

## Induced smectic phase in a nematic liquid crystal mixture

Thein Kyu,<sup>1,\*</sup> Hao-Wen Chiu,<sup>1</sup> and Tisato Kajiyama<sup>2</sup>

<sup>1</sup>*Institute of Polymer Engineering, The University of Akron, Akron, Ohio 44325*

<sup>2</sup>*Department of Chemical Science and Technology, Faculty of Engineering, Kyushu University, Fukuoka 812, Japan*

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It has been recognized experimentally that a smectic phase can be induced in a nematic mixture via strong mesogenic interaction. To account for an induced smectic phase boundary, we have developed a theoretical model by combining Flory-Huggins (FH) theory for isotropic mixing and Maier-Saupe-McMillan (MSM) theory for ordering of smectic-A phase (Sm-A) in the nematic mixture, although smectic ordering is forbidden in the constituent pure nematic liquid crystals. The predictive capability of the present combined FH-MSM model for the coexistence curves of the nematic mixture and the concomitant induced smectic phase boundary has been critically tested with the reported induced smectic phase diagrams. [S1063-651X(97)06606-3]

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### INTRODUCTION

Within the past decade, theoretical predictions and experimental determinations of phase diagrams involving nematics based polymer dispersed liquid crystals (PDLC) have advanced remarkably [1–11]. One notable progress is the development of the generalized theory [3,5,9] that predicts the phase equilibria of two-nematic mixtures based on the combination of the Flory-Huggins (FH) theory [12,13] for isotropic mixing and the Maier-Saupe (MS) theory [14] for nematic ordering. This combined FH-MS theory has been tested favorably well with the experimental phase diagrams of nematic mixtures containing side-chain liquid crystalline polymers (SCLCP) and low molar mass liquid crystals (LC). Recently, we have modified the FH-MS theory to predict phase diagrams of polymer-smectic liquid crystal mixtures by replacing the MS theory for nematic ordering with the Maier-Saupe-McMillan (MSM) theory for smectic ordering [11]. At that time, we recognized, although it is by no means straightforward, that it is possible to further extend this FH-MSM theory to elucidate the phase diagrams of two-smectic mixtures [11].

While the theoretical works on nematic and smectic phase diagrams have made remarkable progress, there are not many experimental phase diagrams for polymer based smectic LC mixtures [1,2,7]. One of the most intriguing phenomena reported experimentally for the binary nematic LC mixtures is the induced smectic phase [1,7]. In a previous paper [9], we have demonstrated that the strong nematic interaction between the dissimilar mesogens can give rise to a more stable nematic (i.e., induced nematic phase) in the nematic mixtures relative to that in the constituent LCs having the same mesogenic moiety. We further conjectured that such a strong mesogenic interaction could induce a smectic phase although smectic ordering is forbidden in the constituent pure LC. The objective of the present study is to prove the above hypothesis. It should be emphasized that the mesogenic interaction under consideration arises purely from the mesogenic moieties of the low molar LC and of the side-chain liquid crys-

talline polymer. However, specific ion-dipole and/or ionic interactions, even if they exist, are excluded in the calculation. Other transitions such as smectic-smectic, crystal-smectic, crystal-nematic transitions of LCs, and glass transition of the polymers, etc., are not taken into consideration.

In this paper we have introduced a theoretical model to account for the induced smectic phase from a nematic mixture by combining the Flory-Huggins theory [12] for isotropic mixing and the Maier-Saupe-McMillan theory [14,15] for induced smectic ordering in nonsmectic liquid crystal mixtures such as nematic mixtures. The predictive capability of the present FH-MSM model for the phase diagrams of the nematic mixture and the concomitant induced smectic phase boundary has been demonstrated by testing critically with the reported induced smectic phase diagrams.

### THEORETICAL MODEL

The total free energy of mixing for a binary nematic mixture may be expressed in terms of a simple addition of the free energy of mixing of isotropic liquids,  $g^i$ , and the free energy of anisotropic ordering of the liquid crystals undergoing induced smectic phase transition,  $g^s$ , i.e.,  $g = g^i + g^s$ , where  $g$  represents the dimensionless total free energy density of the system. The free energy of isotropic mixing of a binary polymer blend can be generally described by the Flory-Huggins theory [12]; viz.,

$$g^i = \frac{G^i}{nkT} = \frac{\phi_1}{r_1} \ln \phi_1 + \frac{\phi_2}{r_2} \ln \phi_2 + \chi \phi_1 \phi_2, \quad (1)$$

where  $k$  is the Boltzmann constant and  $T$  the absolute temperature.  $r_1$  is the number of sites occupied by one liquid crystal molecule and is equal to unity for a low molar mass liquid crystal, whereas  $r_2$  represents the number of segments or sites occupied by a single polymer chain.  $\phi_1$  and  $\phi_2$  represent the volume fractions of the component 1 and 2, respectively, which may be given by [13]

$$\phi_1 = \frac{n_1 r_1}{n_1 r_1 + n_2 r_2}, \quad \phi_2 = \frac{n_2 r_2}{n_1 r_1 + n_2 r_2}, \quad (2)$$

\*Author to whom correspondence should be addressed.

where  $n_1$  and  $n_2$  are the numbers of liquid crystal and polymer molecules, respectively, and  $n = n_1 r_1 + n_2 r_2$ .  $\chi$  is known as the Flory-Huggins interaction parameter which is generally assumed to be a function of reciprocal absolute temperature, viz.,  $\chi = A + B/T$ , where  $A$  and  $B$  are constants [13].

The free energy density of the anisotropic ordering of the nematic liquid crystal mixtures undergoing induced smectic ordering may be represented in terms of the Maier-Saupe-McMillan mean-field theory [15] as

$$g^s = \frac{G^s}{nkT} = -\Sigma_1 \phi_1 - \Sigma_2 \phi_2 - \frac{1}{2} \nu_{11} s_1^2 \phi_1^2 - \frac{1}{2} \nu_{22} s_2^2 \phi_2^2 - \nu_{12} (s_1 s_2 + \beta \sigma_1 \sigma_2) \phi_1 \phi_2, \quad (3)$$

where  $\Sigma_1$  and  $\Sigma_2$  represent the decrease of entropy due to the alignment of individual LC molecules of component 1 and the mesogenic group of component 2, respectively.  $\beta$  represents a dimensionless cross-interaction strength between two dissimilar mesogens undergoing the induced smectic ordering and thus will be called, hereafter, the induced smectic interaction parameter.  $\nu_{11}$  and  $\nu_{22}$  are the nematic interaction parameters of the pure components whereas  $\nu_{12}$  represents the cross-nematic interaction between the dissimilar mesogens which is defined as [9]

$$\nu_{12} = c \sqrt{\nu_{11} \nu_{22}}. \quad (4)$$

The phase transition from an induced smectic to an induced nematic phase in the nematic mixtures may be described through temperature dependence of the cross-nematic interaction  $\nu_{12}$  which is further coupled with the induced smectic interaction parameter  $\beta$  through the last term of Eq. (3). The physical meaning of  $\beta$  in the nematic mixture is similar to that of the McMillan parameter ( $\alpha$ ) [15] defined originally for the pure smectic, but two dissimilar nematic mesogens are involved in forming a single smectic due to the strong cross-mesogenic interaction. The magnitude of  $\beta$  in the LC mixture is twice the  $\alpha$  value of the pure smectic. As in the case of  $\alpha$  in the McMillan theory [15], the magnitude of the induced smectic interaction parameter  $\beta$  generally determines whether a phase transition occurs from a smectic to an isotropic phase directly or through a nematic phase.

The nematic order parameters  $s_1$  and  $s_2$  and the smectic order parameters  $\sigma_1$  and  $\sigma_2$  are further defined as [14–16]

$$s_j = \frac{1}{2} \langle 3 \cos^2 \theta_j - 1 \rangle, \quad (5)$$

$$\sigma_j = \frac{1}{2} \langle \cos(2\pi z/d_j) (3 \cos^2 \theta_j - 1) \rangle, \quad (6)$$

in which  $\theta_j$  is the angle between the director of a liquid crystal molecule belonging to the component  $j$  ( $j=1$  or  $2$ ) and the reference axis  $z$ .  $d_j$  represents the interlayer distance of component  $j$  in the smectic phase. The angle brackets  $\langle \rangle$  denotes the ensemble average which is defined as

$$\langle \cos(2\pi z/d_j) \cos^2 \theta_j \rangle = \int \int \cos(2\pi z/d_j) \cos^2 \theta_j f(z, \cos \theta_j) dz d\Omega_j, \quad (7)$$

where  $\Omega_j$  denotes the solid angle. The induced nematic order parameter and the induced smectic order parameter are further coupled through an orientation distribution function normalized by a partition function. The normalized orientation distribution function  $f(z, \cos \theta_j)$  is considered to be symmetric around the reference axis which may be expressed by

$$f(z, \cos \theta_j) = \frac{1}{4\pi Z_j} \exp\left(\frac{1}{2} m_{n,j} (3 \cos^2 \theta_j - 1)\right) \times \exp\left(\frac{1}{2} m_{s,j} \cos(2\pi z/d_j) (3 \cos^2 \theta_j - 1)\right), \quad (8)$$

where  $Z_j$  is the partition function defined as

$$Z_j = \int \int \exp\left(\frac{1}{2} m_{n,j} (3 \cos^2 \theta_j - 1)\right) \times \exp\left(\frac{1}{2} m_{s,j} \cos(2\pi z/d_j) (3 \cos^2 \theta_j - 1)\right) dz d\Omega_j, \quad (9)$$

in which  $m_{n,j}$  and  $m_{s,j}$  are dimensionless nematic and smectic mean-field parameters, respectively, which characterize the strength of the respective potential fields [16]. The order parameters  $s_j$  and  $\sigma_j$  can then be related to  $Z_j$  through

$$s_j = \int \int f(z, \cos \theta_j) \frac{1}{2} (3 \cos^2 \theta_j - 1) dz d\Omega_j = \frac{1}{Z_j} \frac{dZ_j}{dm_{n,j}}, \quad (10)$$

$$\sigma_j = \int \int f(z, \cos \theta_j) \frac{1}{2} \cos(2\pi z/d_j) (3 \cos^2 \theta_j - 1) dz d\Omega_j = \frac{1}{Z_j} \frac{dZ_j}{dm_{s,j}}, \quad (11)$$

and the entropy  $\Sigma_j$  can be deduced as

$$\Sigma_j = - \int \int f(z, \cos \theta_j) \ln[4\pi f(z, \cos \theta_j)] dz d\Omega_j = \ln Z_j - m_{n,j} s_j - m_{s,j} \sigma_j. \quad (12)$$

It should be pointed out that the integration over  $z$  in Eqs. (7) and (9)–(12) must be carried out in the limit of  $[0, d_j]$  and then normalized by the layer distance  $d_j$ .

The orientational order parameters  $s_j$  and  $\sigma_j$  may be evaluated by minimizing the free energy of anisotropic ordering with respect to the order parameters, i.e.,

$$\frac{\partial g^s}{\partial s_j} = 0 \quad \text{and} \quad \frac{\partial g^s}{\partial \sigma_j} = 0, \quad (13)$$

which yields the equations

$$\frac{\partial g^s}{\partial s_1} = m_{n,1} - \nu_{11}s_1\phi_1 - \nu_{12}s_2\phi_2 = 0, \quad (14)$$

$$\frac{\partial g^s}{\partial s_2} = m_{n,2} - \nu_{22}s_2\phi_2 - \nu_{12}s_1\phi_1 = 0, \quad (15)$$

$$\frac{\partial g^s}{\partial \sigma_1} = m_{s,1} - \nu_{12}\beta\sigma_2\phi_2 = 0, \quad (16)$$

$$\frac{\partial g^s}{\partial \sigma_2} = m_{s,2} - \nu_{12}\beta\sigma_1\phi_1 = 0. \quad (17)$$

It is obvious from Eqs. (14)–(17) that the orientational order parameters ( $s_j$  and  $\sigma_j$ ) depend on the volume fraction, the smectic cross-interaction strength ( $\beta$ ), and the nematic interaction parameters ( $\nu_{11}$ ,  $\nu_{22}$ , and  $\nu_{12}$ ). Moreover,  $\nu_{11}$  and  $\nu_{22}$  are given to be inversely proportional to temperature [8,9], i.e.,

$$\nu_{11} = 4.541 \frac{T_{\text{NI},1}}{T}, \quad \nu_{22} = 4.541 \frac{T_{\text{NI},2}}{T}, \quad (18)$$

where  $T_{\text{NI},j}$  is the nematic-isotropic (NI) transition temperature of the component  $j$ .

The phase transition from an induced smectic phase in the nematic mixture to an isotropic directly or via an induced nematic phase may be described through the  $\beta$  value. For a given set of  $c$  and  $\beta$  values, the temperature dependence of  $s_j$  and  $\sigma_j$  in the mixtures can be evaluated numerically as a function of composition ( $\phi_1$ ) by means of Eqs. (14)–(18). Once the temperature dependence of the order parameters has been determined, the free energy of anisotropic ordering can be calculated accordingly. The equilibrium coexistence points of the phase diagram can then be computed by using a double tangent method. Regarding the details of phase diagram calculation, interested readers are referred to our previous paper [9].

## RESULTS AND DISCUSSION

The question of whether or not an induced smectic phase occurs in a nematic mixture can be considered via the temperature dependence of the coupling term involving the cross-nematic interaction ( $\nu_{12}$  or  $c$ ) and the cross-smectic interaction parameter  $\beta$ , i.e., the last term in Eq. (3). Physically, the  $c$  parameter represents the relative strength of the cross-nematic interaction of the dissimilar mesogens with respect to that in the same mesogens. It has been shown previously [9] that when  $c < 1$ , the cross-interaction is weak, thus the nematic phase is favored to form in the pure constituents; no smectic phase can be induced. However, when  $c > 1$  where the cross-interaction becomes stronger than that in the same mesogens, the nematic becomes more stable in the mixture relative to that in the pure constituent nematics. This situation gives rise to an induced nematic phase. On the other hand, the induced smectic interaction parameter  $\beta$  in the LC mixture has a similar characteristic of the McMillan smectic interaction parameter  $\alpha$  for the smectic layer ordering in the pure smectic LC, i.e., the magnitude of  $\beta$  deter-

mines whether phase transition occurs from an induced smectic to an isotropic phase directly or through an induced nematic phase. Depending on the magnitude of the coupling between the cross-nematic interaction parameter  $c$  (or  $\nu_{12}$ ) and the cross-smectic interaction parameter  $\beta$  a smectic phase may be induced in a nematic mixture.

Figure 1 shows the temperature dependence of the nematic and smectic orientational order parameters for a hypothetical PDLC system in which the constituent nematic LCs have the nematic-isotropic transition temperatures ( $T_{\text{NI}}$ ) at 60 °C and 50 °C, respectively. The constituent nematic LCs have no smectic phase, but smectic can be induced in their mixtures due to the strong cross-mesogenic interactions, e.g., in the case of  $c = 1.2$  and  $\beta = 1.5$ . The nematic ordering takes place at higher temperatures in the middle composition (50:50) as compared to those of the compositions rich in either component (e.g., the 75:25 ratio), which suggests that the nematic phase in the mixture appears more stable relative to that in the constituent nematic LC. The induced smectic phase occurs in the intermediate compositions as the smectic ordering is forbidden in the constituents. As in the case of the induced nematic, the induced smectic is more stable at the 50:50 ratio as compared to that at the 75:25 composition.

Figure 2 depicts the temperature dependence of the corresponding free energy curves for anisotropic ordering of the nematic mixtures as a function of composition. The concave free energy curve at 60 °C suggests that the nematic phase is more stable in the mixture as compared with that in the pure state. This is exactly what was pointed out previously [9], that a nematic phase can exist above the nematic-isotropic transition temperatures of the constituents due to the strong cross-mesogenic interaction ( $c = 1.2$ ). Upon lowering temperature, a subtle change in the curvature can be noticed at some intermediate compositions, which implies that the two free energy curves, attributable to the induced nematic and smectic ordering, overlap. The broad concave curvature of the free energy curve is analogous to the induced nematic ordering while the narrower one is due to the induced smectic ordering. The anisotropic free energy is lowest at the middle composition (e.g., 50:50) and at a lower temperature (e.g., 0 °C), suggesting that the induced smectic is most stable in the mixture. The contribution of the induced smectic ordering to the total free energy decreases with increasing temperature and virtually diminishes at 60 °C above which only the nematic phase can exist in the mixtures.

Since the phase diagram of the nematic mixture has been dominated by the LC ordering, the interaction parameters associated with the FH theory play no crucial role in the calculations of the induced nematic phase boundaries, but it can exert some minor effects on the induced smectic phase boundaries. Figures 3(a)–3(c) depict the transition temperature versus composition phase diagram as a function of the induced smectic parameter  $\beta$  keeping the  $c$  parameter constant (i.e.,  $c = 1.2$ ). Similar to the McMillan parameter  $\alpha$  [15], liquid crystal phase transition occurs in the smectic-nematic-isotropic sequence when  $\beta < 1.96$ , otherwise it occurs directly from the induced smectic phase to the isotropic phase. Although the choice of the FH parameters may not affect the induced nematic phase boundaries, it is required to provide some reasonable values to reflect the width of the induced smectic phase boundaries, e.g.,  $\chi = 379/T$  and

$r_2/r_1=2.25/1$ . Figure 3(a) shows a temperature versus composition phase diagram encompassing an isotropic ( $I$ ), a  $N+I$  coexistence region, a single phase nematic ( $N$ ),  $N+Sm-A$  coexistence regions, and a single phase smectic  $Sm-A$  region in the descending order of temperature. The  $Sm-A$  and  $N+Sm-A$  coexistence phases exist only in the mixtures since the constituent LC has no smectic phase. Hence it is reasonable to conclude that these smectic phases must be induced by the strong mesogenic interaction similar to that of the induced nematic phase in the  $N+I$  coexistence region which is more stable than that of the constituents.

When the value of  $\beta$  changes from 1.2 to 1.5, the  $N+I$  coexistence region is virtually unchanged, but the  $Sm-A$  and  $N+Sm-A$  coexistence regions shift significantly to higher temperatures, reflecting the greater stability of the induced smectic phase as shown in Fig. 3(b). Concurrently, the convexity of the smectic region gets slightly sharper while the  $N+Sm-A$  region becomes wider. When  $\beta$  is increased to 2.0 (i.e.,  $\beta > 1.96$ ), the smectic-isotropic transition occurs in the intermediate compositions without passing through the nematic phase [Fig. 3(c)]. The induced smectic transition ( $Sm-A$  and  $I+Sm-A$ ) temperatures increase appreciably. When the temperature is below the two peritectic lines (dotted lines), two separate nematic phases form from their isotropic states, i.e.,  $I+Sm-A$  transforms to  $N_1+Sm-A$  and  $N_2+Sm-A$ . In the high compositions of either constituent, the transition occurs from an isotropic to the pure nematic through the  $I+N$  coexistence regions in the descending order of temperature. It is interesting to note that the induced nematic phase in the  $I+N$  regions is more stable than that in the neat constituents.

While the theoretical prediction on the rich variety of phase boundaries in the two-nematic mixtures appears interesting, it is imperative to compare it with the experimental results to validate our hypothesis. Figure 4 depicts the experimental phase diagram for a mixture of a side-chain liquid crystalline polymer (4-polymethoxyphenyl 4'-propyloxy benzoate methyl siloxane) and a low molar mass liquid crystal (4-cyano 4'-pentyloxy biphenyl, 5OCB) [1]. The  $c$  parameter was determined experimentally to be 1.105 from the azeotropic point [9]. Hence  $\beta$  is the only adjustable parameter which can influence the induced smectic boundaries, but not to the  $I+N$  coexistence curves. The FH parameters were set as  $r_2/r_1=9/1$ ;  $\chi=-10+3628/T$ . Beyond our expectation,  $\beta=1.87$  gives a remarkable fit for both the induced smectic as well as the  $I+N$  coexistence boundaries.

In the case of temperature versus composition phase diagram of a mixture of two low molar mass liquid crystals (4-*n*-pentyl-4'-cyanobiphenyl and 4-*n*-butyl-4'-methoxyazoxybenzene) [7], the comparison was made with the calculated coexistence curves of the induced nematic and the induced smectic coexistence phase boundaries (solid lines) using the indicated  $c$  and  $\beta$  values along with the FH parameters of  $r_2/r_1=1/1$  and  $\chi=-5+2332/T$ . As can be seen in Fig. 5, the fit for the  $I+N$  coexistence region appears satisfactory, although the induced smectic boundaries show some departure from the experimental data points. Nevertheless, our calculation captures the trend remarkably well, attesting to the rigor of the predictive capability of our theoretical simulation.

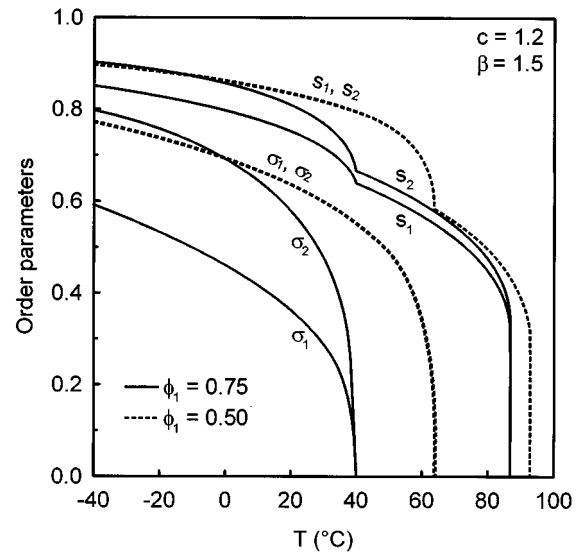


FIG. 1. Temperature dependence of the nematic and smectic order parameters for a mixture comprised of two nematic liquid crystals with  $T_{NI,1}=60$ ;  $T_{NI,2}=50$  °C;  $c=1.2$  and  $\beta=1.5$ , exhibiting an induced smectic phase for two compositions  $\phi_1=0.5$  (dashed lines) and  $\phi_1=0.75$  (solid lines).

## CONCLUSIONS

We have demonstrated that the combined Flory-Huggins–Maier-Saupe-McMillan theory is capable of predicting successfully a rich variety of induced smectic phase diagrams in the binary nematic liquid crystal mixtures using the induced smectic interaction parameter  $\beta$ , which indicates whether the induced smectic mesophase of LC transforms to the isotropic phase directly or through the induced nematic phase. In the former case, the phase diagram consists of the  $I+N$  coexist

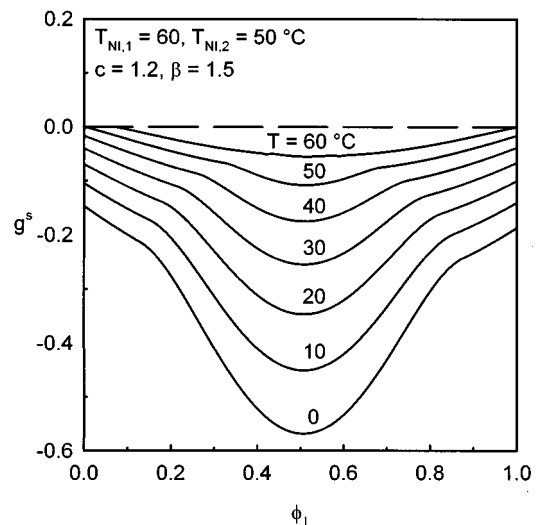


FIG. 2. Variation of combined nematic and induced smectic free energies versus composition at various temperatures for a mixture of two nematic liquid crystals with  $T_{NI,1}=60$  °C;  $T_{NI,2}=50$  °C;  $c=1.2$  and  $\beta=1.5$ .

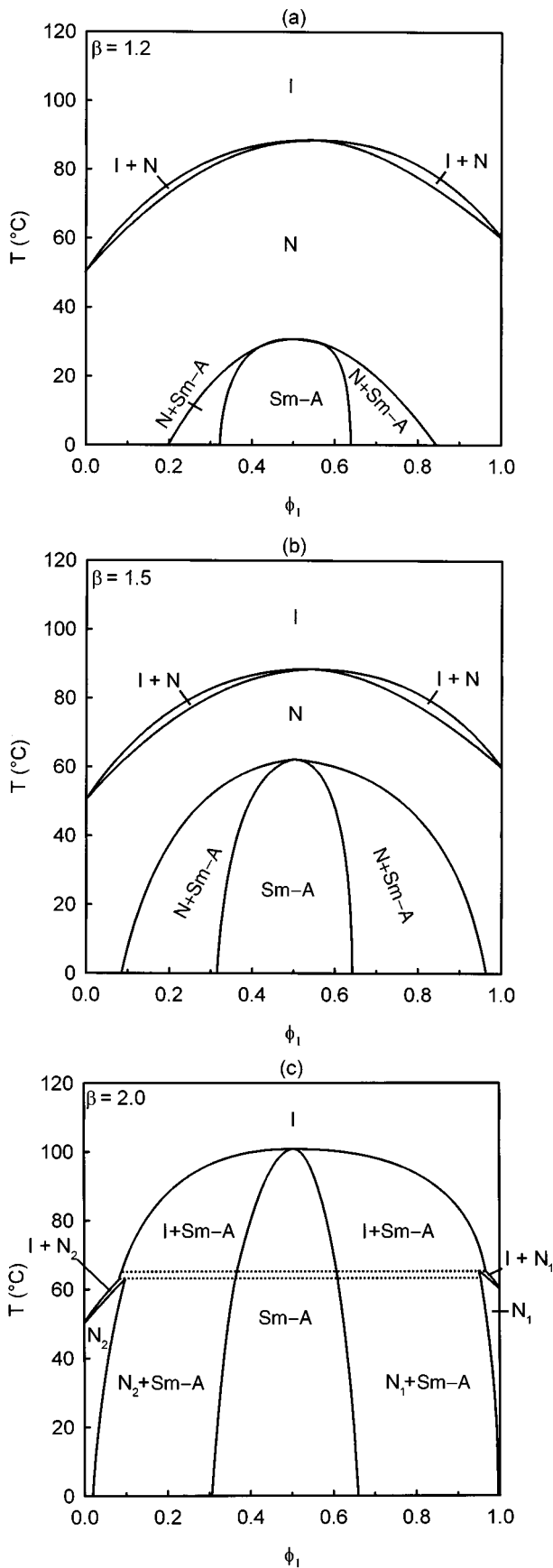


FIG. 3. Temperature versus composition phase diagram showing an induced smectic phase from a mixture of binary nematic liquid crystals having  $T_{NI,1} = 60^\circ\text{C}$ ,  $T_{NI,2} = 50$ ; and  $c = 1.2$  showing the influence of  $\beta$ ; (a) 1.2, (b) 1.5, and (c) 2.0.

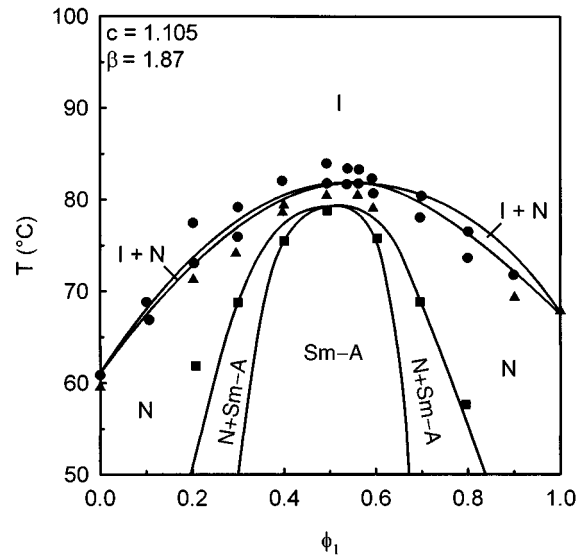


FIG. 4. Temperature versus composition phase diagram for a mixture of side-chain liquid crystalline polymer (4-polymethoxyphenyl 4'-propyloxy benzoate methyl siloxane) and a low molar mass liquid crystal (4-cyano 4'-pentyloxy biphenyl, 5OCB) in comparison with the calculated coexistence curves for induced nematic and induced smectic phase boundaries (solid lines). The filled triangles and circles represent the NI transitions as obtained from differential scanning and calorimetry and optical microscopy, respectively. The filled square symbol represents the induced smectic-nematic transitions as obtained from the optical microscopic investigation (Ref. [1]).

ence region, the pure nematic ( $N$ ), the induced smectic,  $Sm-A$ , the  $I+Sm-A$ , and the  $N+Sm-A$  coexistence regions. In the latter case, the calculated phase diagram consists of the  $I+N$  region, the single phase nematic ( $N$ ), the induced

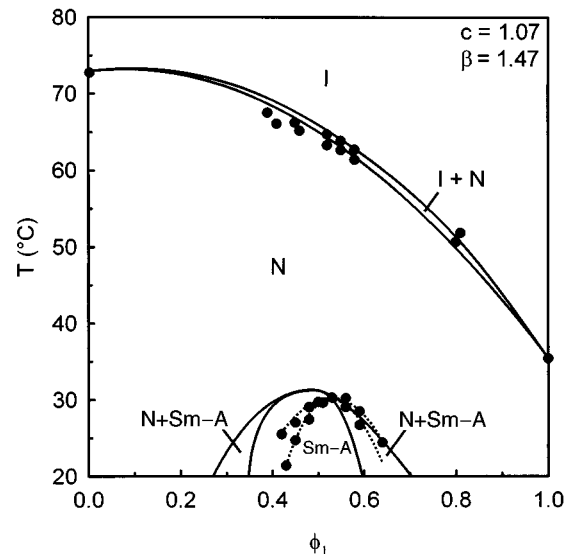


FIG. 5. Temperature versus composition phase diagram for a mixture of two low molar mass liquid crystals (4- $n$ -pentyl-4'-cyanobiphenyl and 4- $n$ -butyl-4'-methoxy azoxybenzene) in comparison with the calculated coexistence curves for induced nematic and induced smectic coexistence phase boundaries (solid lines). The experimental data points and the dotted lines were from Ref. [7].

smectic, and the nematic-smectic ( $N+Sm-A$ ) regions. To the best of our knowledge, the present theoretical calculation is the first in predicting possible coexistence of the induced nematic plus induced smectic ( $N+Sm-A$ ) region and also in verifying the theoretical prediction with the experimental results of the induced smectic phase in the nonsmectic LC mixtures.

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