 + (S_{12} - S_{21}) X_A - S_{22} = 0.
\]
For an unimodular matrix $M$, the $n$-th power of $M$ can be linearized as (Yariv 1992; Wang, Pan and Yang 1999)

$$M^n = U_n M - U_{n-1} I,$$  \hfill (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 Eq.(14), we obtain

$$M^n_A = E^n_l M_A - E^n_{l-1} I,$$  \hfill (15)

$$M^n_B = F^n_n M_B - F^n_{n-1} I,$$  \hfill (16)

where $E^n_l = (\alpha^n_+ - \alpha^n_-)/(\alpha_+ - \alpha_-)$, $F^n_n = (\beta^n_+ - \beta^n_-)/(\beta_+ - \beta_-)$, $\alpha_\pm = (X_A \pm \sqrt{X_A^2 - 4})/2$ and $\beta_\pm = (X_B \pm \sqrt{X_B^2 - 4})/2$. Then from Eqs.(15) and (16), the matrix $M^n_A M^n_B$ in Eq.(9) can be written explicitly as

$$M_{AB} = M^n_A M^n_B = \begin{pmatrix}
(E_l X_a - E_{l-1}) (F_n X_b - E_{n-1}) - E_l F_n & -(E_l X_a - E_{l-1}) F_n + E_l E_{n-1} \\
E_l (F_n X_b - E_{n-1}) - E_{l-1} F_n & -E_l F_n + E_{l-1} E_{n-1}
\end{pmatrix}$$  \hfill (17)

The trace of the matrix $M_{AB}$ is

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

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

$$M^{-N}_{AB} = G_{N-1} M_{AB} - G_{N-2} I,$$  \hfill (19)

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

Using Eq.(15)-(19), we can express the total transfer matrix $R$ and $S$ in terms of $X_A, X_B, E_l F_n$, and $G_N$. We can get an explicit expression for equations (11) and (13) for the
Curie temperature by substituting the matrix elements of $R$ and $S$ into Eq.(11) and (13), respectively. The results are tedious, we only give numerical results below.

Fig.1 gives the dependence of the reduced Curie temperature $t_C$ against the reduced exchange constant $j_A$ in model I and II. The Curie temperature increases with increase of $j_A$. 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 the increase of $j_A$ can also be seen in Fig.2. Fig.2 shows the dependence of the reduced Curie temperature $t_C$ against the reduced exchange constant $j_A$ for different $\omega$ in model I. The transverse field causes a reduction of the Curie temperature. In other words, the Curie temperature decreases with increase of $\omega$.

Fig.3 shows the dependence of the Curie temperature on the number of elementary units $N$ in model I. $t_0$ in the figure is the Curie temperature of the corresponding infinite superlattice. The Curie temperature of the infinite superlattice can be determined from the following equation (Wang, Pan and Yang 1999)

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

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

**III. 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 $B$. By 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. The
finite superlattice is more realistic than the infinite superlattice in experiments. We hope that the present work will have relevance to some future experiments.

Captions:

Fig.1, The dependence of the reduced Curie temperature $t_C$ against the reduced exchange constant $j_A$ in model I and II. The parameters $j_B = 1, l = n = N = 2$, and $\omega = 0.5$.

Fig.2, The dependence of the reduced Curie temperature $t_C$ against the reduced exchange constant $j_A$ for different $\omega$ in model I. The parameters $j_B = 1$, and $l = n = N = 2$.

Fig.3, The dependence of the Curie temperature on the number of elementary units $N$ in model I. The parameters $j_A = 1.2, j_B = 1, l = n = 2$, and $\omega = 0.5$.

References

Barnas, J. (1992). *Phys.Rev.B.* 45, 10427.

Binder, K. (1987). *Ferroelectrics* 35, 99.

Blinc, R., and Zeks, B. (1972). *Adv.Phys.* 1, 693.

Bouziane, T., Saber, M., Belaaraj, A., and Ainane, A. (1999). *J.Magn.Magn.Materials* 195, 220.

Cottam, M.G., Tilley, D.R. and Zeks, B. (1994). *J.Phys.C* 17, 1793.

de Gennes, P.G. (1963). *Solid State Commun.* 1, 132.

Elliot, R.J., Gehring, G.A., Malogemoff, A.P., Smith, S.R.P., Staude, N.S., and Tyte, R.N. (1971). *J.Phys. C* 4, L179.

Iijima, K., Terashima, T., Bando, Y., Kamigaki, K. and Terauchi, H. (1992). *Jpn.J.Appl.Phys.* 72, 2840.

Kanno, I., Hayashi, S., Takayama, R. and Hirao, T. (1996). *Appl.Phys.Lett.* 68, 328.

Qu, B.D., Zhong, W.L. and Zhang, P.L. (1994). *Phys.Lett.A* 189, 419.
Qu, B.D., Zhong, W.L. and Zhang, P.L. (1995). *Jpn.J.Appl.Phys.* **34**,4114.
Schwenk, D., Fishman, F. and Schwabl, F. (1988). *Phys.Rev.* B **38**,11618.
Schwenk, D., Fishman, F. and Schwabl, F., (1990). *J.Phys.:Condens.Matter* **2**,6409.
Stinchcombe, R.B. (1973). *J.Phys.C* **6**, 2459.
Sy, H.K. (1993). *J.Phys.:Condens.Matter* **5**, 1213.
Tabata, H., Tanaka, H. and Kawai, T. (1994). *Appl.Phys.Lett.* **65**,1970.
Tilley, D.R. and Zeks, B. (1984). *Solid State Commun.* **49**,823.
Tilley, D.R.(1988). *Solid State Commun.* **65**,657.
Tsurumi, T., Suzuki, T., Yamane, M.and Daimon, M.(1994). *Jpn.J.Appl.Phys.* **33**,5192.
Wang, X.G., Pan, S.H. and Yang, G.Z. (1999). *J.Phys.:Condens.Matter* **11**, 6581.
Wang, X.G., Pan, S.H. and Yang, G.Z. (1999). *Solid State Commun.* **113**, 59.
Wang, Y.L. and Cooper, B. (1968). *Phys.Rev.* **173**, 539.
Wiener-Avnear, E.(1994). *Appl.Phys.Lett.* **65** ,1784.
Yariv, A. and Yeh, P. (1992). *Optical Waves in Crystals* (John Wiley & Sons, New York).
Zhao, T., Chen, Z.H., Chen, F., Shi, W.S.,Lu, H.B. and Yang, G.Z. (1999). *Phys.Rev.* B **60**,1697.
Zhong, W.L. and Smith, S.R.P. (1998). *J. Korea Physical Society* **32**,S382.
Zhou, J.H. and Yang, C.Z. (1997). *Solid State Commun.* **101**,639.
Figure 2
Figure 3