

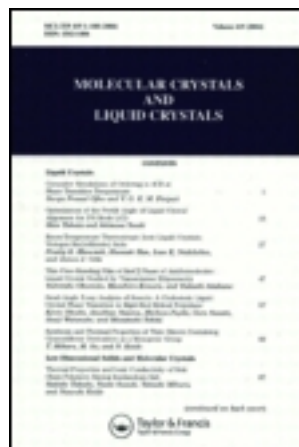
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### Correlation Functions at Antiferroelectric Smectics by Means of Transfer Matrix Method

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## CORRELATION FUNCTIONS AT ANTIFERROELECTRIC SMECTICS BY MEANS OF TRANSFER MATRIX METHOD

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*The mechanism of successive phase transitions occurring at antiferroelectric smectics is well described by ANNNI model. Here the transfer matrix method is generalised so as to be applied to this model with long range interaction. The correlation function is obtained, which shows oscillating behaviour with varying wave number due to the complex eigenvalue of transfer matrix. In the framework of this method the critical temperature of the model is estimated by replacing an intralayer interaction by the molecular field.*

**Keywords:** antiferroelectric smectics; incommensurability; block-spin; transfer matrix method; correlation function

## INTRODUCTION

In antiferroelectric smectics, various types of layer stacking appear because of a frustration among interactions [1,2]. Various types of models have been introduced to explain a successive phase transitions occurring in the antiferroelectric smectic materials [2–9]. Among them, an axial next nearest neighbour Ising model (ANNNI model [10,11]) with the third nearest neighbour interaction is successful [3,4] in the light of the recent experimental results on the structures of the mesophases [12–14]. This model is characterised by the discrete variable and frustration among interactions.

In the theoretical studies of the successive phase transitions, the mean field approximation (or, the phenomenological theory) has been utilized

exclusively [3–9], which is useful to see the global feature of the phenomena in spite of its simplicity. However, such theory is not adequate in case the correlation is to be discussed. In the previous work [15], anisotropic character of the antiferroelectric smectics is studied on the basis of the transfer matrix method [16,17], where the correlation in the direction of layer normal is shown to be quite weak in comparison with the one in the smectic layer, while long range order is isotropic. This result illustrates that the study of the correlation functions is effective to elucidate an anisotropic property of the system.

The ANNNI model consists of the nearest neighbour interaction  $J_1$  and of the second nearest neighbour one  $J_2$  ( $<0$ ) along layer normal together with the nearest neighbour interaction  $J$  within the layer, and due to the negativeness of  $J_2$  the system is strongly frustrated. In the article, the correlation in this frustrated model is studied by generalising the transfer matrix method by introducing a block-spin composed of a couple of spins in the adjacent layer. We calculate the correlation functions without the intralayer interaction in the parameter range of ferroelectric phase ( $\text{SmC}^*$ ), antiferroelectric phase ( $\text{SmC}_A^*$ ), ferrielectric phase ( $\text{SmC}_\gamma^*$ ), another type of antiferroelectric phase (AF) and an intermediate one with period 6. To test an accuracy of the present method, the critical temperature is obtained by replacing the intralayer interaction by a molecular field.

## BLOCK-SPIN MODEL

The Hamiltonian of the ANNNI model is given by

$$H = -J \sum_{(i,j)} s_i s_j - J_1 \sum_i s_i s_{i+1} - J_2 \sum_i s_i s_{i+2} - E \sum_i s_i, \quad (1)$$

where  $s_i$  denotes the Ising spin ( $\pm 1$ ) designating the directions of molecular tilt and  $E$  an external field. As the Hamiltonian contains second nearest neighbour interaction, it is impossible to apply the transfer matrix method directly to this model. Here, we rewrite the Hamiltonian into a block-spin model to be tractable by the transfer matrix method, in which a couple of spins in the successive two layers (say, block-layer) are combined into a block-spin and the interaction is only in the nearest neighbour pairs of block-spins. The  $n$ -th block-spin  $\sigma_n$  is defined by a vector with four components corresponding to states of succeeding couple of spins,  $(\uparrow\uparrow), (\uparrow\downarrow), (\downarrow\uparrow)$  and  $(\downarrow\downarrow)$ ;

$$\begin{aligned}
(\uparrow\uparrow) \Rightarrow \sigma_n &= \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix}, & (\uparrow\downarrow) \Rightarrow \sigma_n &= \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix}, \\
(\downarrow\uparrow) \Rightarrow \sigma_n &= \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix}, & (\downarrow\downarrow) \Rightarrow \sigma_n &= \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix}.
\end{aligned}$$

Then, the spin Hamiltonian (1) is written in terms of the block-spins as

$$H = \sum_{n,m} \sigma_n^T \cdot \mathbf{J}_{xy} \cdot \sigma_m + \sum_n \sigma_n^T \cdot \mathbf{J}_z \cdot \sigma_{n+1} + (\mathbf{S} + \mathbf{E}) \cdot \sum_n \sigma_n, \quad (2)$$

in which matrices,  $\mathbf{J}_{xy}, \mathbf{J}_z$  and the vectors,  $\mathbf{S}, \mathbf{E}$  are given by

$$\begin{aligned}
\mathbf{J}_{xy} &= \begin{pmatrix} -2J & 0 & 0 & 2J \\ 0 & -2J & 2J & 0 \\ 0 & 2J & -2J & 0 \\ 2J & 0 & 0 & -2J \end{pmatrix}, \\
\mathbf{J}_z &= \begin{pmatrix} -J_1 - 2J_2 & -J_1 & J_1 & J_1 + 2J_2 \\ J_1 & J_1 - 2J_2 & -J_1 + 2J_2 & -J_1 \\ -J_1 & -J_1 + 2J_2 & J_1 - 2J_2 & J_1 \\ J_1 + 2J_2 & J_1 & -J_1 & -J_1 - 2J_2 \end{pmatrix}, \\
\mathbf{S} &= (-J_1 \quad J_1 \quad J_1 \quad -J_1), \quad \mathbf{E} = (-2E \quad 0 \quad 0 \quad 2E),
\end{aligned}$$

respectively. In Eq. (2), the first term shows the interaction between neighbouring block-spins in the same block-layer, the second one the interaction between neighbouring block-spins in each succeeding block-layers, the traverse vector  $\mathbf{S}$  the interaction within the block-spin and  $\mathbf{E}$  the external field applied to the block-spin.

Here, the intralayer interaction is replaced by a molecular field (MF), in which  $\sigma_m$  in Eq. (2) is replaced by  $\langle \sigma_n \rangle$ , where the bracket  $\langle \dots \rangle$  means the thermal average. Consequently, Eq. (2) is reduced to the compact form as

$$H = \sum_n (\sigma_n^T \cdot \mathbf{A}_n \cdot \sigma_n + \sigma_n^T \cdot \mathbf{J}_z \cdot \sigma_{n+1}), \quad (3)$$

in which the matrix  $A_n$  is given with coordination number  $z$  in the layer by

$$A_n = \begin{pmatrix} a_1 & & \infty \\ & a_2 & \\ & & a_3 \\ \infty & & & a_4 \end{pmatrix}, \quad \begin{aligned} a_1 &= -2zJx_n - J_1 - 2E, \\ a_2 &= -2zJy_n + J_1, \\ a_3 &= 2zJy_n + J_1, \\ a_4 &= 2zJy_n + J_1, \end{aligned}$$

where we use the following notations

$$x_n = \langle \uparrow\uparrow \rangle_n - \langle \downarrow\downarrow \rangle_n \quad y_n = \langle \uparrow\downarrow \rangle_n - \langle \downarrow\uparrow \rangle_n, \quad (4)$$

and  $\langle \uparrow\uparrow \rangle_n$  denotes the probability that the  $n$ th block-spin takes  $(\uparrow\uparrow)$ , and those probabilities satisfy

$$\langle \uparrow\uparrow \rangle_n + \langle \downarrow\downarrow \rangle_n + \langle \uparrow\downarrow \rangle_n + \langle \downarrow\uparrow \rangle_n = 1.$$

The parameter  $x_n$  is nothing but a order parameters of the  $n$ -th block-spin (that is,  $n$ th block-layer), and determined self-consistently together with  $y_n$  from the free energy mentioned in the below.

By introducing the MF within the layer, the system is reduced to the block-spin chain in the direction of layer normal ( $z$ -axis) with nearest neighbour interaction. Thermodynamics of this system can be calculated by the transfer matrix method with parameters,  $x_n$  and  $y_n$ .

Assume that the order is periodic with period  $p$  in the  $z$ -direction. Then, a transfer matrix  $M_p$  is defined by

$$M_p = LK_1 LK_2 \dots LK_p \quad (5)$$

where

$$(K_n)_{\alpha,\gamma} = \exp\{-\beta(A_n)_{\alpha,\gamma}\} \quad (L)_{\alpha,\gamma} = \exp\{-\beta(J_z)_{\alpha,\gamma}\}$$

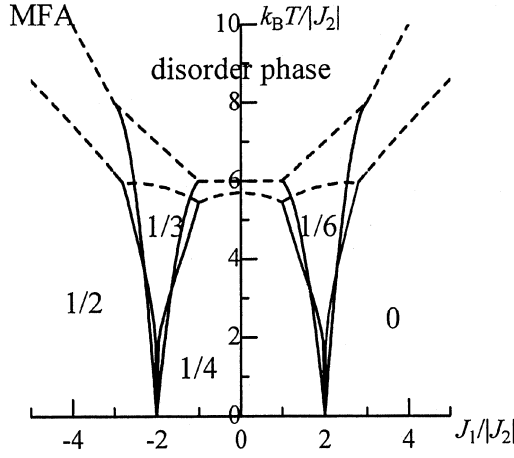
It is noticed that as the matrix  $J_z$  is not symmetrical,  $M_p$  is non-Hermitian and we obtain complex eigenvalues. Because of this fact, the incommensurate structure appears.

## FREE ENERGY AND PHASE DIAGRAM

The free energy  $F_p$  per block-spin is given by

$$F_p = \frac{1}{p} \left\{ -\beta^{-1} \ln(\lambda_p(k_0)) + zJ \sum_{n=1}^p (x_n^2 + y_n^2) \right\}, \quad (6)$$

where  $\lambda_p(k_0)$  is the maximum eigenvalue that is a function of  $x_n$  and  $y_n$ . The second term in Eq. (6) comes from the correction of the MF. The order



**FIGURE 1** Phase diagrams.

parameter  $x_n$  and  $y_n$  are determined by the self-consistent equations;

$$\frac{\partial F_p}{\partial x_n} = 0 \quad \text{and} \quad \frac{\partial F_p}{\partial y_n} = 0 \quad (n = 1, 2, \dots, p)$$

In the ANNNI model countable phases appear forming devil's staircase. However, it is enough to take into account of only several main phases for the purpose of understanding what happens in the system. Here, phase diagram is obtained practically by taking account of five ordered phases with wave number  $q = (1/p) = 0, 1/6, 1/4, 1/3$  and  $1/2$  as shown in Figure 1, where the external field  $E$  is absent and the following unit is used,  $zJ = 4$ .

The phase boundaries marked by MFA shows the one obtained by the ordinary molecular field approximation for ANNNI model. By comparing both phase boundaries, the approximant of the present theory is estimated.

## BLOCK-SPIN CHAIN ALONG LAYER NORMAL

For simplicity, we calculate the correlation functions along layer normal without the molecular field due to the interaction in the same layer, that is,  $x_n = y_n = 0$ . Even in this case without long range order, the incommensurate feature with decaying oscillation is observed in the correlation function.

By inserting  $x_n = y_n = 0$  in Eq. (5), the transfer matrix  $M_p$  is reduced to a form ( $=M$ ) which is independent of  $n$ , and  $M$  is given by

$$\mathbf{M} = \begin{pmatrix} e^{2\beta(J_1+J_2)} & 1 & e^{-2\beta J_1} & e^{-2\beta J_2} \\ 1 & e^{-2\beta(J_1-J_2)} & e^{-2\beta J_2} & e^{2\beta J_1} \\ e^{2\beta J_1} & e^{-2\beta J_2} & e^{-2\beta(J_1-J_2)} & 1 \\ e^{-2\beta J_2} & e^{-2\beta J_1} & 1 & e^{2\beta(J_1+J_2)} \end{pmatrix}.$$

Eigenvalues  $\lambda_k$  ( $k=1\sim 4$ ) of  $\mathbf{M}$  are obtained exactly as

$$\lambda_1 = \xi_+ + \eta_+, \quad \lambda_2 = \xi_+ - \eta_+ \quad \text{and} \quad \lambda_3 = \lambda_4^* = \xi_- + i\eta_-, \quad (7)$$

where

$$\begin{aligned} \xi_{\pm} &= \exp(2\beta J_2) \cosh(2\beta J_1) \pm \exp(-2\beta J_2), \\ \eta_{\pm} &= \frac{1}{2} \sqrt{8\{\cosh(2\beta J_1) \pm 1\} \pm 2 \exp(4\beta J_2) \{\cosh(4\beta J_1) \pm 1\}}. \end{aligned}$$

The maximum eigenvalue is  $\lambda_1$  and  $\lambda_3$  and  $\lambda_4$  are complex conjugate each other.

The thermal average of the block-spin,  $\langle \sigma \rangle$  is given by

$$\langle \sigma \rangle = \begin{pmatrix} \langle \uparrow\uparrow \rangle \\ \langle \uparrow\downarrow \rangle \\ \langle \downarrow\uparrow \rangle \\ \langle \downarrow\downarrow \rangle \end{pmatrix} = \begin{pmatrix} \langle \sigma_1 \rangle \\ \langle \sigma_2 \rangle \\ \langle \sigma_3 \rangle \\ \langle \sigma_4 \rangle \end{pmatrix}.$$

It is noticed that  $\langle \sigma_k \rangle$  ( $k=1\sim 4$ ) are expressed in terms of the unitary matrix  $\mathbf{U}$  which diagonalises the transfer matrix  $\mathbf{M}$  as

$$\langle \sigma_k \rangle = \mathbf{U}_{1,k}^{-1} \mathbf{U}_{k,1}.$$

We define here a group of correlation functions between 0-th block-spin and  $n$ th one,  $\Gamma(n)$  as follows,

$$\Gamma(n) \equiv \begin{pmatrix} \langle \langle \uparrow\uparrow; \uparrow\uparrow \rangle \rangle_n & \langle \langle \uparrow\uparrow; \uparrow\downarrow \rangle \rangle_n & \langle \langle \uparrow\uparrow; \downarrow\uparrow \rangle \rangle_n & \langle \langle \uparrow\uparrow; \downarrow\downarrow \rangle \rangle_n \\ \langle \langle \uparrow\downarrow; \uparrow\uparrow \rangle \rangle_n & \langle \langle \uparrow\downarrow; \uparrow\downarrow \rangle \rangle_n & \langle \langle \uparrow\downarrow; \downarrow\uparrow \rangle \rangle_n & \langle \langle \uparrow\downarrow; \downarrow\downarrow \rangle \rangle_n \\ \langle \langle \downarrow\uparrow; \uparrow\uparrow \rangle \rangle_n & \langle \langle \downarrow\uparrow; \uparrow\downarrow \rangle \rangle_n & \langle \langle \downarrow\uparrow; \downarrow\uparrow \rangle \rangle_n & \langle \langle \downarrow\uparrow; \downarrow\downarrow \rangle \rangle_n \\ \langle \langle \downarrow\downarrow; \uparrow\uparrow \rangle \rangle_n & \langle \langle \downarrow\downarrow; \uparrow\downarrow \rangle \rangle_n & \langle \langle \downarrow\downarrow; \downarrow\uparrow \rangle \rangle_n & \langle \langle \downarrow\downarrow; \downarrow\downarrow \rangle \rangle_n \end{pmatrix}.$$

For example

$$(\Gamma(n))_{1,4} \equiv \langle \langle \uparrow\uparrow; \downarrow\downarrow \rangle \rangle_n = \langle \uparrow\uparrow; \downarrow\downarrow \rangle_n - \langle \uparrow\uparrow \rangle_n \langle \downarrow\downarrow \rangle_n,$$

where  $\langle \uparrow\uparrow; \downarrow\downarrow \rangle_n$  means the joint probability for a couple of states that 0-th block-spin takes  $(\uparrow\uparrow)$  and  $n$ th one  $(\downarrow\downarrow)$ . The element  $\Gamma(n)_{\alpha,\gamma}$  ( $\alpha, \gamma = 1\sim 4$ ) is calculated exactly as;



$$\Gamma(n)_{\alpha,\gamma} = U_{\gamma,1} U_{1,\alpha}^{-1} \sum_{k=2}^4 \left( \frac{\lambda_k}{\lambda_1} \right)^n U_{\alpha,k} U_{k,\gamma}^{-1}. \quad (8)$$

In terms of block-spin correlation functions  $\Gamma(n)$ , we can express the spin-spin correlation functions between 0th and  $R$ th spins,  $\langle\langle\uparrow; \uparrow\rangle\rangle_R$ , which shows the correlation that both 0th and  $R$ th spins take  $\uparrow$ . Here, we obtain

$$\begin{aligned} R=0 \quad \langle\langle\uparrow; \uparrow\rangle\rangle_R &= \langle\uparrow\rangle - \langle\uparrow\rangle^2, \\ R=1 \quad \langle\langle\uparrow; \uparrow\rangle\rangle_R &= \langle\uparrow\uparrow\rangle - \langle\uparrow\rangle^2, \\ R=2n \quad \langle\langle\uparrow; \uparrow\rangle\rangle_R &= \langle\langle\uparrow \circ; \uparrow \circ\rangle\rangle_n \\ &= \sum_{\alpha=1}^2 \sum_{\gamma=1}^2 \Gamma(R/2)_{\alpha,\gamma}, \\ R=2n+1 \quad \langle\langle\uparrow; \uparrow\rangle\rangle_R &= \langle\langle\uparrow \circ; \circ \uparrow\rangle\rangle_n \\ &= \sum_{\alpha=1}^2 \left\{ \Gamma((R-1)/2)_{\alpha,1} + \Gamma((R-1)/2)_{\alpha,3} \right\}. \end{aligned}$$

On the basis of spin-spin correlation function, we can discuss the incommensurate structure along layer normal. The structure (or, modulated structure) is characterized by the factor  $(\lambda_k/\lambda_1)^{R/2}$  in  $\Gamma(R/2)_{\alpha,\gamma}$  in Eq. (8). Let us rewrite  $\lambda_k/\lambda_1$  in the form as,

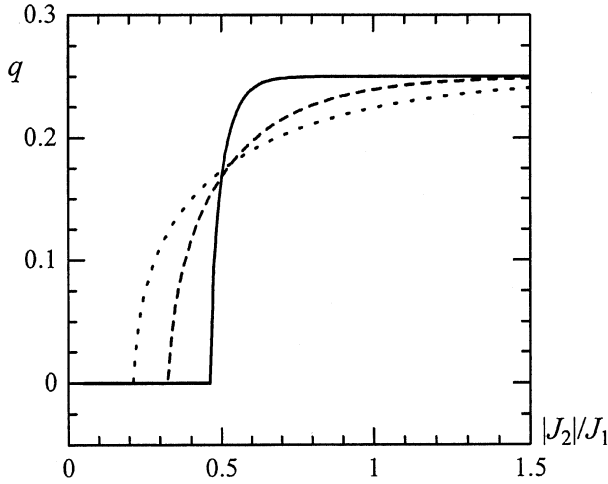
$$\frac{\lambda_k}{\lambda_1} \equiv e^{-2(\gamma_k - 2\pi i q_k)} \quad (k = 2 \sim 4), \quad (9)$$

where  $\gamma_k$  is a damping factor coming from the real part of  $\lambda_k$  and  $q_k$  the wave number coming from the imaginary part. Both parameters depend on the temperature and the ratio of interaction parameters. Substituting Eq. (9) to Eq. (8), we obtain

$$\Gamma(R/2)_{\alpha,\gamma} = \sum_{k=2}^4 e^{-\gamma_k R} \left\{ a'_{k,\alpha,\gamma} \cos(2\pi q_k R) - a''_{k,\alpha,\gamma} \sin(2\pi q_k R) \right\}, \quad (10)$$

where  $a'_{k,\alpha,\gamma}$  and  $a''_{k,\alpha,\gamma}$  mean a real part and a imaginary one of the matrix  $U_{\gamma,1} U_{1,\alpha}^{-1} U_{\alpha,k} U_{k,\gamma}^{-1}$  in Eq. (8), respectively. As we can see in Eq. (7),  $\lambda_1$  and  $\lambda_2$  are real, so that the wave number  $q_2$  is equal to zero. The term of  $k=2$  in Eq. (10) gives purely damping contribution. On the other hand, non-vanishing wave number  $q_3$  ( $=-q_4$ ) comes from the complex value of  $\lambda_3$  ( $=\lambda_4^*$ ), which is truly the wave number  $q$  of the incommensurate structure. Thus, we obtain

$$q \equiv q_3 = \frac{1}{4\pi} \arg\left(\frac{\lambda_3}{\lambda_1}\right). \quad (11)$$



**FIGURE 2** Incommensurate phase.

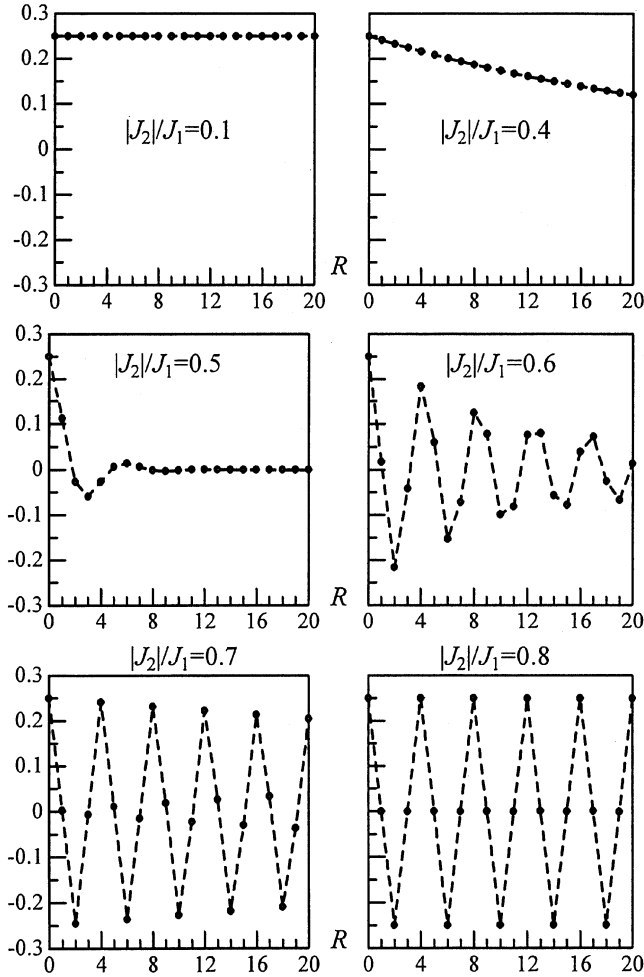
In Figure 2,  $q$  is shown as a function of  $|J_2|/J_1$  for temperature  $T=0.1$  (solid line), 0.5 (broken line) and 1.0 (dotted line).

At the absolute zero temperature, the phase transition between ferroelectric phase ( $q=0$ ) and antiferroelectric one, AF ( $q=1/4$ ) occurs at  $|J_2|/J_1=0.5$ , while no phase transition occurs at finite temperature. However we see the remarkable change of  $q$  even in finite temperature, that is,  $q$  changes from zero continuously at a certain critical value of  $|J_2|/J_1$ , beyond which  $q$  increases up to  $1/4$ . The critical value decreases as the temperature increases.

The spin-spin correlation function  $\langle\langle\uparrow; \uparrow\rangle\rangle_R$  is plotted as a function of “distance”  $R$  for several values of  $|J_2|/J_1$  in Figure 3a ( $T=0.1$ ) and Figure 3b ( $T=0.5$ ). We see that the purely damping feature is observed for  $|J_2|/J_1$  smaller than the critical value, beyond which the damping oscillation appears. It is noticed that the degree of damping (or, an inverse of correlation length) decreases as  $|J_2|/J_1$  increases, and in both limits of zero and infinity of  $|J_2|/J_1$ , the damping can be neglected, even though long range order is absent.

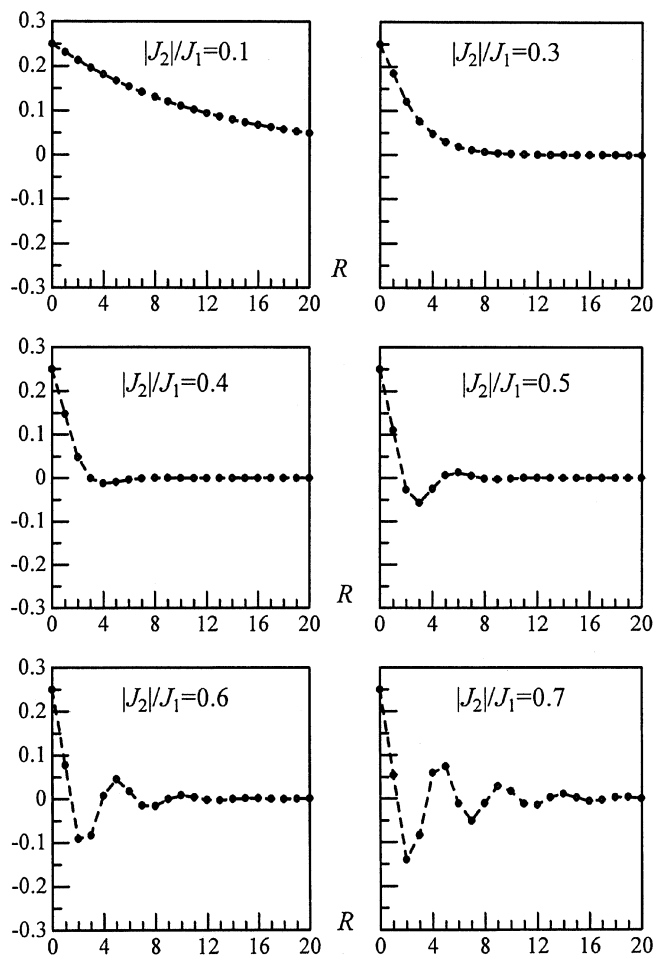
## SUMMARY

In order to clarify the mechanism of successive phase transition occurring at antiferroelectric smectics, the correlation function at the ANNNI model is studied. The block-spin composed of a couple of spins adjacent in the layer normal direction is introduced, by which the second nearest



**FIGURE 3a**  $\langle\langle\uparrow;\uparrow\rangle\rangle_R$  vs.  $R$  at  $T=0.1$ .

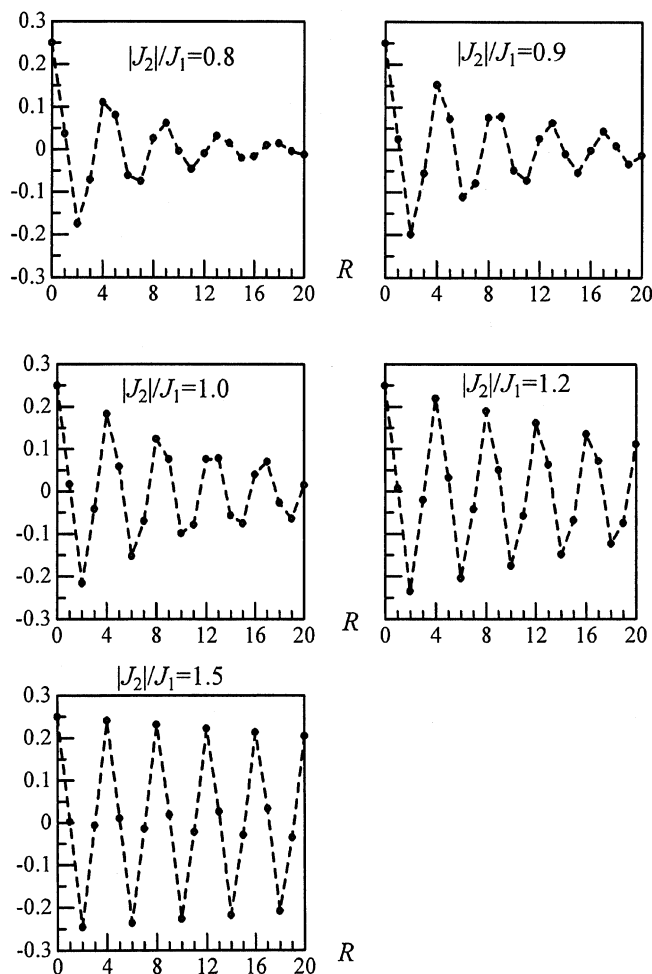
neighbour interaction appearing in ANNNI model is tractable in the method of transfer matrix. Though the practical calculation is carried out at the linear chain where the long range order appears only at absolute zero temperature, incommensurate character due to the frustration is observed in the correlation function with damped oscillation for the separation of spins. This oscillation comes from the non-Hermit property of transfer matrix, and wave number of oscillation changes as the ratio of the interaction parameters changes, which agrees qualitatively with the phase diagram of ANNNI model in 3-dimension. By replacing the interaction within



**FIGURE 3b**  $\langle\langle\uparrow; \uparrow\rangle\rangle_R$  vs.  $R$  at  $T=0.5$ .

the smectic layer by the molecular field, the phase diagram is derived in the block-spin formalism together with the transfer matrix method.

In the previous study of correlation functions at ferro- and antiferroelectric smectic phases, the system is free from frustration. On the other hand in the present study, the correlation function is obtained at the linear chain system with full frustration. As no long range order appears in this system, the present result corresponds qualitatively to the disordered region of 3-dimensional system. In this respect, it is quite interesting that the incommensurability due to the frustration is observed even at the correlation of fluctuating variable. Nevertheless, the study of correlation

**FIGURE 3b** Continued.

function at ordered phase with frustration is an open problem to be solved. Though it is hard to solve for general case, we may obtain the correlations for the restricted phases such as  $\text{SmC}_\gamma^*$  and AF in a near future.

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