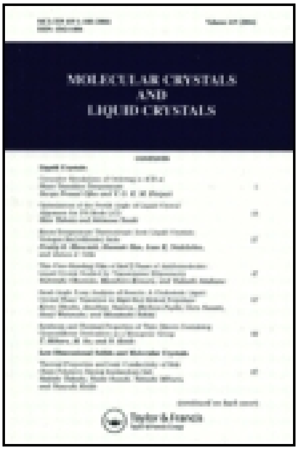

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The phase transitions between ferrielectric phases nearby AF phase and ferroelectric one in the external field are studied by an extended ANNNI model, in which a temperature-dependent first neighbor interaction $J_1 = ST - J_0$ is taken into account. Phase diagram of the external field vs. J_0 exhibits the continuous phase transition from the ferrielectric phase to the ferroelectric one and seems to explain a new type of V-shaped response observed recently at ferrielectric phase in a certain smectic material.

Keywords: ferrielectric smectics; continuous phase transition; ANNNI model

INTRODUCTION

In ferroelectric liquid crystals, it is well-known that between SmC* and SmC$^*_A$, various kinds of phases ; SmC*, FI, AF, FI$_H$, SmC$^*_\gamma$, FI$_L$ and SmC$^*_A$, appear in order as the temperature decreases[1~3]. Although not all of those phases appear in every materials, it is remarkable that several chiral smectic materials undergo such successive phase transitions between SmC* and SmC$^*_A$ phases. The mechanism of such successive phase transitions have been explained by an extended ANNNI model[4~6] (hereafter, refer to E-ANNNI model), in which the third neighbor interaction J_3 is added to ANNNI model[7,8] composed of the first neighbor one J_1 and the second neighbor one J_2 being negative.

In experiments of the ferroelectric or antiferroelectric smectic materials in the electric field, several interesting phenomena have been found[9,10]. In order to clarify those phenomena, we studied the phase diagram of the electric field vs. J_1 by the use of the E-ANNNI model with the electric field[11,12]. From analysis of the phase diagram, we suggested that the phases FI_L and FI_H are of the structures with the wave numbers $q = 2/5$ and $2/7$ in respectively. Furthermore, for the temperature, corresponding to nearby the critical temperature without electric field , it was shown that the transition of second order occurs, especially the continuous transitions from mesophases with $q < 1/4$ (the phase of $q = 1/4$ is AF one) to ferroelectric one occur at the critical temperature. Recently, a new type of V-shaped response of the transmittance of light in the electric field is observed at a ferrielectric phase in a certain binary mixture of smectic material[13,14]. That phase seems to exhibit the continuous transition obtained here.

In this paper, we forcus the modulated phases near AF phase in the electric field and study the continuous transition from these phases to the uniform phase in detail.

E-ANNNI MODEL

One expects that J_1 increases as temperature increases because of the increase of excluded volume of molecules with the temperature. Then, instead of J_1 we use $J_1(T)$ as the first neighbor interaction depending on temperature T, defined by $J_1(T) = ST - J_0$ [4,5], where J_0 must be positive so that SmC_A^* phase is stable in the low temperature region.

In the mean field approximation, the free energy of the E-ANNNI model with the external field E per molecule is given as[11,12]

$$\begin{aligned} F_p &= \frac{1}{p}\sum_{i=1}^{p}\Big[-\frac{zJ}{2}\sigma_i^2 - J_1(T)\sigma_i\sigma_{i+1} - J_2\sigma_i\sigma_{i+2} - J_3\sigma_i\sigma_{i+3} - E\sigma_i \\ &+\frac{k_BT}{2}\{(1+\sigma_i)\ln(1+\sigma_i) + (1-\sigma_i)\ln(1-\sigma_i)\}\Big], \qquad (1) \end{aligned}$$

where p denotes the period of a ordered phase for the axial direction, J the interaction between molecules in a same layer and z the coordination number in a layer. The variable σ_i ($= \sigma_{i+np}$ with integer n) represents the order parameter which is determined by minimizing the free energy F_p with respect to σ_i, that is, by

$$\begin{aligned} E &= -zJ\sigma_i - J_1(T)(\sigma_{i-1} + \sigma_{i+1}) - J_2(\sigma_{i-2} + \sigma_{i+2}) \\ &-J_3(\sigma_{i-3} + \sigma_{i+3}) + \frac{k_BT}{2}\ln\left\{\frac{1+\sigma_i}{1-\sigma_i}\right\} \qquad (i = 1,2,3,\cdots,p).(2) \end{aligned}$$

On the other hand, in the case of continuous transition, transition point is determined by an instability point of the uniform phase. The condition of the instability of the uniform phase is given by[12]

$$\frac{k_BT}{1-\sigma_0^2} = \alpha_p(q_m), \qquad (3)$$

where σ_0 means a order parameter of the uniform phase on the instability surface $E - J_0 - T$. The $\alpha_p(q_m)$ expresses the maximum value of

$$\alpha_p(q) = zJ + 2\{J_1(T)\cos(2\pi q) + J_2\cos(4\pi q) + J_3\cos(6\pi q)\} \qquad (4)$$

with respect to q, where q is the wave number of the modulated phase. The phase with the wave number q_m is just one appearing at the transition point.

PHASE DIAGRAM

Here, all energy parameters are scaled in the unit $|J_2|$, that is $|J_2| = 1$, and $J = 1$, $J_2 = -1$ and $J_3 = 0.3$ are adopted, additionally we choose $z = 6$ and $S = 2$. The phase diagram of T vs.J_0 for $E = 0$ near AF phase

($q = 1/4$) is shown in Fig.1, where phases are indicated by the values of q. The dotted line, which is obtained from eq.(3) with $\sigma_0 = 0$, shows the critical temperature taking a minimum value at $T = 8$. It is noted that the uniform phase SmC* ($q = 0$) appear at high temperature side of disordered phase when $S > 1/2$.

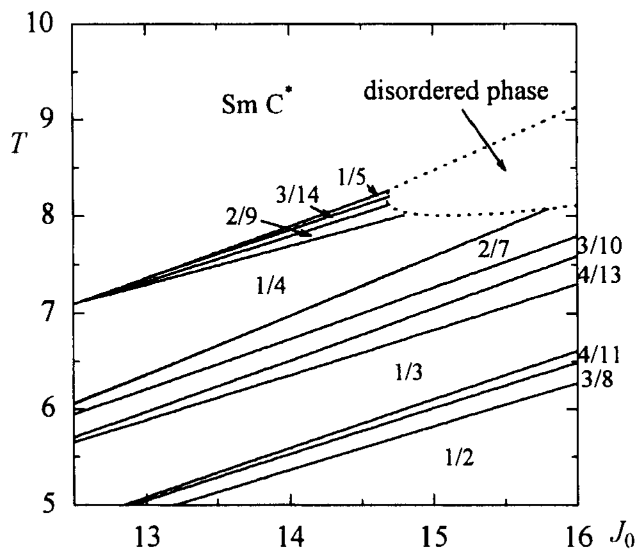

FIGURE 1 : Phase diargam T vs. J_0 at $E = 0$.

The phase diagrams of E vs. J_0 near $q = 1/4$ are shown in Fig.2a at $T = 7$ and in Fig.2b at $T = 8$, respectively. The solid and the broken lines represent the coexisting and the critical lines, respectively, the dotted line the instability line obtained from eq.(3). The phase diagram at $T = 7$ shows that the transition from modulated phase to uniform phase is the first order.

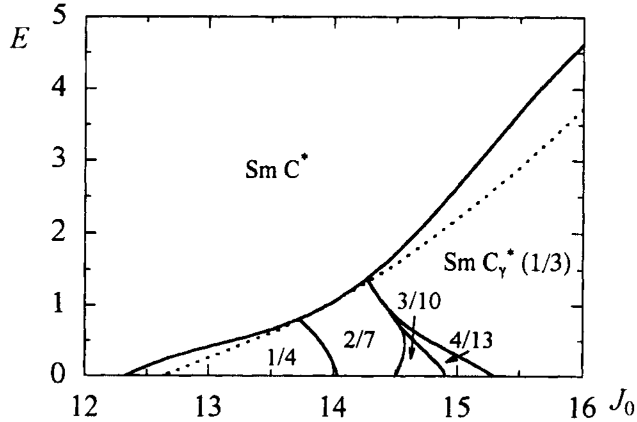

FIGURE 2a : Phase diagram E vs. J_0 for $T = 7$.

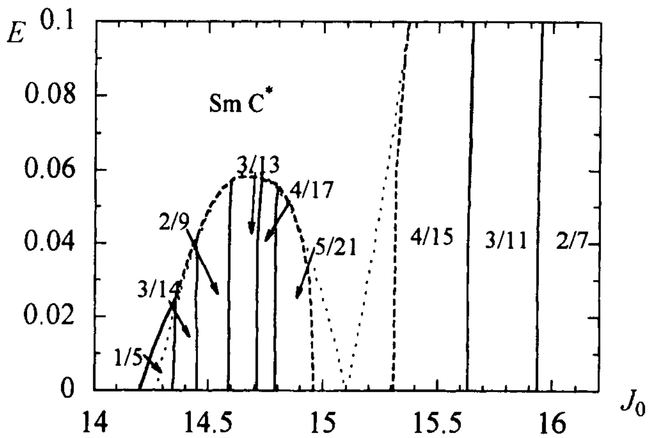

FIGURE 2b : Phase diagram E vs. J_0 for $T = 8$.

On the other hand, at $T = 8$, we have shown previously[12] that the continuous phase transiton from the modulated phase to the uniform one occurs near the phase $q = 1/4$, in which phases with $q = 1/5$, $3/14$, $2/9$, $3/13$ and $2/7$ are taken into account. In order to study the phase diagram between the phases $q = 3/13$ ad $2/7$ precisely, here phases with $q = 4/17$, $5/21$, $4/15$ and $3/11$ are taken into account in addition to those mentioned above. The critical line for the modulated phases with $q < 2/7$ agrees approximately with the instability line except for the phases with $q = 5/21$ and $4/15$, which are adjacent to the phase $q = 1/4$. If this analysis is continued by taking account of more phases between $q = 5/21$ and $4/15$, the critical line seems to agree completely with the instability line in the interval between $q = 3/14$ and $2/7$. Thus, we are led to the conclusion that the second order transition truely occurs and the transition temperature is given by the instability condition of eq.(3) with continuous wave number q_m.

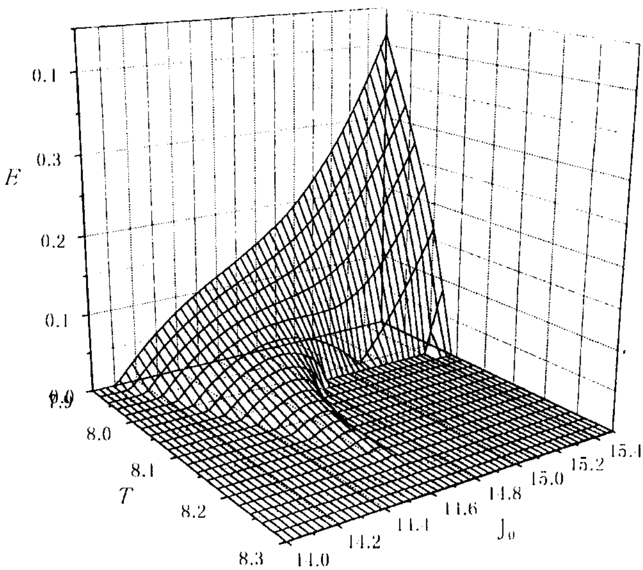

FIGURE 3 : Phase diargam $E(J_0,T)$

Consequently, we conclude that the phase diagram nearby $T = 8$ with $q < 2/7$ is replaced by the critical surface $E(J_0, T)$ obtained from eq.(3), blow of which modulated (incommensurate) phases with continuous wave number q_m become stabe. The phase diagram $E(J_0, T)$ near $T = 8$ is shown in Fig.3, in Fig.4, the modulated phases with the typical wave number q_m are shown. It is noted that the region of the modulated phases with $q < 1/4$ disappear above $T \simeq 8.26$.

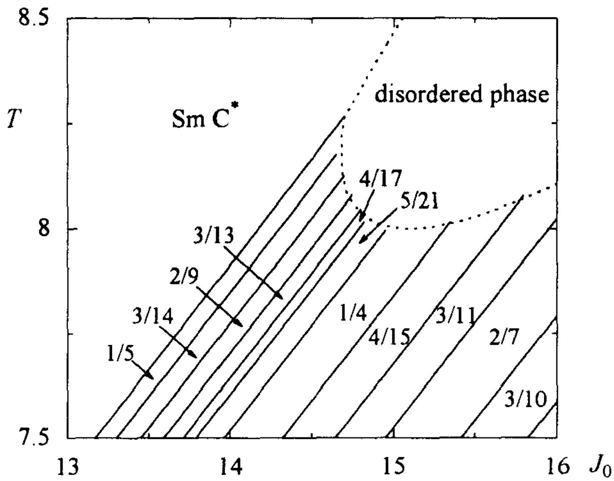

FIGURE 4 : Modulated phases q_m on the critical surface.

SUMMARY

The phase diagram $E(J_0, T)$ near the critical temperature of AF phase is obtained, where the phase transition between the uniform phase and the modulated one is continuous and the modulated phase changes continuously to some extent in the area near the AF phase.

As mentioned above, recently the V-shaped response of the transmittance of light to the external field at the antiferroelectric smectic materials in the planer texture is reported[14] and such response is assumed to come from some kind of randomness of the layers. However the sharpness of the response curve suggests the existence of a long range order and the response without hysteresis should be attributed to the continuous phase transiton. While, at the free standing film of the same material, successive phase transition is observed, in which jumps of the helical pitch are observed in the ferrielectric phase with the change of temperature[14]. The phases of this situation are considered to be far from the critical temperature and located at the region that the devil's staircase appear. On the other hand, in the former case of the homogeneous sample, the order is suppressed by the walls and the sequence of phases is not observed. In the present stage, a structure of the phase named as SmX* characterized by the V-shaped response is not clear, the phase is supposed to correspond to the one near the critical temperature where the phase changes continuously, provided that some long range order exists.

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