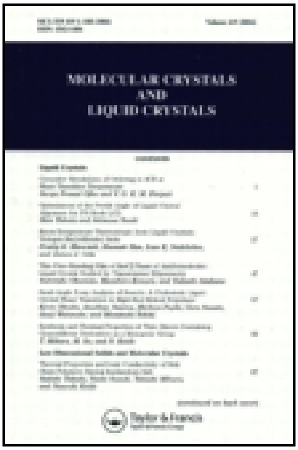

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Satoshi Tanaka [a] & Mamoru Yamashita [b]

[a] Department of Physics, Kinki University, Kowakae 3-4-1, Higashi-Osaka, Osaka, 577-8502, JAPAN

[b] Department of Physics Engineering, Faculty of Engineering, Mie University, Kamihama-cho 1515, Tsu, 514-8507, JAPAN

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On the Hysteresis-Free Response of Antiferroelectric Smectics – the Case of Mesophases with Long Period –

SATOSHI TANAKA[a] and MAMORU YAMASHITA[b]

[a]*Department of Physics, Kinki University, Kowakae 3-4-1, Higashi-Osaka, Osaka, 577-8502, JAPAN and* [b]*Department of Physics Engineering, Faculty of Engineering, Mie University, Kamihama-cho 1515, Tsu 514-8507, JAPAN*

In order to elucidate the hysteresis-free response (V-shaped response) observed at a transmittance of light in antiferroelectric smectic materials, the dynamical responses of the order parameters to an electric field are studied on the basis of an extended ANNNI model with $J_3 < 0$. It is shown that the V-shaped responses appear at mesophases with long period near the ferroelectric phase in low frequency range.

Keywords: Antiferrielectric smectics; dynamical responses; hysteresis-free response; ANNNI model

INTRODUCTION

In some antiferroelectric smectic materials, a V-shaped response without threshold (or hysteresis-free response) has been observed at a transmittance of light to electric field[1,2]. In addition, various types of responses such as normal, abnormal and crossed shapes, have also been found[3~5] in the vicinity of temperature and frequency range where the V-shaped response occurs. These responses are observed at S_X near SC*[3~5]. The origin of the phenomena is anticipated as the result of a weakened interaction due to the competition between ferro- and antiferroelectric behaviors and to frustration[3~5], whereas an interpretation on the mechanism of the V-shaped response is still controversial[5~10].

The present authors have studied a dynamics of metamagnetism with no frustration[8] as well as an extended ANNNI model (EAM) with $J_3 > 0$ (see eq.(1))[9]. Especially in this case, the effect of the polarization on the cell surfaces is taken into account and the responses of order parameters to the external field are studied analytically and numerically. As the results, various types of responses are proved to appear depending on two relaxation times of the order parameter and the surface polarization, where the V-shaped response comes out at the low frequency range in the mesophases near SC* and a quasi V-shaped one also appear at a critical frequency at which the effect of the two relaxation times compensate to each other.

Recently Takeuchi *et al.*[10] have stressed an importance of phases with long period near SC* and obtained the phase diagrams for the various values of J_3 (including the negative values) in the absence of the external field on the basis of EAM.

In this context, we obtain here the phase diagram in the presence of the external field in the case of $J_3 < 0$, and discuss the dynamics of the order parameters. Alike the case with $J_3 > 0$, the sequence of response shape is derived, while in the phase with the wave number $q = 1/10$ a shape without darkest level is observed.

PHASE DIAGRAMS

In the mean field approximation, the free energy per molecule for the EAM can be written as[11]

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

where J denotes the interaction parameter in a same layer, J_1, J_2 and J_3 those between the first, second and third nearest neighboring layers, respectively and E the electric field. The p means the period for the axial direction of an ordered structure and z the coordination number in a layer. The variable σ_i $(= \sigma_{i+np}$ with the integer $n)$

represents an order parameter determined from

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

Let us study the phase diagrams in $J_3 < 0$, where $J/|J_2| = 1$, $J_2/|J_2| = -1$ and $J_3/|J_2| = -0.3$ and $z = 6$ are adopted, in addition J_1, T and E are also scaled in the unit $|J_2|$.

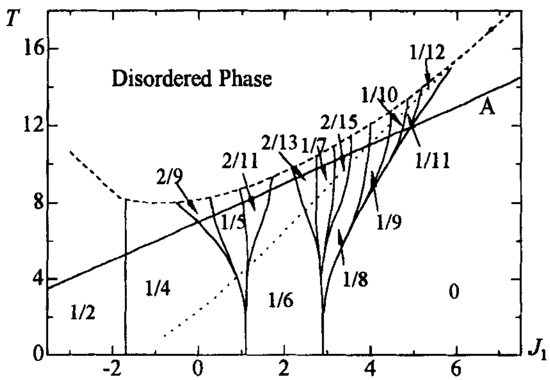


FIGURE 1

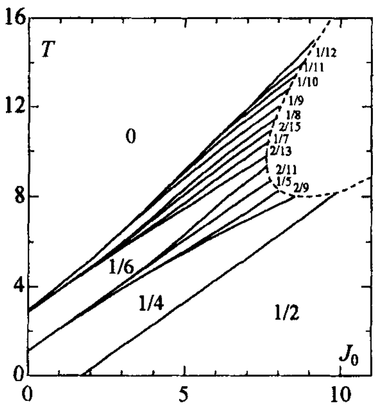


FIGURE 2

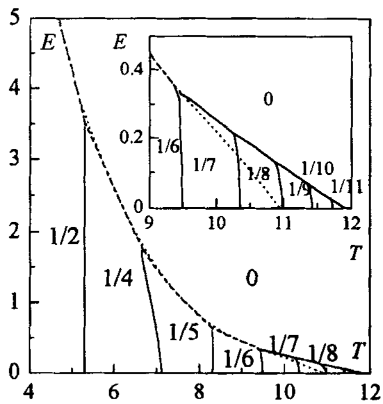


FIGURE 3

The Figure 1 illustrates the phase diagram; T vs. J_1 at $E = 0$, where the modified phases are indicated by the wave number q. The solid lines show the coexisting curves, the broken one the second order transition line and the dotted one the instability of uniform phase. The modulated phases with long period (or small q) appear in the considerably wide range near SC* phase ($q = 0$), compared with the case of $J_3 > 0$[12]. The parameter J_1 depends generally on the temperature T as $J_1 = ST - J_0$, where S is the packing entropy and J_0 the bare interaction. The phase diagram; T vs. J_0 at $E = 0$ is illustrated in Figure 2. The A-line on Figure 1 shows the case of S=1 and J_0=7. The phase diagram; E vs. T along the A-line is shown in Figure 3, where the field-induced second order transition occurs in the wide range of T ($1/4 \geq q \geq 1/6$), while the field-induced first order one occurs in the higher range of T ($q \leq 1/6$) near SC*. This fact is different from the case of $J_3 > 0$[13~15] because the frustration is enhanced in the case of $J_3 < 0$ compared with $J_3 > 0$.

DYNAMICAL RESPONSES

Next we discuss the dynamical responses of the order parameters to the external field. The master equations of $\sigma_i(t)$ $(i = 1, 2, \cdots, p)$ at a time t are given by[9]

$$\tau_0 \frac{d\sigma_i}{dt} = -\sigma_i + \tanh(\beta < h_i >) + f_i(t) \tag{3}$$

$$h_i = zJ\sigma_i + J_1(\sigma_{i+1} + \sigma_{i-1}) + J_2(\sigma_{i+2} + \sigma_{i-2}) + J_3(\sigma_{i+3} + \sigma_{i-3}) + E(t), \tag{4}$$

where τ_0 is a relaxation time of the order parameters and $f_i(t)$ the random force acting on $\sigma_i(t)$. The $E(t)$ is an effective field including the effect of polarization on the cell surface[16]. For the sinusoidal external field with the frequency ω; $E_{\text{ex}}(t) = E_{\text{e0}} \sin(\omega t)$, the $E(t)$ is given by

$$E(t) = E_0\{\sin(\omega t + \theta_{\text{S}}) - \sin\theta_{\text{S}} \exp(-t/\tau_{\text{S}})\}, \tag{5}$$

where τ_{S} is a relaxation time of the polarization on the cell surface and E_0 and θ_{S} are defined by

$$E_0 = E_{\text{e0}}\sqrt{1 - \frac{2c - c^2}{1 + (\omega\tau_{\text{S}})^2}} \quad \text{and} \quad \tan\theta_{\text{S}} = \frac{c\omega\tau_{\text{S}}}{1 + (\omega\tau_{\text{S}})^2 - c}, \tag{6}$$

where c is the parameter less than unity.

If $\omega\tau_0$ is small enough, then $\sigma_i(t)$ can follow the change of the effective field $E(t)$ instantaneously and the response is characterized by the static property. While, in the case that $\omega\tau_0$ is not so small and E_0 is large enough compared with the critical field E_c (E in Figure 3), $\sigma_i(t)$ becomes the uniform value $\sigma_0(t)$ except for a quarter period in the beginning, which delays from the change of $E(t)$. Consequently, after sufficiently long time it is adequate to assume the following form[9];

$$\sigma_0(t) \approx \sigma_A \sin(\omega t + \theta_S - \theta_0), \tag{7}$$

where θ_0 is determined by the following form;

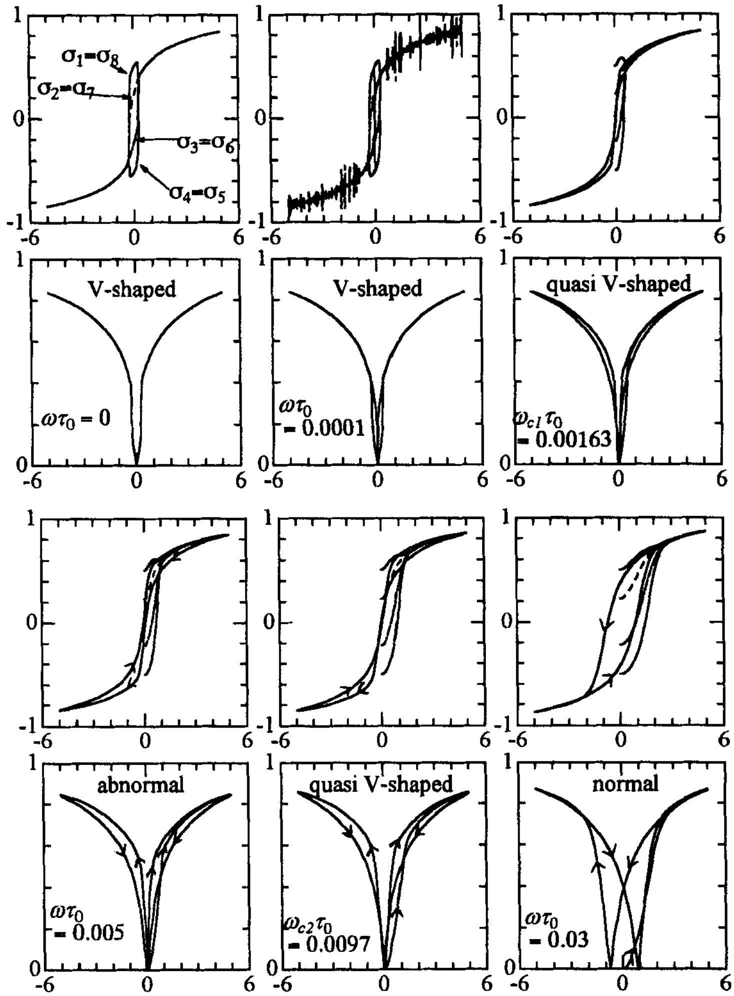
$$\frac{1}{\omega\tau_0}\tan\theta_0 - 1 = \frac{\tanh\{\frac{\beta\hat{J}}{\omega\tau_0}\sin\theta_0 \tanh(\beta E_0 \sin\theta_0)\}}{\cos\theta_0 \tanh(\beta E_0 \sin\theta_0)}, \tag{8}$$

where $\hat{J} = zJ + 2(J_1 + J_2 + J_3)$. At the point $\theta_S = \theta_0$ ($\omega = \omega_c$), $\sigma_0(t)$ viblates in phase with $E_{\rm ex}(t)$, as the result the quasi V-shaped response appears. In the case of $\omega > \omega_c$ (or $\omega < \omega_c$), the response becomes normal one (or abnormal one).

Now we study the dynamical responses of the order parameters σ_i numerically, in which three phases along the A-line (see Figure 1); two mesophases with long period $q = 1/8$ ($T = 10.6$) and $q = 1/10$ ($T = 11.6$) and the AF phase with $q = 1/4$ ($T = 6$) are analyzed for different values of $\omega\tau_0$. The parameters; $\tau_s/\tau_0 = 100$ and $E_{c0} = 6$ are adopted.

The Figure 4 shows the responses of the mesophase with $q = 1/8$ which is expected as S_X, where upper figures illustrate the σ_i vs. $E_{\rm ex}(t)$ and the lower ones the $|\sigma_0|$ corresponding to the transmittance of light. The sharp and V-shaped response with small hysteresis appears at fairy low frequency because of the field-induced first order transition (see Figure 3) and of $\beta\hat{J} = 1$. As $\omega\tau_0$ increases, the structure of the sublattice disappears gradually and finally the normal response appears. Among these responses, two quasi V-shaped responses is observed at $\omega = \omega_{c1}$ and $\omega = \omega_{c2}$.

In the case of the mesophase with $q = 1/10$ near the ferroelectric phase, $|\sigma_0|$ vs. $E_{\rm ex}(t)$ is shown in Figure 5, in which the responses become the normal ones even at low frequency and show the character of the ferroelectric one because $\beta\hat{J}$ ($= 1.06$) is larger than unity. This tendency becomes larger as $\omega\tau_0$ increases. These responses have been already observed by experiment (see Figure 3(e) in ref.[4]).



σ1=σ8
σ2=σ7
σ3=σ6
σ4=σ5
V-shaped
ωτ0 = 0
V-shaped
ωτ0 = 0.0001
quasi V-shaped
ωc1τ0 = 0.00163
abnormal
ωτ0 = 0.005
quasi V-shaped
ωc2τ0 = 0.0097
normal
ωτ0 = 0.03

FIGURE 4

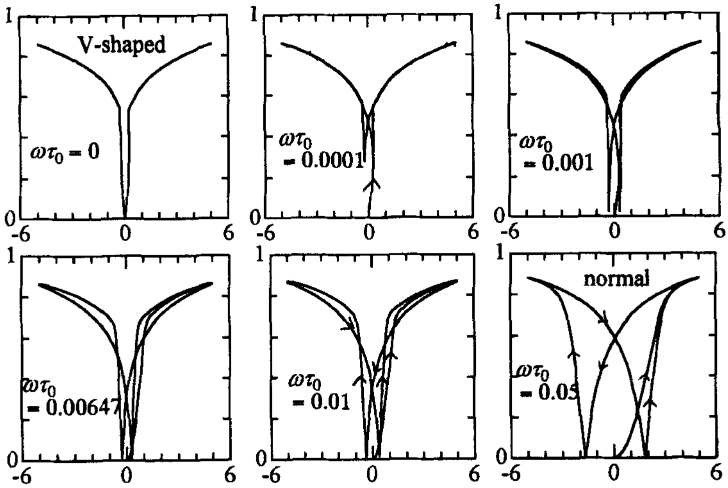


FIGURE 5

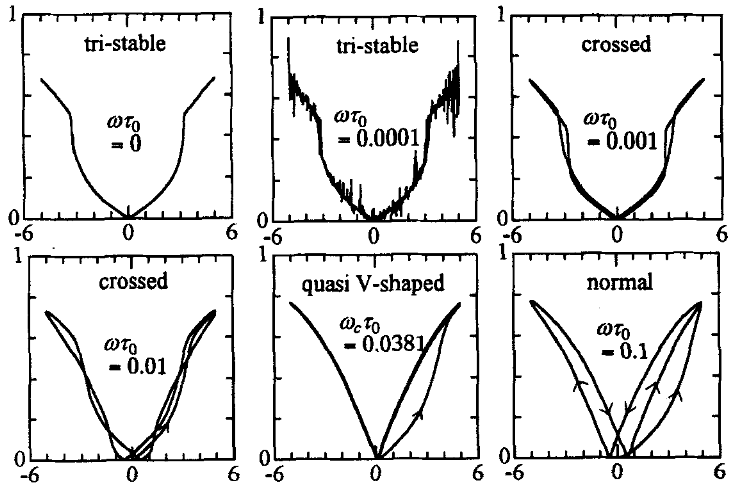


FIGURE 6

The responses of $|\sigma_0|$ in the AF phase with $q = 1/4$ are also shown in Figure 6 in order to compare with the mesophases with long period. The tri-stable responses characterized by the antiferroelectric ones appear at low frequency, especially at $\omega\tau_0 = 0.01$, the structure of sublattice appear only near $E_{\mathrm{ex}}(t) = 0$, then the complex response such as crossed one is exhibited.

SUMMARY

Dynamic response of antiferroelectric smectics as well as static properties are studied mainly at mesophases with long period near SC* on the basis of extended ANNNI model. The change of the response shape with the increasing frequency reveals the similar behavior as the one obtained previously[8,9], while for the phase with $q = 1/10$ the ferroelectric response is reproduced, which corresponds to the one observed experimentally at $\mathrm{S_X}$[4].

For the high frequency at which the sublattice structure is lost after an initial period, the response is described by the uniform order parameter $\sigma_0(t)$ which is governed solely by the effective field $\hat{J}$[9]. Eventually, the response is related closely to the ferroelectric critical line, $\beta\hat{J} = 1$, together with the instability line of the uniform phase shown by dotted line in Fig. 1. The threshold frequency, beyond which the initial sublattice structure is lost, depends on this instability line. For the parameters chosen at the phase with $q = 1/8$ ($\beta\hat{J} = 1$), the uniform phase is unstable, and under this circumstance no sign of hysteresis is observed in Figure 4. On the other hand for those at $q = 1/10$ ($\beta\hat{J} = 1.06$), the uniform phase is stable and two phases with orders, σ_0 and $-\sigma 0$, coexist at the vanishing value of the field, while the periodic mesophase is a true equilibrium phase. Thus, the V-shaped response occurs for the case $\beta\hat{J} \leq 1$, and the shape can be interpreted as belong to the Langevin type[3~5], even though we have no ground to conclude that the phase in the absence of the electric field is the ramdomized phase.

In the present study, response of mono-domain is exclusively concerned. The importance of the effect of walls to disturb ferrielectric phase and SC* has been pointed out[3~5]. In this respect, the response of the system with inhomogeneity due to the wall is an interesting problem to be challenged.

References

[1] S. Inui, N. Iimura, T. Suzuki, H. Iwane, K. Miyachi, Y. Takanishi and A. Fukuda: *J. Mater. Chem.* **6**, (1996) 671.

[2] A. Fukuda and T. Matsumoto: Proc. 4th Int. Display Workshops. (The Institute of Television Engineers of Japan and The Society for Information Display, Tokyo, 1997) p. 355.

[3] S.S. Seomun, T. Gouda, Y. Takanishi, K. Ishikawa and H. Takezoe: *Liq. Cryst.* **26** (1999) 151.

[4] S.S. Seomun, Y. Takanishi, K. Ishikawa, H. Takezoe and A. Fukuda: *Jpn. J. Appl. Phys.* **38** (1999) 3586.

[5] T. Matsumoto, A. Fukuda, M. Johno, Y. Motoyama, T. Yui, S.S. Seomun and M. Yamashita: *J. Mater. Chem.* **9** (1999) 2051.

[6] P. Rudquist, J.P.F. Lagerwall, M. Buivydas, F. Gouda, S.T. Lagerwall, N.A. Clark, J.E. Maclennan, R. Shao, D.A. Coleman, S. Bardon, T. Bellini, D.R. Link, G. Natale, M.A. Glaser, D.M. Walda, M.D. Wand and X.-H. Chen: *J. Mater. Chem.* **9**, (1999) 1257.

[7] B. Park, M. Nakata, S.S. Seomun, Y. Takanishi, K. Ishikawa and H. Takezoe: *Phys. Rev. E* **59**, (1999) R3815.

[8] S. Tanaka and M. Yamashita: to be published in *Mol. Cryst. Liq. Cryst.*.

[9] S. Tanaka and M. Yamashita: *J. Phys. Soc. Jpn.* **69** (2000) No. 7.

[10] M. Takeuchi, K. Chao, T. Ando, T. Matsumoto, A. Fukuda and M. Yamashita: to be publised in to *Ferroelectrics*.

[11] M. Yamashita and S. Miyazima: *Ferroelectrics* **148** (1993) 1.

[12] M. Yamashita: *Ferroelectrics* **181** (1996) 201.

[13] M. Yamashita and S. Tanaka: *Jpn. J. Appl. Phys.* **37** (1998) L528.

[14] S. Tanaka and M. Yamashita: *Jpn. J. Appl. Phys.* **38** (1999) L139.

[15] S. Tanaka and M. Yamashita: *Mol. Cryst. Liq. Cryst.* **328** (1999) 39.

[16] A.D.L. Chandani, C. Yumin, S.S. Seomun, Y. Takanishi, K. Ishikawa and H. Takezoe: *Liq. Cryst.* **26** (1999) 167.