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INFLUENCE OF FLUCTUATIONS
ON THE ORDER PARAMETERS
IN NEMATIC MIXTURES

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1. Introduction

Mixtures of nematic liquid crystals are widely used in practice, e.g. in LCD's and for this reason they are extensively investigated by experimentalists and theoreticians too. The microscopic theory of nematogens and their mixtures has been established in the last decades to propose the macroscopic physical properties from the molecular data of the constituents. Although it is perhaps impossible to perform first principles calculations of the mixture's properties more and more details of the molecular structure were incorporated into the theory. In the framework of the mean field approximation (MFA) some essential features of the phase transition nematic-isotropic and the behaviour of the order parameters are well understood. Nevertheless there are some discrepancies between the mean field treatment of nematic mixtures and the measured relation between the component order parameters $S_1$ and $S_2$ of the constituents ($S_1$ and $S_2$ are the second rank order parameters $S_i = \langle P_i (\cos(\Theta_i)) \rangle$; $\Theta_i$ is the angle between the director and the principal molecular axis). According to the theory of Palffy-Muhoray et al. [1] this relation should be of the form

$$S_1 = f(z)$$

$$S_2 = f(r z),$$

with $r = (T_2 v_2 / T_1 v_1)^{1/2}$ ($T_1/T_2$ clearing point ratio, $v_1/v_2$ molecular volume ratio of the components). Eqs. (1) are a parameter representation of the function $S_2(S_1)$. To discuss this relation it is not necessary to know the order parameters as a function of temperature. Hence we do not deal with the temperature dependence of $z$. If the anisotropic intermolecular potential is assumed to be
proportional to $P_2(\cos(\theta_{12}))$ ($\theta_{12}$ is the angle between the principal axis of both molecules) the function $f(z)$ is given by

$$f(z) = \frac{d}{dz} \ln \int_0^1 P_2(\xi) \exp[z P_2(\xi)] d\xi.$$  \hspace{1cm} (2)

The mean field result (1),(2) is valid if

$$u_{12} = (u_{11}u_{22})^{1/2}$$ \hspace{1cm} (3)

where the $u_{ij}$ are the interaction constants of the anisotropic forces between the molecules. This geometric mean rule is often used to reduce the number of parameters, the observed violations of (3) are small and not important for our considerations [2,3,4]. $u_{11}$ and $u_{22}$ are connected with the transition temperatures of the pure components, namely

$$u_{11} = 4.54153 v_1 k T_m$$

which is known from simple Maier-Saupe theory [5].

It is a strange fact that relation (1) does not depend on the composition of the mixture! This could be a hint that the HFA fails qualitatively to explain the order parameter relation $S_2(S_1)$. To confirm this idea we consider a fictive mixture with a low concentration of component 2 which has a considerably higher transition temperature than the (host) component 1 ($r$ is large). According to (1) a moderate value of $S_1$ corresponds to a component order parameter $S_2$ tending to 1 since $f(z \to \infty) = 1$. This is a direct consequence of the mean field picture which considers a molecule of component 2 in the effective "field" of the other molecules. From a microscopic point of view the molecule 2 should be affected by the random motion of the host molecules resulting in a decreasing value of its order parameter. Thus, the order parameters of both components differ much less than predicted by the HFA. To confirm this consideration we compare Eqs. (1),(2) with some experimental and computer simulation data (Fig.1). The $^{13}$-NMR measurements where performed by Grande et al. [8] whereas the Monte Carlo results are taken from Hashim et al. [7]. It is obvious that the HFA cannot explain neither the experimental nor the simulation data. The model used in [7] reflects the situation discussed above, e.g. a molecule of component 2 is surrounded by the molecules of component 1 only. $T_2/T_1$ is large ($r = 2$) and the HFA of the model yields exactly the result (1),(2).

2. The Model

In this section a restricted orientation lattice model is introduced which simply permits non mean field calculations. Since we are not interested in the details of the thermodynamics of nematic mixtures but in the influence of fluctuations on the order parameters some essential simplifications were made. We assume that the axially symmetric non-polar rigid molecules are living on a simple cubic 3-dimensional lattice (the practical and conceptional problems of lattice models for liquid crystals have been discussed elsewhere [14]). Further, the allowed orientations of the molecules are
restricted to the \(x\), \(y\) and \(z\) - axis of the lattice. Thus, there are no peculiarities with the symmetries of the lattice and molecular orientations (a similar model was discussed in [8]). Now we consider the intermolecular potential, which is, in general, of the form \(V(\mathbf{r}, \Theta_1, \Theta_2, \varphi_{12})\), where \(\mathbf{r}\) is the intermolecular distance vector in the principal axis frame of molecule 1 and \(\Theta_1\) and \(\Theta_2\) is the angle between the long axis of both molecules. The \(\varphi_{12}\) dependence of \(V\) is ignored in this paper. We mention that this potential is non separable which is of some importance for the existence of a phase transition in the system (see [9,10]). Taking into account only the nearest neighbor interactions one yields four invariant pair configurations which are shown in the scheme below.

\[
\begin{align*}
(a) & \quad (b) & \quad (c) & \quad (d) \\
V = -J & \quad V = 0 & \quad V = 0 & \quad V = 0
\end{align*}
\]

Since we neglected the \(\varphi_{12}\) dependence of the potential configurations (c) and (d) have the same energy which is set to be zero. Now we mention that in nematic the molecules are interacting via anisotropic attractions and hard core repulsions too. Assuming that in case (b) attraction and repulsion forces will compensate, the potential \(V\) is fully determined by the energy of configuration (a) which should be negative to prefer the parallel alignment of the molecules in the nematic phase. Of course, this is for technical convenience, but one can extend the model and take different interaction constants for all configuration (a) - (c).

Now we introduce "spin" variables \(\sigma_i = 1, 2, 3\) for the three possible orientations of a molecule at lattice site \(i\). The values \(\eta_{ij} = 1, 2, 3\) label the direction of the link between the two sites \(i\) and \(j\). As a result of the considerations above the hamiltonian of the model reads

\[
H = -J \sum_{\langle ij \rangle} \left(1 - \delta_{\sigma_i R_{ij} \sigma_j} \right) \delta_{\sigma_i \sigma_j} \delta_{\eta_i \eta_j} \tag{4}
\]

\(<\langle ij \rangle\) denotes summation over nearest neighbor (nn) pairs only.) The extension of this model to a binary mixture with quenched disorder yields a hamiltonian given by

\[
H_{\text{mix}} = \sum_{\langle ij \rangle} J_{n_i n_j} (1 - \delta_{\sigma_i R_{ij} \sigma_j}) \delta_{\sigma_i \sigma_j} \delta_{\eta_i \eta_j} \tag{5}
\]

\(J_{11}, J_{22}, J_{12} = J_{21}\) are the interaction parameters between the two kinds of molecules, the randomly chosen numbers \(n_i\) are equal to \(1, 2\) if a lattice site is occupied by a molecule of component 1,2.

Since model (4) is very similar to the 3-state Potts model it is of some interest to investigate its properties in the pure nematogenic case. Besides the MFA we employed (I) a cluster variation (CV) technique, (II) a cluster mean field (CMF) approximation and performed (III) Monte Carlo simulations of the model.

3. The cluster approximations

In order to get some corrections to the mean field picture the approximation scheme of the cluster variation method is utilized [11,12], restricted to the two-cluster case or pair approximation [12]. In the framework of this theory the entropy \(\Sigma\) and internal energy \(U\) of the system (4) are given by

\[
\frac{\Sigma}{kN} = -\sum_{\sigma} \frac{1}{p_{\sigma}} \ln p_{\sigma} - \sum_{\alpha \beta} \frac{1}{p_{\alpha} p_{\beta}} \ln \frac{p_{\alpha} p_{\beta}}{p_{\alpha} + p_{\beta}} \tag{6a}
\]

\[
\frac{U}{JN} = -\sum_{\sigma} \left(1 - \frac{1}{p_{\sigma}}\right) p_{\sigma}^2 \tag{6b}
\]

where \(p_{\sigma}\) is the probability to find a molecule in state \(\sigma\) and \(p_{\alpha \beta}\) is the probability to find a molecule in state \(\sigma\) and in the \(Y\)-direction a molecule in state \(\beta\). Eq. (6b) follows immediately from (4) whereas (6a) is an extension of a formula given in [12].

In the uniaxial nematic phase the director is assumed to be aligned with the \(z\)-axis (\(\phi = 3\)); hence \(p_1 = p_2 = (1-p_3)/2\), \(p_3 = \chi\), \(\rho = \rho_2(\cos(\phi)) = (3\chi - 1)/2\) (\(\phi\) angle of the principal molecular axis with respect to the \(z\)-axis). Taking into account the symmetries of the system the probabilities \(\rho_{\alpha \beta}\) may be expressed in terms of \(\chi\).
The free energy \( F = U - T \sum \) (\( J = k = 1 \)) reads now

\[
F/N = -2(1-a-b-c-d-2e) - 5T(xln(x)+(1-x)ln((1-x)/2)) \\
+T((x-2a)ln(x-2a)+4aln(a)+(1-x-2a-2b)ln((1-x)/2-a-b) \\
+2bln(b)+2(x-c-e)ln(x-c-e)+6eln(c)+4eln(e)+4dln(d) \\
+(x-2c-2e)ln((1-x)/2-c-e)+(1-x-2e-2d)ln((1-x)/2-e-d)).
\] (7)

Minimizing \( F \) with respect to \( a, \ldots, e \) one yields \( 8 \) coupled equations, which, in part, can be solved analytically. After some algebra we get

\[
a = (1-x)/2 - b(1-q) \quad , \quad q = \exp(1/T) \]
(8a)

\[0 = b(2(1-q^2) + b(x+q)x - (1-x)^2)/4 \]
(8b)

\[d = (1-x)/2 - c - x^2q \]
(8c)

\[0 = e + x + e \]
(8d)

\[= \frac{x - c - e}{c^2(q - 1) + x - c}
\]
\[q - 1 \]

\[= \frac{1 - x}{2} \]
\[1 - x \]
\[0 = (\frac{1 - x}{2} - a - d)(\frac{1 - x}{2} - e - d) - d^2 \]
(8e)

\[1 - x \]
\[5 \]
\[2 \]
\[5 \]
\[1 - x \]
\[0 = (\frac{1 - x}{2} - x)(x - e - c) - e - a - b \]
(8f)

For a given \( x \) and \( c \) one immediately solves (8a-d). Then Eq. (8e) may be solved numerically and one calculates the value of \( a, \ldots, e \) for all possible \( x \). Finally it is easy to solve Eq. (8f) on a computer. From this procedure one obtains the order parameter \( S \), the values of \( \Sigma \), \( U \) and \( F \) as a function of the temperature \( T \). The phase transition temperature \( T_{n1} \) is determined by \( F(T_{n1}, S(T_{n1})) = F(T_{n1}, 0) \).

The pair approximation allows the calculation of the short range correlation function \( K(R_{12}) = \langle P_2(\cos(\theta(R_{12}))) \rangle \), where \( \theta(R_{12}) \) is the angle between two nn molecules separated by \( R_{12} \). The long range correlation function tends to \( S^2 \) as \( |R_{12}| \rightarrow \infty \). The result is

\[K^U = K(3) = 1 - 3(2a + b) \]
(8)

\[K^L = K(2) = K(1) = 1 - 3(c + d + e) \]

In contrast to the 3-state Potts model and the Lebwohl-Lasher model (lattice version of the Heisen-Saupe theory [13]) the short range correlation function is anisotropic; at \( T_{n1} \) one obtains \( K^U = 0.38, K^L = 0.415, S^2 = 0.274 \).

Now we turn to question of extending the CV technique to mixtures. Since the number of equations would be considerably increased the application of this method seems to be complicated. Therefore, a second approximation scheme (cluster mean field CBF) is proposed which is simpler to manage. The basic assumption is that only short range correlations are important. This is not so restrictive, even at \( T_{n1} \) as shown in computer simulation experiments [7, 14]. Thus, the six nearest neighbours of a molecule are almost statistically independent from another, and so
\[ p_3 = x = \frac{\sum_{(s_1)_{nn}} (\prod_{i=1}^{8} p_{s_i}) \exp[-E(s_1(s_1)_{nn})/kT]}{\sum_{s=1} \exp[-E(s_1(s_1)_{nn})/kT]} \]  

(10)

\[ = X(S,T), \]

where \( E(s_1(s_1)_{nn}) \) is the energy of a molecule in state \( s_1 \) surrounded by the six nearest molecules in the states \( s_1, \ldots, s_5 \). Eq. (10) is a self-consistent equation for \( x \) or \( S \) which has to be solved numerically. To determine the phase transition temperature one needs the free energy and hence the entropy in an appropriate approximation. According to the arguments which led to the CV formula for \( S \), we get

\[ \Sigma /2kN = \sum_{\alpha} p_{\alpha} \ln p_{\alpha} \]

\[ + \sum_{\alpha, (s_1)_{nn}} p(\alpha, (s_1)_{nn}) \ln \{ p(\alpha, (s_1)_{nn}) \}, \]

(11)

where \( p(\alpha, (s_1)_{nn}) \) is the probability for a configuration \( (s=s_\alpha, s_1, \ldots, s_5) \). The joint probability \( p(\alpha, (s_1)_{nn}) \) is proportional to \( \exp[-E(\alpha, (s_1)_{nn})/kT] \).

For comparison we briefly recall the mean field theory of (4) which is the same as for the 3-state Potts model (besides a rescaling of \( J \) due to the maximum energy per lattice site of 3J in the Potts model and 2J in our model). The free energy in MFA is \( (k = J = 1) \)

\[ F(T,S)/N = -2(1 + S^2)/3 + T(x \ln(x) + (1-x) \ln((1-x)/2)) \].

Minimizing \( F \) with respect to \( S=(3x-1)/2 \) one obtains

\[ S = \frac{\exp(4S/T) - 1}{\exp(4S/T) + 2}, \quad T_m = 1/ln2, \quad S(T_m) = 0.5. \]

The entropy jump \( \Delta \Sigma \) and the latent heat \( \Delta U \) are

\[ \Delta \Sigma /N = \frac{\ln 2}{3}, \quad \Delta U /N = 1/3. \]

In table 1 the results of the CV, CMF and mean field calculations are shown.

<table>
<thead>
<tr>
<th>MFA</th>
<th>CV</th>
<th>CMF</th>
</tr>
</thead>
<tbody>
<tr>
<td>( T_m )</td>
<td>1.44</td>
<td>1.205</td>
</tr>
<tr>
<td>( \Delta U )</td>
<td>0.333</td>
<td>0.250</td>
</tr>
<tr>
<td>( \Delta \Sigma )</td>
<td>0.231</td>
<td>0.207</td>
</tr>
<tr>
<td>( T_m' )</td>
<td>0.5</td>
<td>0.525</td>
</tr>
</tbody>
</table>

Tab. 1.

As expected from non mean field calculations the transition temperatures and the jumps of configurational entropy and internal energy are considerably reduced with respect to the MFA results, whereas the differences between the CV and CMF method are not so significant. As a consequence, it is sufficient to use the CMF approach in order to deal with binary mixtures (Sec.5).

4. The Monte Carlo simulations

We turn now to the computer simulation of the model (4) to test the consistence and reliability of the cluster approximations. As known from the 3-dimensional 3-state Potts model it is rather difficult to get sensible informations about the nature of the phase transition (15,16), large lattice sizes (\( >30 \times 30 \times 30 \)) and long computer runs are necessary. Nevertheless small scale simulations are useful too, taking into account the possible failure of the calculations in the vicinity of the transition point.

We have employed the standard heat-bath Monte Carlo technique with periodic boundary conditions which is more advantageous for discrete systems than the Metropolis algorithm. The properties evaluated by the simulations were the internal energy \( U \) and the order parameter \( S \). Due to the fluctuations of the director within the laboratory frame one has to average the largest eigenvalue of the tensor order parameter \( \Sigma = \left( \langle s^r \rangle s^s \right) - \langle s^r \rangle \langle s^s \rangle \) during the simulation run, where \( l_{sr} \) is the
direction cosine of the molecular axis with the lattice \(\mathbf{a}\) axis. \(<...>\) denotes the average over one lattice configuration (for a detailed discussion see [14]).

Fig. 2: Order parameter \(S\) versus temperature \(T\) for the model (4). MFA (a), CMF approximation (b), CV method (c), Monte Carlo data (d). The arrows indicate the transition temperatures of the approximations (a)-(d) and the corresponding values of the 3-state Potts model (these temperatures are rescaled by a factor of 2/3 to obtain the same transition temperature in MFA). The values for the CV (e) and the Monte Carlo method (f) were taken from [15] and [18], respectively. The estimated order parameter in the nematic and disordered phase at \(T_{ni}\) is indicated by the dashed arrows.

In Fig. 2 and Fig. 3 the simulation results of a 8x8x8 lattice are shown. We performed 500 Monte Carlo steps per site (MCS) for equilibration and 500 MCS to take data. The estimated transition temperature is about 1.16 but this is truly a rough value since finite size effects and possible metastable states were not considered [11]. From a long run (\(2 \cdot 10^6\) MCS at \(T = 1.16\)) the variance of the distribution function of the order parameter was determined indicating that the system is courting between an ordered and disordered phase. The estimated upper and lower values of \(S\) at \(T_{ni}\) are indicated by the dashed arrows in Fig. 2. For temperatures well below 1.16 the errors of the simulation data are sufficient small and hence a sensible comparison with the other methods is possible. From Fig. 1 and Fig. 2 it is obvious that the cluster approximations yield a satisfactory improvement with respect to the MFA whereas the CV technique is slightly better than the CMF approximation. Further, the rescaled transition temperatures of the 3-state Potts model are well above the corresponding temperatures of model (4). So, the thermodynamics of (4) is more affected by fluctuations and one expects similar consequences in the case of binary mixtures. Finally, the simple CMF approximation is sufficient to describe the influence of fluctuations and one may apply it to the mixture Hamiltonian (5).

Fig. 3. Internal energy versus order parameter for the model (4). The MFA yields simply \(U = -2(1 + 2S^2)/3\)
5. The model mixture

Now, the considerations of Sec. 3 and 4 are extended to the case of a binary nematic mixture whose hamiltonian is given by (5). As mentioned above the distribution of the two kinds of molecules is assumed to be unchanged during a macroscopic time interval. So, we exclude the formation of a two phase region in the phase diagram actually observed in experiments. Since we are interested in the relation between the order parameters in the nematic phase this assumption is not so restrictive.

To begin with the mean field theory of (6) one starts with the free energy

$$3F/N = 4 \left[ J_{11}x_m^2 + 2J_{12}x_m(1-x_m)S_1 + J_{22}(1-x_m)^2 \right] + 3kT \left[ x_m \sum_n p_n^{14} \ln p_n^{14} + (1-x_m) \sum_n p_n^{12} \ln p_n^{12} \right] \text{ const.,}$$

(12)

where $x_m$ is the mole fraction of the component 1 and the probabilities $p_n^{14}$, $p_n^{12}$ are related to the order parameters via $S_1 = 3(p_n^{14} - 1)/2$, $S_2 = 3(p_n^{12} - 1)/2$. Once more the geometric mean rule $J_{12} = (J_{11}J_{22})^{1/2}$ for the interaction parameters is used. Thus, the theory contains two parameters, the mole fraction $x_m$ and the ratio $J_{22}/J_{11} = r^2$ which is equal to the ratio of the transition temperatures $T_1/T_2$.

From (11) we return to Eqs. (1) but the function $f(z)$ has to be replaced by $[\exp(z) - 1]/[\exp(z) + 2]$. In comparison with the theory of Paffy-Kohoray et al. [1] the restricted orientation model yields an order parameter difference $S_2 - S_1$ somewhat enhanced for $S_1 > 0.4$ (Fig.1). Nevertheless, the qualitative behaviour of both models is the same.

Now we turn to the CFM approximation of (5). In the limiting cases $x \to 0$ and $x \to 1$ each molecule of the minority component is surrounded only by molecules of the other component and so the solution of the problem is given by the equations

$$S_1 = 3(X(T,S_1)-1)/2 , \quad S_2 = 3(X(T,r,S_2)-1)/2 , \quad (x_m = 1)$$

$$S_1 = 3(X(T,r,S_2)-1)/2 , \quad S_2 = 3(X(T,S_2)-1)/2 , \quad (x_m = 0),$$

where $X(T,S)$ was defined by (10). The case $0 < x < 1$ is more complicated because the averages are to be taken over all possible configurations of the constituents in the cluster of the six nearest neighbours. Nevertheless, these averages may be calculated exactly (on a computer) since all sums are finite. Finally one obtains the following equations:

$$S_1 = x_1(S_1,S_2,x_m,r) + \frac{1}{2} , \quad S_2 = x_2(S_1,S_2,x_m,r) + \frac{1}{2},$$

(13)

with

$$X_i = \sum_{\{s_1\}_{nn}} \sum_{\{n_1\}_{nn}} \left( \prod_{i=1}^{8} \frac{\exp[-E_i(3,s_1,n_1,r)/kT]}{\sum_{s=1}^{3} \exp[-E_i(s,s_1,n_1,n_1,r)/kT]} \right)$$

where $E_i(s,s_1,n_1,n_1,r)$ is the energy of a molecule in state $s_1$ for component $i$.

From the solutions of Eqs. (13) one yields the desired relation between the order parameters.

In addition to this the configurational entropy is determined by the same approximation which led to Eq. (11). This enables us to investigate the influence of the fluctuations on the phase diagram but not carried out in this paper.

6. Results and Discussion

We begin with the limiting case of $x_m \to 1$, $r \to \infty$ in order to verify the considerations made in Sec.1. We employed the CFM approximation for model (5) and the corresponding 3-state Potts model where the $J_{22}R_{ij}^3$ term in (5) has been dropped. The results for $S_2 - S_1$ are shown in Fig.4. It is obvious that the MFA fails qualitatively to predict the relation between the order parameters. In addition the $S_2 - S_1$ values in Fig.4 reveal that the 3-state Potts model is less sensitive to fluctuations as proposed by the higher transition temperature in the pure nematic case.

As known from other cluster theories [17] the deviations from the MFA are increasing with a decreasing number of nearest neighbours. Now, one may explain the results for model (5) by an effective reduction of the coordination number due to the $J_{12}$ dependence of the potential (expressed by the $J_{22}R_{ij}^3$ term). Thus the details of the interparticle potential are important for the interpretation of the observed order parameter differences.

Now we turn to the case of finite but large value of r to compare the
results of the CMF approximation with some simulation data. Once more the Monte Carlo simulations were performed on a 8x8 lattice with periodic boundary conditions. According to the value of \( x_m \), a random distribution of the molecules on the lattice sites was generated. Actually we repeated the simulations with different random number sequences to get reliable data. Unfortunately it is rather difficult to deal with \( x_m \to 1 \) or \( x_m \to 0 \) since the number of molecules of the minority component is too less to get satisfactory statistics. Hence we performed calculations for \( x_m = 0.5 \) to compare with the CMF results at \( x_m = 0 \) (Fig.5).

![Diagram](image)

**Fig.4:** Order parameter difference \( S_2 - S_1 \) versus \( S_1 \) in CMF approximation for the model (5) (a) and the 3-state Potts model (b) in the case of \( x_m \to 1 \) and \( r \to \infty \). The straight line is the MFA result.

Because of the lack of computer time (only 1000 MCS after reaching equilibrium) the simulation data are not very accurate. Nevertheless the MFA cannot explain the simulation data but on the other hand the CMF approximation underestimates the order parameter difference; the data are rather consistent with \( x_m = 0 \).

In order to get a sensible explanation of these discrepancies one has to perform more accurate computer simulations perhaps at \( r \) values of experimental relevance (\( r \approx 1.3 \)).

![Graph](image)

**Fig.5:** Order parameter difference versus \( S_4 \) for \( r = 2 \); MFA (a), CMF approximation (\( x = 0.5 \) (b), \( x = 0 \) (c)) and Monte Carlo data for \( x_m = 0.5 \) (\( r \)).

Finally we note again that the simple approximation made in Sec. 5 are sufficient to explain the order parameter differences in binary nematic mixtures whereas the MFA fails qualitatively. Furthermore we mention the importance of the \( F_{12} \) dependence of the intermolecular potential which led to the model (5). In comparison with the 3-state Potts model this model exhibits an increased sensitivity to fluctuations and yields an anisotropy of the short range correlation functions. This properties are of considerable interest for the interpretation of the measurements of liquid crystalline mixtures.
References


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Influence of Fluctuations on the Order Parameters in Nematic Mixtures

NMR measurements and computer simulations suggest that the component order parameters $S_1$ and $S_2$ of binary nematic mixtures differ much less from each other than proposed by mean field theories. Using a simple restricted orientation model for the nematogens and going beyond the mean field approximation one may explain the observed behaviour of the order parameters. The calculated relations between $S_1$ and $S_2$ are confirmed by Monte-Carlo simulations of the model.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

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