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A simple model of liquid-crystalline magnetic suspension of anisometric particles

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ABSTRACT

On the base of molecular-statistical approach we study the phase transition between the ordered (ferromagnetic) and disordered (paramagnetic) phases in liquid-crystalline suspensions of magnetic nanoparticles in an external magnetic field. The free energy and equations of magnetic and orientational equilibrium are obtained in the framework of spherical approximation.

It is well known that liquid crystals (LCs) have low anisotropy of diamagnetic susceptibility, this is why strong magnetic field (H~1 kOe) is required to control their orientational structure. In 1970 Brochard and de Gennes [1] proposed the idea to create highly dispersed liquid-crystalline suspensions of magnetic nanoparticles using nematic LCs as a carrier, which were called ferronematics (FNs). The initial magnetic susceptibility of FNs is several orders of magnitude higher than that of pure nematic, which allows to control orientation of FNs' structure by relatively weak magnetic fields (H~10 Oe).

Using the molecular-statistical approach we study external magnetic field induced orientational transitions in FNs on the base of exactly solvable mean-field spherical model. We consider the case of soft and planar anchoring between the particles and the director, so in the magnetic field absence the director and long axes of the particles are parallel. Let the external magnetic field $\mathbf{H} = (0,\ 0,\ H)$ be along the z-axis. Equilibrium orientational and magnetic structure of FN corresponds to a free energy minimum.

We describe the orientation of rod-like molecule at point $\mathbf{r}_\alpha$ by traceless symmetric second rank tensor

$$\nu_{ik}^{\alpha} = \sqrt{\frac{3}{2}}\left(\nu_{\alpha i}\nu_{\alpha k} - \frac{1}{3}(\boldsymbol{\nu}_\alpha)^2\delta_{ik}\right), \tag{1}$$

where the unit vector $\boldsymbol{\nu}_\alpha$ is aligned along the main axis of nematic molecule ($\alpha = \overline{1,\ N_n}$). For the impurity particle we have analogously

$$e_{ik}^{\beta} = \sqrt{\frac{3}{2}}\left(e_{\beta i}e_{\beta k} - \frac{1}{3}(\mathbf{e}_\beta)^2\delta_{ik}\right), \tag{2}$$

where the unit vector $\mathbf{e}_\beta$ is directed along the main axis of anisometric particle ($\beta = \overline{1,\ N_p}$). Macroscopic tensors of orientation of the suspension components can be obtained by statistical averaging of tensors (1) and (2):

$$\begin{aligned}\eta_{ik} &\equiv <\nu_{ik}^{\alpha}> \\ &= \sqrt{\frac{3}{2}}\eta\left(n_i n_k - \frac{1}{3}\delta_{ik}\right), \qquad S_{ik} \\ &\equiv <e_{ik}^{\beta}> \\ &= \sqrt{\frac{3}{2}}S\left(n_i n_k - \frac{1}{3}\delta_{ik}\right).\end{aligned} \tag{3}$$

We assume that the orientations of impurity particles and LC molecules are coupled in equilibrium state, therefore the tensors (3) can be written in terms of a unit vector $\mathbf{n}$, called the director, that corresponds to the preferred orientation direction of LC molecules. Here we have introduced the scalar order parameters $\eta \equiv <P_2(\mathbf{n}\boldsymbol{\nu})>$ and $S \equiv <P_2(\mathbf{ne})>$, where P_2 is the second Legendre polynomial.

The orientational interaction energy of FN considered as a mixture of N_n nematic molecules and N_p magnetic nanoparticles, can be written as [2]

$$\mathcal{H} = -\frac{1}{2}\sum_{\alpha\neq}^{N_n}\sum_{\beta} A(r_{\alpha\beta})\nu_{ik}^{\alpha}\nu_{ik}^{\beta} - \sum_{\alpha}^{N_n}\sum_{\beta}^{N_p} B(r_{\alpha\beta})\nu_{ik}^{\alpha}e_{ik}^{\beta} - \sum_{\beta}^{N_p}\boldsymbol{\mu}_\beta\mathbf{H}. \tag{4}$$

Here the first contribution is the interaction energy between nematic molecules, the second is the interaction energy between nematic molecules and nanoparticles and the last is the particles' magnetic moments $\boldsymbol{\mu}_\beta = M_s v_p \mathbf{e}_\beta$ energy in the external magnetic field $\mathbf{H} = (0,\ 0,\ H)$ [$\mathbf{e}$ is a magnetization unit vector, M_s is the saturation magnetization of magnetic particles material]. Hereinafter the summation over repeated tensor indices is implied.

In Eq. (4) the interaction energies $A(r_{\alpha\beta})$ and $B(r_{\alpha\beta})$ depend on the

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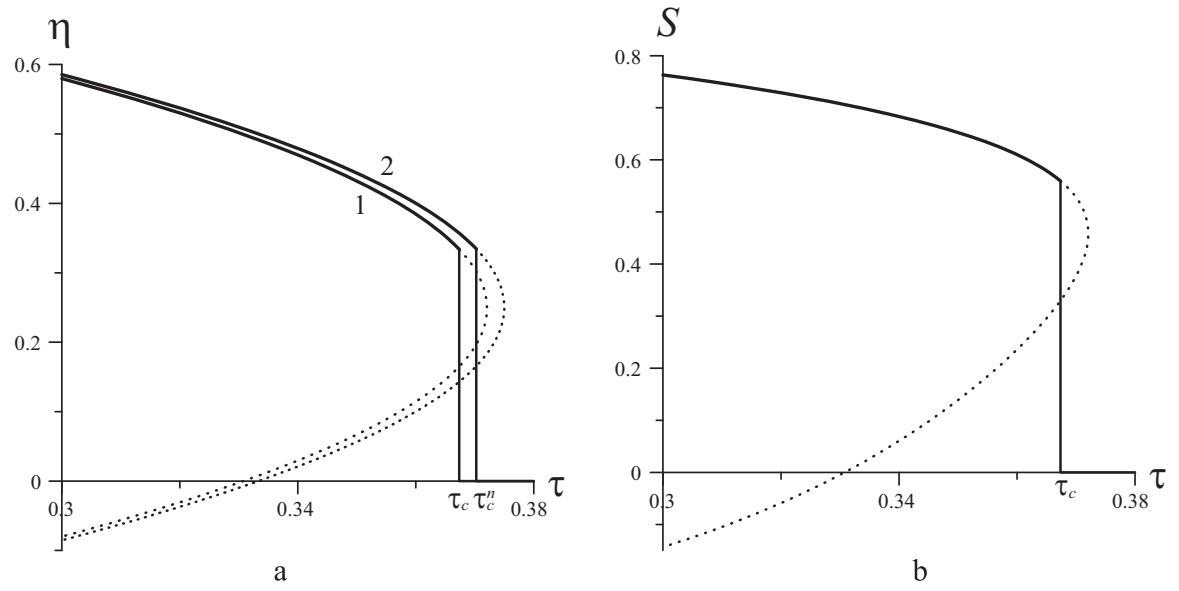

Fig. 1. Temperature dependences of the LC-matrix order parameters η (a) and dispersed nanoparticles S (b) in the field absence for ferronematic (curve 1, $\tau_c = 0.3674$, $\eta_c = 0.3337$, $S_c = 0.5596$) and pure nematic (curve 2, $\tau_c^n = 0.3702$, $\eta_c^n = 0.3345$) for $y_p = 0.01$, $\gamma = 0.06$ and $\omega = 2$.

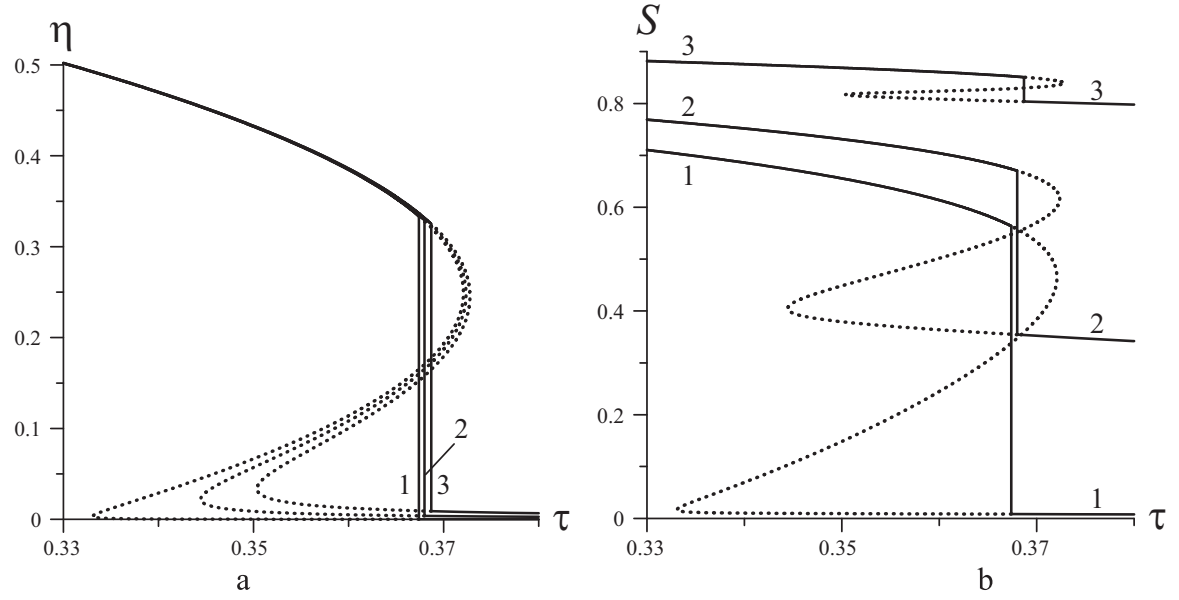

Fig. 2. Temperature dependences of the LC-matrix order parameters η (a) and dispersed nanoparticles S (b) under magnetic field for $y_p = 0.01$, $\gamma = 0.06$ and $\omega = 2$. Curves 1 – $h = 0.1$, $\tau_{c1} = 0.3674$, $\eta_{c1}^{up} = 0.3336$, $\eta_{c1}^{down} = 0.0001$, $S_{c1}^{up} = 0.5636$, $S_{c1}^{down} = 0.0083$. Curves 2 – $h = 1$, $\tau_{c2} = 0.3680$, $\eta_{c2}^{up} = 0.3296$, $\eta_{c2}^{down} = 0.0039$, $S_{c2}^{up} = 0.6705$, $S_{c2}^{down} = 0.3544$. Curves 3 – $h = 5$, $\tau_{c3} = 0.3687$, $\eta_{c3}^{up} = 0.3246$, $\eta_{c3}^{down} = 0.0091$, $S_{c3}^{up} = 0.8506$, $S_{c3}^{down} = 0.8036$.

distance $r_{\alpha\beta} = |\mathbf{r}_\alpha - \mathbf{r}_\beta|$ between the molecules or molecules and nanoparticles. The energy $A(r_{\alpha\beta}) > 0$, that corresponds to a minimum of energy (4) when molecules long axes are parallel, and $B(r_{\alpha\beta})$ can be of any sign. We consider the case $B(r_{\alpha\beta}) > 0$, then energy (4) minimum in the field absence corresponds to a parallel orientation of long axes of the molecules and impurity particles [the planar anchoring, $\boldsymbol{\nu}\|\mathbf{e}$]. We also assume the low concentration of dispersed particles, that allows to neglect magnetic dipole-dipole interactions between the particles in the suspension.

Using the mean field approximation, from Eq. (4) we obtain [2,3]

$$\mathcal{H} = \lambda\left\{N_n\left(\frac{1}{2}y_n\eta_{ik} + y_p\omega\gamma S_{ik}\right)\eta_{ik} - \sum_{\alpha}^{N_n}(y_n\eta_{ik} + y_p\omega\gamma S_{ik})\nu_{ik}^{\alpha} - \sum_{\beta}^{N_p}(y_n\omega\eta_{ik}e_{ik}^{\beta} - (\mu\mathbf{H}/\lambda)\mathbf{e}_\beta)\right\}. \quad (5)$$

Here we have introduced the notations for the mean field parameters

$$\sum_{\alpha}^{N_n} A(r_{\alpha\beta}) = \frac{N_n}{V}A,\ \sum_{\alpha}^{N_n} B(r_{\alpha\beta}) = \frac{N_n}{V}B,\ \sum_{\beta}^{N_p} B(r_{\alpha\beta}) = \frac{N_p}{V}B,\ \lambda = \frac{A}{v_n}$$

[V is FN volume, v_n is molecular volume, v_p is impurity particle volume], and volume fractions of the components $y_p = N_p v_p/V$, $y_n = N_n v_n/V = 1 - y_p$, and dimensionless parameters $\gamma = v_n/v_p$, $\omega = B/A$. The parameter ω describes the relative role of anisotropic orientational interactions between the nanoparticles and LC molecules, the parameter γ describes a relative particle volume.

To calculate the statistical integral for the suspension we use the so-called spherical approximation, which is well known from the theory of magnetism. In this case instead of considering $\boldsymbol{\nu}_\alpha$ and $\mathbf{e}_\beta$ to be unit vectors one can allow these vectors to have any length under the so-called spherical constraint, that the sum of squares of their length