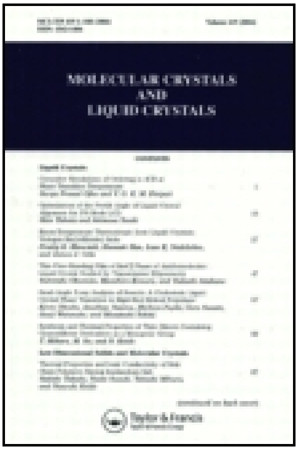

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Metamagnetism and V-Shaped Response in Anti-Ferroelectric Smectic Liquid Crystals

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In order to find a clue to elucidation of the thresholdless response or V-shaped response observed in antiferroelectric smectic materials, an antiferroelectric phase of the Ising model with layered structure, which is known as metamagnetism, is studied statically and dynamically under an external field. It is shown that the phase exhibiting a continuous change driven by the field is not necessarily a disordered phase, but such phenomenon occurs in the phase with long range order.

Keywords: antiferroelectric smectics; V-shaped response; metamagnetism

INTRODUCTION

In ferroeletric and antiferroelectric liquid crystal, the bistable or the tristable switching behaviors are observed in a response of a transmittance of light against an external field[1], which correspond to a first order transition. Recently a V-shaped response with thresfoldless field or no hysteresis has been observed in some antiferroelectric smectic materials[2,3], showing nothing but the second order phase transition. So far, several interpretations about this phenomenon are proposed, one of which is, e.g., such that in the field-induced transition from the antiferroelectric phase to the ferroelectric one, the antiferroelectric phase is not a phase with the true long range interlayer ordering but a quasi-randomized one with sufficiently weak interlayer correlation[3–6] which should be described in terms of the two-dimensional Langevin function. Anyway, the weakened interlayer interaction in cooperation with frustration between interactions is attributed to this phenomenon[7].

On the other hand, We discussed the successive phase transitions in the antiferroelectric smectic liquid crystals by extended ANNNI model with third neighbor interaction[8-10], and it is clarified that the second order transition driven by the external field occurs from some ferrielectic phases to uniform one, suggesting a possibility of the V-shaped response[11-13].

In this note we discuss the field-induced transition without the frustration, i.e., the metamagnetism, to study the mechanism of the V-shaped response phenomenon by using the dynamical mean field theory, because the metamagnetism is the typical model, and here the case of the weak interlayer interaction is focussed. It is shown that the V-shaped response occurs at the antiferroelectric one with true long range order, in which the field-induced second order transition occurs.

MODEL AND PHASE DIAGRAMS

The antiferroelectric smectics can be described in terms of the antiferromagnetic Ising model with layered structure after the case of metamagnetism. The Hamiltonia is given by

$$H = -J \sum_{<i,j>} s_i s_j - J_1 \sum_i s_i s_{i+1} - \mu E \sum_i s_i \tag{1}$$

where the spin s_i takes ± 1 representing the tilt direction of i-th molecular long axis in case of the antiferroelectric smectics, the first summation is taken all over the nearest neighbouring pairs in the same layer and the second one is for the pairs among the succeeding layers and μ and E denote the dipole moment and the external field, respectively. The energy parameter J_1 changes the sign due to the competition between electrostatic and steric interactions in the successive phase transitions. In this note J_1 is chosen to be negative exclusively.

In the mean field approximation, the thermodynamical potential Φ per molecule is expressed as

$$\begin{aligned}\frac{\Phi}{zJ} &= \frac{1}{2}\Bigg[-\frac{1}{2}(\sigma_1^2+\sigma_2^2)+\hat{J}_1\sigma_1\sigma_2-\hat{E}(\sigma_1+\sigma_2)\\ &+\frac{\hat{T}}{2}\{(1+\sigma_1)\ln(1+\sigma_1)+(1-\sigma_1)\ln(1-\sigma_1)\\ &+(1+\sigma_2)\ln(1+\sigma_2)+(1-\sigma_2)\ln(1-\sigma_2)\}\Bigg], \qquad (2)\end{aligned}$$

where $\hat{T}$, $\hat{J}_1$ and $\hat{E}$ are the dimensionless parameters defined as follows:

$$\hat{T}=\frac{k_BT}{zJ},\quad \hat{J}_1=\frac{z_1|J_1|}{zJ}\quad\text{and}\quad \hat{E}=\frac{\mu E}{zJ}. \qquad (3)$$

The z is the coordination number in the same sublattice and z_1 the one between the different sublattice. The thermal averages σ_1 $(=< s_i >)$ and σ_2 $(=< s_{i+1} >)$ are the sublattice moments, determined by

$$\begin{aligned}\sigma_1 &= \tanh\Big\{\frac{1}{\hat{T}}(\sigma_1-\hat{J}_1\sigma_2+\hat{E})\Big\}, \qquad (4)\\ \sigma_2 &= \tanh\Big\{\frac{1}{\hat{T}}(\sigma_2-\hat{J}_1\sigma_1+\hat{E})\Big\}. \qquad (5)\end{aligned}$$

The FIGURE 1-a shows the phase diagram of $\hat{T}$ vs. $\hat{J}_1$, where $\hat{T}_N$ and $\hat{T}_{tc}$ denote the Neel temperature and the tricritical temperature and are given by[14]

$$\hat{T}_N=1+\hat{J}_1,\qquad \hat{T}_{tc}=(1-\frac{\hat{J}_1}{3})\hat{T}_N. \qquad (6)$$

The FIGURE 1-b shows the phase diagrams of $\hat{E}$ vs. $\hat{T}$ for several values of $\hat{J}_1$, where the solid lines represent coexisting lines, that is, the field-induced first order transition from the antiferroelectric phase to the unform one and the broken lines the second order transition lines on which the critical field $\hat{E}_c$ is expressed by

$$\hat{E}_c=-(2-\hat{T}_N)\sqrt{1-\frac{\hat{T}}{\hat{T}_N}}+\frac{\hat{T}}{2}\ln\left[\frac{1+\sqrt{1-\frac{\hat{T}}{\hat{T}_N}}}{1-\sqrt{1-\frac{\hat{T}}{\hat{T}_N}}}\right]\quad \hat{T}_{tc}<\hat{T}<\hat{T}_N. \qquad (7)$$

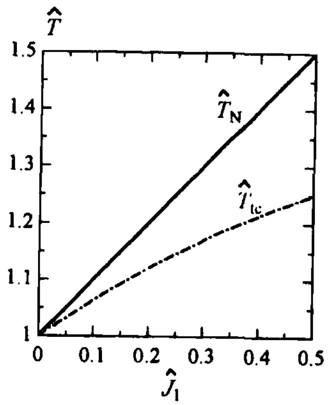

Fig. 1-a : Phase diagram of $\hat{T}$ vs. $\hat{J}_1$.

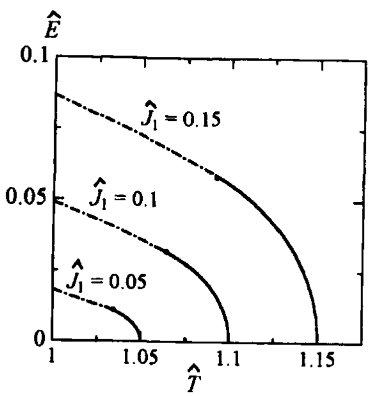

Fig. 1-b : Critical field $\hat{E}_c$ vs. $\hat{T}$. The circlepoint shows the tricritical point.

DYNAMICAL RESPONSE

Now let us consider the dynamics of the sublattice moment σ_1, σ_2 driven by an AC external field. On the basis of dynamical mean field approximation[15], the equations of motion $\sigma_1(t)$ and $\sigma_2(t)$ are represented by

$$\tau \frac{d\sigma_1}{dt} = -\sigma_1 + \tanh\left\{\frac{1}{\hat{T}}(\sigma_1 - \hat{J}_1\sigma_2 + \hat{E}(t))\right\} \tag{8}$$

$$\tau \frac{d\sigma_2}{dt} = -\sigma_2 + \tanh\left\{\frac{1}{\hat{T}}(\sigma_2 - \hat{J}_1\sigma_1 + \hat{E}(t))\right\}, \tag{9}$$

where τ is a relaxation time related to the spin-spin correlation and hereafter we assume it to be constant for simplicity. The $\hat{E}(t)$ is the effective field acting on the moment and is assumed to be given as

$$\hat{E}(t) = \hat{E}_{ex}(t) - \hat{E}_s(t), \tag{10}$$

where $\hat{E}_{ex}(t)$ is the external field applied to the cell and $\hat{E}_s(t)$ the field due to the porlarization on cell surface induced by the external field. The importance of $\hat{E}_s(t)$ has been reported from view of experiment[16,17]. Here, we assume $\hat{E}_s(t)$ to be determined by the following phenomenological equation;

$$\frac{d\hat{E}_s(t)}{dt} + \gamma(\hat{E}_s(t) - c\hat{E}_{ex}(t)) = 0 \tag{11}$$

where γ is a damping parameter of the polarization on the surface and c is a parameter less than unity. For the external field given by

$$\hat{E}_{ex}(t) = \hat{E}_0 \sin(\omega t), \tag{12}$$

$\hat{E}_s(t)$ and $\hat{E}(t)$ are easily obtained as follows;

$$\hat{E}_s(t) = \frac{c\hat{E}_0}{\sqrt{1+\tan^2\theta}} \sin(\omega t - \theta) \tag{13}$$

and

$$\hat{E}(t) = \hat{E}_0(t)\sqrt{1 - \frac{2c - c^2}{1+\tan^2\theta}} \sin(\omega t + \delta) \tag{14}$$

where

$$\tan\theta = \frac{\omega}{\gamma}, \qquad \tan\delta = \frac{c\tan\theta}{1+\tan^2\theta - c} \tag{15}$$

$\hat{E}(t)$, $\hat{E}_s(t)$ and $\hat{E}_{ex}(t)$ are illustrated in FIGURE 2. By the use of eq.(14) as the effective field, the eqs.(8) and (9) can be solved numerically.

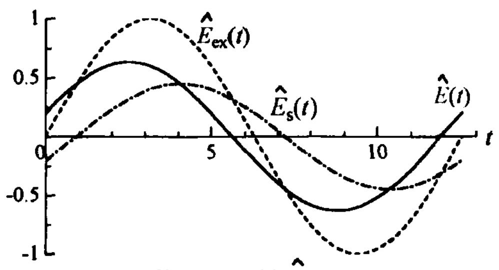

Fig.2 : Effective Field $\hat{E}(t)$. $\omega = 0.5$, $\gamma = 1$, $c = 0.5$.

Let us focus the response of the averaged moment $\sigma(t) = \{\sigma_1(t) + \sigma_2(t)\}/2$ against the external field. By the reason that the transmittance of light I is proportional to $\sin^2\Theta$, where Θ denotes an apparent tilt angle, we assume I to be proportional to σ^2.

At a first step, we consider the case of $\hat{J}_1 = 0.05$, and $\hat{T} = 1.04$, in which the field-induced second order (or continuous) transition occurs from the antiferroelectric phase to the uniform one at $\hat{E} = \hat{E}_c$ as is shown in FIGURE 1-b. Hereafter, we adopt $\gamma = 1$ and $c = 0.1$.

FIGURES 3-a and 3-b show the retardation (or advance) of $\sigma(t)$ against $\hat{E}_{ex}(t)$ at $\tau = 0.004$. At $\omega = 10$, the phase of σ is advanced against $\hat{E}_{ex}(t)$, so that the response of σ^2 against $\hat{E}_{ex}(t)$ becomes a normal one. While, for $\omega = 1$, the phase of σ^2 is retarded against $\hat{E}_{ex}(t)$, as the result abnormal response appears. These types of response have been confirmed with respect to a external field with triangular wave[16,17].

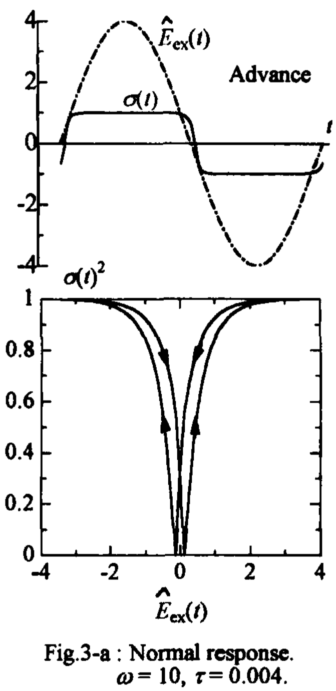

Fig.3-a : Normal response. $\omega = 10$, $\tau = 0.004$.

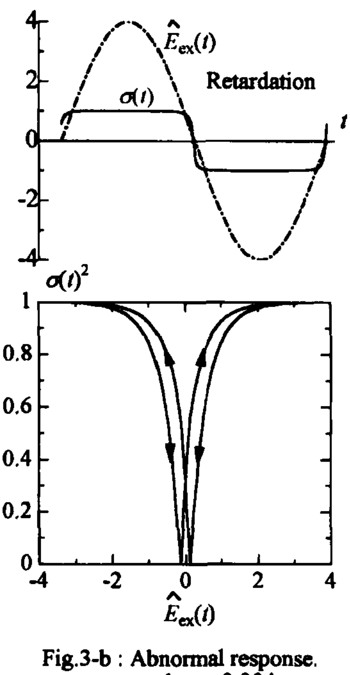

Fig.3-b : Abnormal response. $\omega = 1$, $\tau = 0.004$.

In FIGURES 4-a~ 4-c, the responses of σ^2 against the external field $\hat{E}_{ex}(t)$ are illustrated for $\tau = 0.1$, 0.02 and 0. In the case of $\tau = 0.1$, the normal responses appear during $\omega = 0.1 \sim 10$, in which the hystereses becomes large according as ω increases because of a large relaxation time, and the behavior of response at ω =10 shows that the fast change of the external field can not trail the change of the dipole moment. In the case of τ = 0.02, the V-shaped response appears at ω = 0.1 and at ω = 0.5, the abnormal response being approximate V-shaped one, however, at ω = 1, small crossed response, which is middle one between the abnormal response (ω =0.5) and the normal one (ω =10), appears. Finally, in the case of τ =0, in which the dipole moment σ can respond instantaneously to the effective field retarded against the external field, the V-response appears at low or high frequency (ω =0.1, 10) because the phase shift δ is negligible small, and at middle frequency (ω =0.5, 1), the abnormal responses appear.

From these circumstances, it is clear that the type of response is related to ω/γ and τ sensitively even if $\hat{J}_1$ and $\hat{T}$ are fixed.

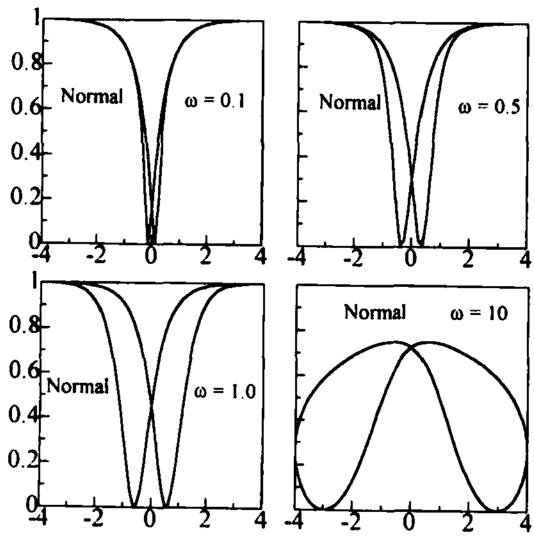

Fig.4-a : τ= 0.1

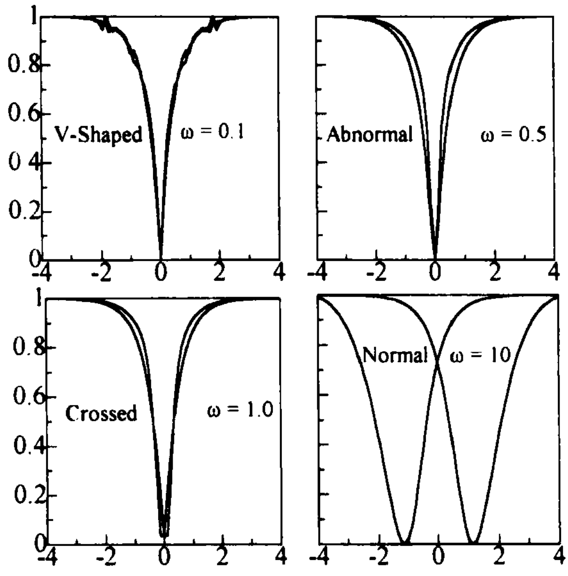

Fig.4-b : $\tau = 0.02$

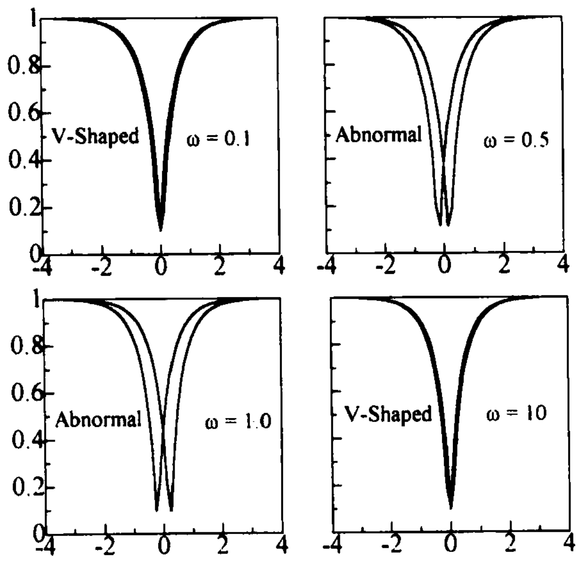

Fig.4-c : $\tau = 0$

SUMMARY AND DISCUSSION

At the phenomena of V-shaped response of antiferroelectric smectics, the weakened interaction between neighboring layer and the frustration are considered to be essential. In this note, we focus our attention to the former, and the metamagnetism is studied as a typical case, in which the possibility of the continuous phase transition is pointed out in the region of small value of $\hat{J}_1$. The dynamics of transition from the antiferroelectric phase to the uniform one driven by the electric field is studied in this region, where the effect of polarization on the cell surface is taken into account. Depending on the ω/γ and τ, various types of the response are obtained, those of which are an abnormal response[16,17] and a crossed one[17] observed already by experiment. The V-shaped response occurs at very low frequency (or static external field) or high frequency for $\tau = 0$.

The relaxation time τ, which is related to the spin-spin correlations, depends in general on both ω and temperature[15], and it is not easy to estimate it. We utilize constant value of τ together with the value $\gamma = 1$. In this respect, the correspondance of the present calculations to the observed ones[16,17] is insufficient, and accordingly the present results are the qualitative ones in nature.

The V-shaped response is observed at various phases. The Sx* phase[16] is one of such notable phases, which is a sort of the ferrielectric phase and considered to occur as a effect of the frustration. Though some features of the V-shaped response are clarified by the present study on the basis of the metamagnetism with dynamiccal treatment, it is not adequate to elucidate the whole concepts of the V-shaped response. In future we will challenge this problem by taking into account the frustration.

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