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REENTRANT MESOMORPHISM INDUCED BY EXTERNAL FIELD

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Abstract The influence of an external orienting field on reentrant phase transitions taking place in smectic A liquid crystals with positive molecular anisotropy of magnetic susceptibility is studied on the basis of a thermodynamic potential of the $X_{1,0}$ type catastrophe. Topological classification of bifurcation sets and phase diagrams of the model are performed. It is predicted the possibility of induction of reentrant phases under the action of the external field. Conditions for experimental observation of this effect are discussed.

INTRODUCTION

To date, a variety of liquid crystal substances has been discovered whose intricate behavior can be explained in terms of coupled order parameters only. Among such systems are those which have layered orientationally ordered structure and demonstrate reentrant phase transitions (PT) of several types^{1,2}. Investigation of the influence of an external magnetic (or electric) field on reentrant mesomorphism can give new important information about its features and nature. It can be suggested that some mesogeneous systems can exhibit reentrant properties under external field action not possessing them in ordinary conditions. Such a phenomenon, being realized in experiment, can be defined as field induced reentrant mesomorphism. This problem was solved earlier neither theoretically nor experimentally and for the first time is discussed in the present paper.

We generalize a broaden version of the phenomenological de Gennes model of smectic A (SmA) mesophase^{3,4} which describes three types namely SmA-nematic (N)-SmA-isotropic liquid (IL), N-SmA-N-IL, SmA-N-SmA-N-IL of reentrant PT in the absence of the external field. It is necessary to note that this model describes corresponding PT sequences without introduction any a priori dependencies of model parameters on thermodynamic values. The bifurcation analysis (see for details References⁵⁻⁹) of the

model is carried out. All of the types of its separatrices are constructed and on this basis a topological classification of phase diagrams (PD) of smectic A with reentrant behavior in the external field is fulfilled. Below only two interesting results are presented. The first one is description of the possibility of induction of reentrant SmA phase and the second one is the transformation of the SmA-N phase transition in reentrant PT sequence from first to second order under the influence of the external orienting field.

FORMALISM

The free energy potential of smectic A in the external field is taken in the form

$$F = \tau_1 Q^2 / 2 - \beta Q^3 / 3 + \gamma Q^4 / 4 + \tau_2 S^2 / 2 + b S^4 / 4 - \chi Q S^2 + \eta Q^2 S^2 / 2 - \mu Q, \qquad (1)$$

where positive values β, γ, b, χ and η are material constants; the value $\mu = \chi a H^2/3$ ($\chi_z > 0$) describes the dimensionless contribution bound up with the action of the external magnetic field H. Parameters $\tau_k = a_k(T - T_c^k)$, ($a_k > 0$, k = 1, 2) characterize deviation of the system temperature T from temperatures T_c^1, T_c^2 of corresponding mean-field phase transitions bound up with the orientational (Q) and translational (S) order parameters in the case when the latters are non-interacting. The equations of state corresponding to the relation $\nabla F(Q,S) = 0$ can be written in the form

$$\gamma Q^{3} - \beta Q^{2} + \tau_{1}Q - \mu - S^{2}(\chi - \eta Q) = 0, \qquad (2)$$

$$S(\tau_2 - 2\chi Q + \eta Q^2 + bS^2) = 0.$$
 (3)

Determinant of the stability matrix of the potential (1) is equal to zero at the condition

$$b(\tau_1 - 2\beta Q + 3\gamma Q^2 + \eta S^2) - 2(\eta Q - \chi)^2 = 0.$$
 (4)

From a mathematical point of view, the expression (1) corresponds to the $X_{1,0}$ type catastrophe¹⁰. Analysis performed for such a multiparametric potential at unidentified control parameter encounters nontrivial calculational difficulties. It follows from Eqs.(2)-(4) that three critical values of the external field are realized

$$\mu_1^{\bullet} = \beta^3 / (27\gamma^2),$$
 (5)

$$\mu_2^* = \left\{ \mu_2^* + \left[\beta^2 / (9\gamma) + 4\chi^2 / (3b\gamma) \right]^{3/2} \right\} / 2. \tag{6}$$

$$\mu_3^{\bullet} = \chi^2(\beta \eta - 2\gamma \chi)/\eta^3, \tag{7}$$

which exert an influence on the important transformations of the SmA PD topology. First of them (5) corresponds to the situation when the N-pN PT becomes the second order one (Wojtowicz-Sheng¹² effect), the values μ_2^{\bullet} and μ_3^{\bullet} describe the appearance of new SmA-N tricritical point and double tricritical point (TCP), respectively.

RESULTS AND DISCUSSION

Cross-sections of SmA phase diagram by coordinates (τ_2, τ_1) plane are presented in Figure 1. The parameter values are chosen as $\beta=9/2$, $\gamma=1$, b=4, $\chi=3$, $\eta=2$ and provide the equality $\mu_1^*=\mu_3^*$. The PT boundaries of first and second order are drawn by solid and dashed lines, respectively. Thin lines are separatrix curves, solid lines correspond to the condition Q>0, points - to the alternative case. The SmA-N line possesses the TCP. This line is convex at $\mu=0$ (Figure 1) and is finished at the triple point (TP) where three phases namely the SmA, N, IL at $\mu=0$ or the SmA, N, pN at $0<\mu<\mu_1^*$ co-exist simultaneously.

Turning on the field leads to transformation of PD (Figure 1b). The stability area of the pN state, which replaces the IL phase, is decreased; the TCP moves up and right along the SmA-N phase boundary. The N-pN PT becomes second order at $\mu=\mu_1^*$ and disappears at $\mu>\mu_1^*$. At $\mu=\mu_2^*$, the SmA-N-pN TP turns into the second TCP on the SmA-N phase boundary. The rise of the field within the interval $\mu_2^*<\mu<\mu_3^*$ leads to the situation when both TCP move towards each other along the corresponding curve. At $\mu=\mu_3^*$ tricritical points coincide and a new peculiarity - the double TCP⁹ appears in PD (Figure 1c). At $\mu>\mu_3^*$, this singularity disappears and the only second order SmA-N phase boundary remains in phase diagram.

According to the Landau PT theory, temperature evolution of the concrete mesogen is described by the straight line directed from the III quadrant to the I one in the (τ_2,τ_1) plane. If this thermodynamic way is close and above the SmA-N phase boundary, then the only SmA-IL (Figure 2a) or the SmA-pN PT takes place in the absence or presence of the external field, respectively. The rise of the field leads, at the beginning, to

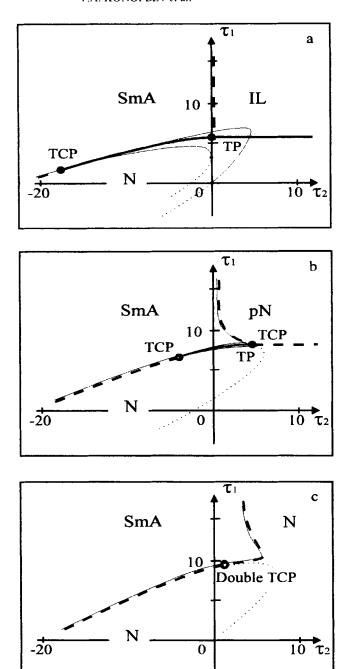


FIGURE 1 Topological types of phase diagrams versus external field: μ =0 (a), μ = μ_1^* (b), μ = μ_2^* (c).

birth of the SmA-N-SmA TP ($0 < \mu < \mu_C$), then to the appearance of the intermediate N phase between two SmA states. So, the SmA-N-SmA_r-pN sequence takes place under the influence of the external field (Figure 2b). In Figure 2 PT orders are shown in parentheses. The following increasing of the field leads to realization of the SmA-N TCP ($\mu = \mu_C$) and replacing of the first order SmA-N PT by the second order one (Figure 2c).

CONCLUSION

Note that the effect of induction of the SmA-N TCP was discussed earlier¹³. Recently Lelidis 14 has represented the experimental evidence of this phenomenon. Our topological analysis of the SmA model (1) shows that this situation takes place under every relationships of the model parameters. In particular, as it is shown in Figures 1,2 the external field can induce the SmA-N TCP (Figure 1b), the SmA_r-N TCP (not shown in Figure 2) and even the new peculiarity - the double TCP (Figure 1c). The phenomenon of induction of the intermediate N-phase under the influence of orienting field was also described in some papers^{15,16}. However, there are no investigations concerning field induced reentrant mesomorphism. We have shown that, if there is the only SmA-IL PT in the absence of the field, its turning on induces the intermediate N state and leads to the SmA-N-SmA_T-pN PT sequence. It can be suggested that the main reason of the transformation of the IL phase to the pN state is the orienting influence of the external field. But the appearance of a new N phase is connected to arising of the additional interaction between mesomorphic particles which is the consequence of the external influence. Thus, the nematic ordering of mesophase becomes more preferable than the layered one in some temperature range.

For experimental evidence of the phenomenon under consideration it is suitable to select mixtures or homologous series of mesogens in which reentrant PT are realized in the absence of the external field. In both cases it is necessary to choose mixture concentration or homolog number scarcely less than reentrant mesomorphism is appeared. In conclusion, we note that values of the external field giving rise to induced reentrance can be significant smaller than the ones of the Wojtowicz-Sheng¹² effect and this permits to

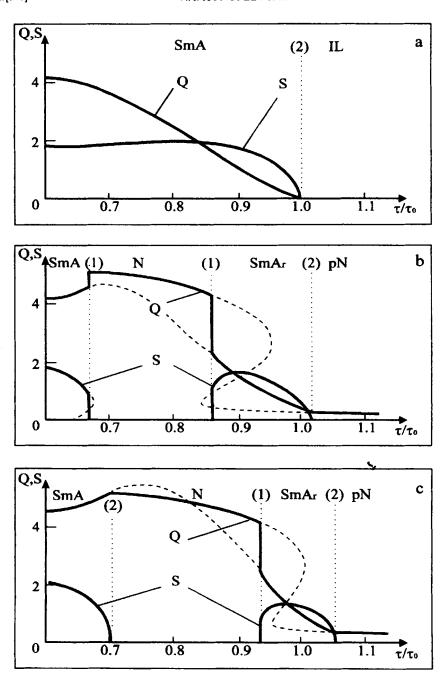


FIGURE 2 Temperature dependencies of orientational (Q) and translational (S) order parameters: μ =0 (a), $0 < \mu < \mu_C$ (b), $\mu > \mu_C$ (c).

hope that reentrant mesomorphism induced by the external field can be realized experimentally.

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