Magnetic field control of the ordering of two-component suspension of hard rods

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The Onsager theory of hard rod dispersion in a neutral solvent is extended to a case of two-component dispersion consisting of both non-magnetic and magnetic rods. It was found that the alignment of magneto-sensitive dispersion component by a magnetic field leads to the alignment of non-magnetic component in the dispersion and to an elimination of the isotropic phase. This effect is significant even at low relative concentrations of magnetic rods and leads to a magnetically induced anisotropy in a non-magnetic dispersion of rods mixed with the magnetic ones.

1. Introduction

In 1925, Zocher [1] discovered that the phase state of V$_2$O$_5$ aqueous colloidal suspension depends on the volume fraction of V$_2$O$_5$: at low fractions, the suspension was optically isotropic; at the volume fraction $\phi = 0.4\%$, the suspension was separated into isotropic and anisotropic phases coexisting in equilibrium; and at $\phi > 0.6\%$, the suspension became optically anisotropic. Such behaviour is typical for what was later called lyotropic liquid crystals [2]. Later, the formation of anisotropic phase in systems of sterically interacting hard rods was theoretically described by Onsager [3]. Onsager compared the orientational entropy of the system of hard rods that results in their orientational disordering and the packing entropy of rods that is related to the excluded volume effect and results in the ordered packing of rods. Depending on the concentration, the competition between those effects resulted in the isotropic, nematic and coexisting nematic–isotropic phases of the hard rod
suspension. In 1984, Lekkerkerker and co-workers [4,5] extended the Onsager theory to the two-component suspension of hard rods with two different lengths. It was found that the orderings of long and short rods are different in the nematic phase, and the longer particles are aligned better than the shorter ones. This conclusion confirmed the suggestion made earlier by Onsager [3].

In this paper, we consider a two-component suspension of hard rods, one of the components being subject to the action of an external magnetic field. Specifically, we apply the extended Onsager–Lekkerkerker theory to describe a two-component suspension of hard rods with two different lengths. One of the suspension components is magnetically sensitive and consists of rods with magnetic dipoles oriented along their long axes. It appears that the phase diagram of such a suspension in the magnetic field is changed. In particular, the orientation of magnetically sensitive rods by the magnetic field results in disappearance of the isotropic phase in the suspension. In other words, magnetically induced orientation of magnetic rods leads to the orientation of the non-magnetic component. This phenomenon is significant even if the concentration of magnetic rods is much lower than that of non-magnetic rods. It results in the strong effects of magnetically induced anisotropy in the hard rod suspensions doped with a small amount of magnetic rods, which was recently observed by Reznikov et al. [6] in a suspension of V2O5 ribbons doped with Fe2O3 nanoparticles.

2. Theory

Consider the dispersion of two kinds of hard rods in a neutral solvent. The rods of the first kind are not sensitive to a magnetic field (rods 1) and the rods of the second kind have magnetic moments along their long axis (rods 2). Let a suspension consisting of $N_1$ rods 1 with length $L_1$ and diameter $D_1$ and $N_2$ rods 2 with length $L_2$, diameter $D_2$ and magnetic moment $m$ be in a dialytic equilibrium with a neutral solvent at temperature $T$ in the magnetic field $H$. Following the approach of Onsager and Lekkerkerker, the excess of Helmholtz free energy over the free energy of the solvent, $\Delta F$, is determined by the functional

$$\frac{\Delta F}{Nk_BT} = \frac{\mu_0}{k_BT} - 1 + \ln c + (1 - x) \ln(1 - x) + x \ln x + (1 - x)\sigma_1 + x\sigma_2$$

$$+ c[b_{11}(1 - x)^2\rho_{11} + 2b_{12}x(1 - x)\rho_{12} + b_{22}x^2\rho_{22}]$$

$$- ax \int \cos \beta f_2(\Omega) \, d\Omega,$$  \hspace{1cm} (2.1)

where $(1 - x)$ is the mole fraction of rods 1, $x$ is the mole fraction of rods 2, $N = N_1 + N_2$ is the total number of particles, $c = N/V$ is the rod concentration, $b_{ij} = \pi(D_i + D_j)L_iL_j/8$, $\beta$ is the angle between $H$ and $m$, $a = (mH)/(k_BT)$, and $\sigma_j$ and $\rho_{ij}$ are the functionals that depend on the angular distribution of the long axis of rods 1, $f_1(\Omega)$, and the angular distribution of the magnetic moments of rods 2, $f_2(\Omega)$,

$$\sigma_j \equiv \int f_j(\Omega) \ln[4\pi f_j(\Omega)] \, d\Omega, \quad j = 1, 2$$  \hspace{1cm} (2.2)

and

$$\rho_{ij} \equiv \frac{4}{\pi} \int |\sin \gamma| f_i(\Omega)f_j(\Omega') \, d\Omega \, d\Omega', \quad i, j = 1, 2,$$  \hspace{1cm} (2.3)

where $\gamma$ is the angle between two vectors defined by the angles $\Omega$ and $\Omega'$. The distribution functions $f_1(\Omega)$ and $f_2(\Omega)$ satisfy the following normalization conditions:

$$\int f_1(\Omega) \, d\Omega = 1$$  \hspace{1cm} (2.4a)

and

$$\int f_2(\Omega) \, d\Omega = 1.$$

(2.4b)
In free energy (2.1), the term $\mu^0$ describes the chemical potential of the system, the term $-1 + \ln c + (1 - x) \ln(1 - x) + x \ln x$ corresponds to the translational entropy of the rods, the term $(1 - x)\sigma_1 + x\sigma_2$ describes the orientational entropy of the rods, and the term $c[b_{11}(1 - x)^2\rho_{11} + 2b_{12}(1 - x)\rho_{12} + b_{22}x^2\rho_{22}]$ describes the packing entropy related to the excluded volume effect. The last term corresponds to the energy of interaction between the magnetic rods and the external magnetic field. Note that the contributions of the terms proportional to $e^p$ with $p \geq 2$ are neglected.

In this article, we consider the case when the magnetic dipole–dipole interaction between rods 2 is much less than the steric interaction between the rods. Moreover, the magnetic interaction between rods 2 and $H$ is lower than the thermal energy $k_B T$, 

$$\lambda \equiv \frac{W_{d-d}}{k_B T} = \frac{m^2 x_c}{k_B T} \ll 1.$$  

Consider a change of phase diagram for a one-component dispersion of rods 1 at its doping with magnetic rods 2 oriented by a strong magnetic field ($\lambda \gg 1$). According to the Onsager method of free energy minimization for monodispersed rods [3], the distribution of rods 1 is described by the trial function 

$$f_1^{tr}(q) = \frac{q}{4\pi \sinh q} \cosh(q \cos \theta),$$  \hspace{1cm} (2.6)

where the parameter $q \geq 0$. In the case of low anisotropy of distribution function $f_1^{tr}$ (i.e. when $q \ll 1$), free energy (2.1) for pure dispersion of rods 1 looks like [3] 

$$\frac{\Delta F(q)}{N k_B T} = \text{const.} + \frac{q^4}{90} - \frac{c b_{11} q^4}{360}. \hspace{1cm} (2.7)$$

When the effective concentration of rods 1, $c b_{11}$, is not larger than the critical value of 4, the minimum of free energy (2.7) corresponds to $q = 0$, and the distribution of rods 1 is isotropic. When $c b_{11} > 4$, the minimum is realized at $q > 0$ and the distribution of rods 1 is anisotropic, which corresponds to the formation of a nematic phase.

The addition of magnetic rods 2 to the dispersion of rods 1 (i.e. at $x > 0$) results in qualitative changes to the system phase diagram. In this case, equation (2.7) takes the form 

$$\frac{\Delta F(q)}{N k_B T} = \text{const.} + (1 - x) \frac{q^4}{90} - (1 - x)^2 \frac{c b_{11} q^4}{360} + 2 c b_{11} x (1 - x) \rho_{12}. \hspace{1cm} (2.8)$$

Here, the term $2 c b_{11} x (1 - x) \rho_{12}$ describes steric interaction between rods 1 and rods 2.

In a strong magnetic field, the amplitude of interaction between rods 2 and the magnetic field is much larger than the steric interaction between the rods, i.e. $a \gg c b_{22}$ and $a \gg c b_{12}$. In this case, the angular distribution of rods 2, $f_{2\infty}$, takes the Boltzmann form: 

$$f_{2\infty}(a) = \frac{a}{4\pi \sinh a} \exp(a \cos \theta). \hspace{1cm} (2.9)$$

According to Onsager [3], integral (2.3) for the distributions $f_1^{tr}$ and $f_{2\infty}$ reads 

$$\rho_{12}[f_1^{tr}, f_{2\infty}] = \frac{2}{\pi \sinh a \sinh q} \int_0^\pi \cosh \sqrt{a^2 + q^2 + 2 a q \cos \gamma \cos \gamma} \cos \gamma \cos \gamma \, d\gamma. \hspace{1cm} (2.10)$$

For a large magnetic field, $a \gg q$ and equation (2.10) can be simplified to 

$$\rho_{12}[f_1^{tr}, f_{2\infty}] = \frac{2}{\pi \sinh q} \int_0^\pi \sinh(q \cos \gamma) \cos \gamma \cos \gamma \, d\gamma = \frac{2 h_1(q)}{\sinh q} \hspace{1cm} (2.11)$$

where $h_1$ is the modified Bessel function of the first kind [7]. For small $q$-values, the power series of (2.11) is 

$$\rho_{12}[f_1^{tr}, f_{2\infty}] = 1 - \frac{q^4}{24} + \cdots, \hspace{1cm} q \ll 1. \hspace{1cm} (2.12)$$
The substitution of expression (2.12) into formula (2.8) and the following minimization of the obtained functional leads to the equation

\[
\frac{1}{Nk_B T} \frac{\partial \Delta F(q)}{\partial q} = (1-x) \frac{2q^3}{45} - (1-x)^2 \frac{cb_{11}q^3}{90} - x(1-x) \frac{cb_{12}q}{6} = 0. 
\] (2.13)

When \( cb_{11}(1-x) < 4 \), this equation has two non-negative roots: \( q' = 0 \) and \( q'' = \sqrt{15cb_{12}x/[4 - cb_{11}(1-x)]} \).

The root \( q'' \) corresponds to the global minimum of free energy (2.8). It means that an addition of magnetic rods 2 aligned by the magnetic field to a dispersion of non-magnetic rods 1 results in the anisotropic distribution of non-magnetic rods 1. The order parameter of this distribution, \( S_1 \), transforms to

\[
S_1(q'') = \frac{q''^2}{15} = \frac{cb_{12}x}{4 - cb_{11}(1-x)}, \quad q'' \ll 1, 
\] (2.15)

provided that the trial function \( f_1 = f_1^e \) and \( q = q'' \).

One can see that for low concentrations of rods 1, \( cb_{11}(1-x) \ll 4 \), which corresponds to the isotropic phase of a pure suspension of rods 1, the order parameter for rods 1 is proportional to the concentration of rods 2, \( cx \), and the amplitude of interaction between rods 1 and rods 2, \( b_{12} \).

For example, for a two-component suspension of rods 1 and rods 2 with equal sizes, equal partial concentrations, \( x = 0.5 \), and the total concentration being half of the critical one, \( cb_{11} = cb_{12} = 2 \), the order parameter of rods 1 is 0.33. This value is high enough to be observed experimentally, e.g. by birefringence measurements.

Expression (2.15) is valid only for the case of strong magnetic field, \( a \gg cb_{12} \) and \( a \gg cb_{22} \), and small ordering of rods 1, \( q \ll 1 \). To obtain the angular distributions of rods 1 and rods 2 without those restrictions and to obtain the phase diagram of two-component suspension at any magnetic field strength, we applied numerical methods aimed at minimizing basic functional (2.1).

For a given thermodynamic state of the system, \((c, x, a)\), one can construct a Lagrange functional

\[
J = \Delta F + \lambda_1' \left( \int f_1(\Omega) d\Omega - 1 \right) + \lambda_2' \left( \int f_2(\Omega) d\Omega - 1 \right), 
\] (2.16)

where \( \lambda_1' \) and \( \lambda_2' \) are Lagrange undetermined multipliers. The minimization of functional (2.16) with respect to the distribution functions \( f_1(\Omega) \) and \( f_2(\Omega) \) results in the Euler–Lagrange equations

\[
\ln 4\pi f_1(\Omega) = \lambda_1 - \frac{8c}{\pi} \int |\sin \gamma(\Omega, \Omega')| [b_{11}(1-x)f_1(\Omega') + b_{12}xf_2(\Omega')] d\Omega' 
\] (2.17a)

and

\[
\ln 4\pi f_2(\Omega) = \lambda_2 - \frac{8c}{\pi} \int |\sin \gamma(\Omega, \Omega')| [b_{12}(1-x)f_2(\Omega') + b_{22}xf_2(\Omega')] d\Omega' + a \cos \beta. 
\] (2.17b)

Here, \( \lambda_1 = -\lambda_1' - 1 \) and \( \lambda_2 = -\lambda_2' - 1 \) are the coefficients that take into account normalization conditions (2.4). Equations (2.17) describe the distributions of rods 1 and rods 2 in a homogeneous region. The biphasic coexistence region is described in appendix A.

There are several methods of solving equations (2.17). In the case of one-component suspension and no magnetic field, equations (2.17) were solved for the first time by Lasher in his work [8]. Then Kayser & Raverché [9] used the bifurcation theory to investigate the existence of all possible multiple solutions of the integral equation. Herzfeld et al. [10] solved the problem by using the numerical iteration method, which we used in our work.

The results of numerical solutions of equations (2.17) in the whole range of rod concentrations allowed us to build up the phase diagram of the system for a given magnetic field value. These diagrams were plotted in the coordinates \((e, x)\), where \( e = cb_{11} = cb_{12} = cb_{22} \) is the effective rod...
concentration, which takes into account the steric interaction between them. As an example, the calculated diagrams for the case of equal sizes of rods 1 and rods 2 are presented in figure 1 for two magnetic field values. At a zero magnetic field, rods 1 and rods 2 are indistinguishable, and the phase diagram is reduced to the case of a one-component system (figure 1a). In agreement with Kayser & Raverché [9] and Herzfeld et al. [10], the system has only one or two local minima of its free energy, depending on the concentration. At \( e \leq 3.29 \) (region I in figure 1a), the global minimum corresponds to the isotropic distribution of rods. At \( e \geq 4.19 \) (region N), the axial anisotropic distribution of rods forms a nematic phase in the suspension. If the concentration falls within the range \( 3.29 < e < 4.19 \) (region \( N + I \)), the system becomes separated into an isotropic phase with the concentration \( e = 3.29 \) and an anisotropic one with \( e = 4.19 \).

In contrast to the cases when the rods are non-sensitive to the magnetic field or when the magnetic field is absent, the system with magnetically sensitive rods is always anisotropic in a magnetic field (figure 1b). Note that if the two-phase region of non-magnetic suspension consists of coexisting isotropic and nematic phases (region \( N + I \)), the two-phase region of magneto-sensitive suspension consists of two nematic phases with different concentrations of rod components (region \( N_1 + N_2 \)).

The suspension in the coexisting phase region \( N_1 + N_2 \) (figure 1b) decomposes into two homogeneous coexistent nematic fractions, \( N_1 \) and \( N_2 \), at the boundary between the coexisting phase region and homogeneous phase region (see appendix A and table 1). The first three columns in table 1 represent the concentration \( x \), the composition \( e \) and the order parameter \( S_1 \) in the phase \( N_1 \) and the last three in the phase \( N_2 \).

The dependencies of the ordering of rods 1 and rods 2 on the magnetic field for the isotropic \((e = 2)\) and nematic \((e = 5)\) suspension phases are presented in figures 2 and 3. In the absence of steric interaction between rods 2 and rods 1, the distribution of rods 2 is described by the Boltzmann function (no-interaction curve in figure 2a). In this case, the magnetic field does not influence the ordering of rods 1, and the order parameter of rods 1 is constant (figure 2b). The steric interaction between the rods in isotropic phase \((e = 2)\) results in an axial orientation ordering of rods 1 along the magnetic field (figure 2). In the case of nematic phase \((e = 5)\), the steric interaction improves the ordering of rods 2 along the magnetic field (figure 3).

The values of the order parameter \( S_1 \) of rods 1 estimated by formula (2.15) for high magnetic fields are depicted in figure 2 by stars. One can see that the approximate formula (2.15) describes
with magnetic nanorods.

Table 1. Parameters of the system at the conjugate points on the phase diagram, figure 1b, at $S_2 \approx 1$.

<table>
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<th>$x$</th>
<th>$e$</th>
<th>$S_1$</th>
<th>$e'$</th>
<th>$x'$</th>
<th>$S'_1$</th>
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<td>0</td>
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Figure 2. Dependencies of the order parameter of (a) rods 2 and (b) rods 1 on the magnetic parameter $a$ for $e = 2$ and various molar fractions $x$. The curve no interaction shows the limit $x \to 0$. The stars show the estimated order parameter.

3. Estimations

An important conclusion of the results of our calculations is that the magnetically induced order parameter of non-magnetic component is high and can be comparable with that of the nematic phase of hard rods. Therefore, strong effects of magnetically induced anisotropy may be expected in the suspensions of hard rods, e.g. in aqueous suspensions of DNA, carbon nanotubes, vanadium pentoxide ribbons, etc. Let us estimate the value of magnetically induced ordering in an aqueous suspension of vanadium pentoxide nanoribbons ($V_2O_5$ ribbons) doped with magnetic nanorods.

quantitatively the rod ordering at high magnetic fields and at low concentrations of rods 2, i.e. at low $x$. 

3. Estimations

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Figure 3. Dependencies of the order parameter of (a) rods 2 and (b) rods 1 on the magnetic parameter \( a \) for \( e = 5 \) and various molar fractions \( x \).

Figure 4. Dependencies of the order parameter of goethite rods and \( V_2O_5 \) rods on the magnetic field: (a) \( \phi_G = 1\% \); \( \phi_V = 0.2\% \) and (b) \( \phi_G = 3\% \); \( \phi_V = 0.2\% \).

We will make estimations for aqueous suspensions of \( V_2O_5 \) ribbons, which are typical systems showing the isotropic–nematic phase transition. Their main physical properties were recently reviewed by Davidson [11]. Typically, \( V_2O_5 \) ribbons are approximately 300 nm long, 25 nm wide and 1 nm thick, and they are charged. The isotropic–biphasic and the biphasic–nematic transitions in the suspension occur at \( V_2O_5 \) ribbon volume fractions of about 0.4 per cent and 0.6 per cent, respectively, which corresponds to the Onsager theory of steric interaction applied to charged hard rods 320 nm in effective length and 55 nm in diameter.

First, consider the magneto-anisotropic properties of \( V_2O_5 \) suspension mixed with ferromagnetic goethite rods. The main physical properties of goethite were described in earlier studies [12,13]. Goethite magnetic nanorods have a lath-like shape of a typical size of \( 330 \times 40 \times 18 \text{ nm}^3 \) and a small permanent magnetic moment of \( 1.1 \times 10^{-17} \text{ emu} \). Here, we neglect the anisotropic magnetic permeability of goethite rods. The suspension of goethite rods has a biphasic–nematic transition at a volume fraction of 7.5 per cent, and it is described by the Onsager theory for charged hard rods (with an effective length of 355 nm and a diameter of 61 nm). The dependence of magnetically induced order parameters of goethite and \( V_2O_5 \) components with the volume concentrations \( \phi_G = 1\% \) and \( \phi_V = 0.2\% \), respectively, on the magnetic field are depicted in figure 4a. One can see a non-threshold ordering of \( V_2O_5 \) ribbons, which is almost proportional to the magnetic field up to \( H = 10 \text{ kOe} \). If the magnetic field increases further, the goethite ordering
saturates. It results in the corresponding saturation of the magnetically induced ordering of the V$_2$O$_5$ ribbons. The coupling between the components can be enhanced if the concentration of one of them is close to the critical value corresponding to the nematic–isotropic transition. The analogous dependencies for a suspension with $\varphi_G = 3\%$ and $\varphi_V = 0.2\%$ are presented in figure 4b. One can see that the ordering of ribbons is close to the ordering of goethite; the components are coupled so strongly that the system behaves almost as a one-component suspension.

Goethite is a weak ferromagnetic material. Therefore, in the estimations shown above, the field and the fraction of magnetic component, which are needed for the ordering of non-magnetic component, are high. At the same time, an effective control of non-magnetic component requires a weak field and a low fraction of magnetic component. From this point of view, doping of V$_2$O$_5$ suspension with strongly ferromagnetic Ni nanorods looks promising. According to Gunter et al. [14], Ni nanorods can possess a magnetic moment of $5 \times 10^{-14}$ emu. In this case, the application of Ni nanorods $240 \times 28$ nm$^2$ in dimensions results in the ordering of V$_2$O$_5$ ribbons (see figure 5 obtained for a relatively small molar fraction of Ni nanorods, $x = 0.135$). One can see that in this case the characteristic magnetic fields needed for the alignment of V$_2$O$_5$ ribbons change from the range of several thousands to tens of oersteds. These fields are of the same order of magnitude as those applied to standard thermotropic liquid crystals in optics devices.

4. Conclusion

The extension of the Onsager–Lekkerkerker theory for a two-component hard rod suspension to the case when one of the components is sensitive to an external field shows that the field-induced ordering of this component leads to the ordering of the other component. In contrast to the cases when the rods are non-sensitive to the magnetic field or when the magnetic field equals zero, the system with magnetic sensitive rods is always anisotropic in a non-zero magnetic field. The phase diagram of magneto-sensitive suspension includes a region where two nematic phases with different component concentrations and orderings coexist in a magnetic field. The effect of field-induced ordering in the field-insensitive component should not be restricted to the considered case of magneto-sensitive rods in a magnetic field. We believe that analogous effects can take place with electric and optical fields as well. It opens good perspectives for the application of Onsager-like liquids in electro-, magneto- and nonlinear optics.

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Appendix A

Here we discuss the conditions of the coexistence region of the suspension. At a given magnetic parameter $a$, the rod suspension has a phase region, where the system is decomposed into two coexisting states $(c, x, a)$ and $(c', x', a)$, and which corresponds to the minimum of total free energy. In this region, the osmotic pressure $\Pi$ and the chemical potentials $\mu_1$ and $\mu_2$ in both phases are equal:

\[
\Pi(c, x) = \Pi(c', x'),
\]

\[
\mu_1(c, x) = \mu_1(c', x'),
\]

\[
\mu_2(c, x) = \mu_2(c', x').
\]

According to Lekkerkerker et al. [4],

\[
\Pi(c, x) = -\left(\frac{\partial \Delta F}{\partial V}\right)_{T, \mu_0, N_1, N_2}
= k_B T \ln [1 + c((1 - x)^2 b_{11} \rho_{11} + 2 x (1 - x) b_{12} \rho_{12} + x^2 b_{22} \rho_{22})],
\]

\[
\mu_1(c, x) = -\left(\frac{\partial \Delta F}{\partial N_1}\right)_{T, \mu_0, V, N_2}
= \mu_1^0 + k_B T \ln c + \ln (1 - x) + \sigma_1 + 2 c((1 - x) b_{11} \rho_{11} + x b_{12} \rho_{12})
\]

and

\[
\mu_2(c, x) = -\left(\frac{\partial \Delta F}{\partial N_2}\right)_{T, \mu_0, V, N_1}
= \mu_1^0 + k_B T \ln c + \ln x + \sigma_2 + 2 c((1 - x) b_{12} \rho_{12} + x b_{22} \rho_{22})].
\]

Lekkerkerker et al. [4] noted that there was one degree of freedom in a suspension of two different kinds of rods. In our system, we have an additional degree of freedom, namely, the magnetic parameter. As a result, we have five independent thermodynamic variables—$a$, $x$, $c$, $x'$ and $c'$—in three equations (A 1) so that our two-component system has two degrees of freedom.

References


