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Landau description of ferrofluid to ferronematic phase transition

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A Landau-type free energy function is presented to describe the phase transitions from an isotropic (superparamagnetic) ferrofluid to a ferromagnetic nematic liquid crystal either directly or via a superparamagnetic nematic liquid crystal. These two nematic phases are usually both called 'ferronematic', although they are distinct phases. Both show nematic ordering, but only the ferromagnetic phase shows spontaneous magnetic ordering, additionally. The interplay of nematic with magnetic order is of special interest. A phase with ferromagnetic order but no nematic order is not possible in the present model, since the former always implies the latter (but not vice versa). In the presence of a strong external magnetic field these transitions are smeared out and the different ferronematic phases become more similar to each other.

1. Introduction and Motivation Ferrofluids are suspensions of nano-sized ferromagnetic particles in some carrier liquid (for example, water or oil) [1]. Without an applied external magnetic field the orientations of the magnetic moments of the particles are random resulting in a vanishing macroscopic magnetization (magnetic disorder). An external magnetic field, however, easily orients the particles' magnetic moments and a large (induced) magnetization is obtained. This "superparamagnetic" property is the basis for many applications.

In 1970 the idea emerged [2] to dope nematic liquid crystals with a small amount of ferromagnetic grains (volume fraction of $10^{-5} - 10^{-4}$) in order to enhance the magnetic susceptibility. The expectation was that a strong coupling between the orientation of the magnetic grains and the nematic ordering induces a spontaneous macroscopic magnetization (ferromagnetic state), which allows easy control of the director alignment by weak external magnetic fields.

Quite rapidly, mixtures of thermotropic calamitic [3, 4] and discotic [5], as well as lyotropic nematics [6] with ferroparticles were produced. However, these systems were more like dirty liquid crystals, where the magnetic additives served for a better orientation of the nematics in an external field. Problems were the stability of these mixtures and the mutual orientation of the director and the magnetization. The experimental situation changed considerably, when it was possible to make stable emulsions, first as ferrosmectic systems [7–9], were the ferromagnetic nano-particles are embedded in the layers. These ferrosmectics are very dilute systems, which prevents their use in applications. Recently, stable ferronematic systems, where the liquid crystal and the magnetic aspects are on equal footing, have drawn increasing attention (apart from other rather exotic phases, like ferrovesicles [10]). Birefringence [11–13] behavior in homogeneous electric [14], and magnetic fields (including the Frederiks transition [13, 15–17]) in inhomogeneous fields [18], and under the influence of bounding surfaces [19] have been investigated.

With the synthesis of thermotropic ferronematics [20] it turned out that the orientation between the magnetization and the nematic director is not completely

rigid. Based on a microscopic treatment of rodlike ferromagnetic grains Burylov and Raikher [21] derived an expression for the free energy of a ferronematic, which treats the orientations of the magnetization and the nematic ordering as separate degrees of freedom. As in Ref. [2] the strength of the magnetization is assumed to be in saturation, even without external fields. However, in the ferronematics produced up to now no indication of a true ferromagnetic behavior (spontaneous magnetization in equilibrium without external fields) has been reported and they all seem to be superparamagnetic like the ordinary isotropic ferrofluids.

Here we discuss possible phase transitions as a function of temperature (or pressure or ferrofluid particle concentration) between an isotropic ferrofluid phase (I), the superparamagnetic (N) and the ferromagnetic (F) nematic phase. Using a Landau-de Gennes expansion of a nematic and a magnetic order parameter up to fourth order, we find that depending on the signs and magnitude of the crosscouplings between nematic and magnetic order there is the possibility to have either a direct FI transition or a two-step NI and FN transition. Similarities to the isotropic-nematic-smectic A transitions in ordinary liquid crystals are discussed. In the ferromagnetic phase the magnetic and the nematic orientations are either parallel or orthogonal. In the present model there is no possibility to have a ferromagnetic phase that lacks nematic ordering. In the last section we show how the jumps of the order parameters are reduced by the presence of an external field and eventually are completely smeared out, if the field is strong enough.

2. Phase Transitions without Field The starting point of our approach is to write down the Landau free energy density f. In order to obtain the isotropic-nematic phase transition as well, we make use of the nematic order parameter originally proposed by de Gennes [22], a symmetric, traceless tensor described by $Q_{ij} = \frac{1}{2}S(3\hat{n}_i\hat{n}_j - \delta_{ij})$. The unit vector $\hat{\boldsymbol{n}}$ describes the orientation of the nematic ordering. Since the nematic order does not discriminate between "up" and "down" along the preferred direction, $\hat{\boldsymbol{n}}$ and $-\hat{\boldsymbol{n}}$ are equivalent and $\hat{\boldsymbol{n}}$ is called a director. Q_{ij} is the simplest tensorial quantity compatible with this up-down symmetry. The quantity S defines the strength of the nematic ordering (the modulus of the nematic order parameter) and is zero (one) for complete disorder (order). Thus in the isotropic phase $S = 0$ and in the nematic phase $S \neq 0$. The magnetic order is described by the magnetization $\boldsymbol{M} = M\hat{\boldsymbol{m}}$ whose modulus M is zero in a (super)paramagnetic and non-zero in a ferromagnetic state. The unit vector denotes the orientation of the magnetic ordering. It is an axial vector, which is odd under time reversal symmetry (like any magnetic field).

Keeping terms up to quartic order the total free energy density near the isotropic/nematic and para-/ferromagnetic transition can be written as:

$$\begin{aligned} f &= f_0 + \frac{A}{2}Q_{ij}Q_{ij} - \frac{B}{3}Q_{ij}Q_{jk}Q_{ki} + \frac{C_1}{4}(Q_{ij}Q_{ij})^2 + \frac{C_2}{4}Q_{ij}Q_{jk}Q_{kl}Q_{li} \\ &\quad + \frac{\alpha}{2}\boldsymbol{M}^2 + \frac{\beta}{4}\boldsymbol{M}^4 + \frac{\gamma}{2}M_iM_jQ_{ij} + \frac{\delta_1}{2}\boldsymbol{M}^2Q_{ij}Q_{ij} + \frac{\delta_2}{2}M_iM_kQ_{ij}Q_{kj} \end{aligned} \qquad (1)$$

where f_0 is the free energy density of the isotropic (superparamagnetic) ferrofluid phase. The change of sign of $A \equiv A_0(T - T^*_{NI})$ and $\alpha \equiv \alpha_0(T - T^*_{FP})$ at the critical temperatures T^*_{NI} and T^*_{FP} corresponds to a hypothetical second order isotropic to nematic, and to the para- to ferromagnetic transition, respectively, if the cross coupling terms ($\sim \gamma$ and $\sim \delta_{1,2}$) are absent. All other coefficients, as well as A_0 and α_0, are assumed to be constant near the transition point. For pressure - or (more likely) - concentration-driven phase transitions one could write $A \equiv A_c(c - c^*_{NI})$ and $\alpha \equiv \alpha_c(c - c^*_{FP})$ with a change in sign at some critical

concentrations of ferroparticles. For simplicity we will stick to the temperature related notation in the following.

In addition to the bi-quadratic couplings between the nematic and magnetic order governed by the coupling coefficients $\delta_{1,2}$, there is also a cubic coupling ($\sim \gamma$), which is most important for these transitions. As will be discussed below, it is responsible for the fact that magnetic order induces the nematic order. As we shall see, negative (positive) values of $\delta_{1,2}$ favor (disfavor) ferromagnetic nematics over the (super)paramagnetic ferronematics.

We assume C_1, C_2, β and $\beta C_{1,2} - \delta_{1,2}^2$ to be positive to guarantee the stability of the isotropic phase at high temperatures and $B > 0$ to get $S > 0$ in the ferronematic phases, which is suitable for rod-like nematogens.

Here we consider phases in which the nematic and magnetic order are spatially invariant, $S = const.$ and $M = const.$, and the ordering directions $\hat{\boldsymbol{n}}$ and $\hat{\boldsymbol{m}}$ make an angle ψ, i.e. $\hat{\boldsymbol{n}} \cdot \hat{\boldsymbol{m}} = \cos\psi$. In that case eq.(1) reads

$$\begin{aligned} f - f_0 &= \frac{3}{4}AS^2 - \frac{1}{4}BS^3 + \frac{9}{16}CS^4 + \frac{1}{2}\alpha M^2 + \frac{1}{4}\beta M^4 + \frac{3}{4}\delta_1 M^2 S^2 \\ &\quad + \frac{1}{8}\delta_2 M^2 S^2 (3\cos^2\psi + 1) + \frac{1}{4}\gamma M^2 S\,(3\cos^2\psi - 1) \end{aligned} \tag{2}$$

where $C \equiv C_1 + C_2/2$. The presence of the cubic terms ($\sim B$ and $\sim \gamma$) describes the first order character of the isotropic ferrofluid to ferronematic transitions. Eq.(2) has a structure quite similar to that of a Landau free energy for the isotropic to smectic A phase transition in conventional thermotropic liquid crystals [23].

Minimization of Eq.(2) with respect to S, M and ψ yields the following three phases:

$$\text{Isotropic Ferrofluid}: \quad S = 0, \quad M = 0, \quad \psi = \text{undefined} \tag{3}$$

$$\text{Paramagnetic Nematic}: \quad S_N = \frac{1}{6C}\left[B + (B^2 - 24AC)^{1/2}\right] > 0, \quad M = 0, \quad \psi = \text{undefined} \tag{4}$$

$$\text{Ferromagnetic Nematic}: \quad S_F > 0, \quad M^2 = -\frac{1}{\beta}\Big(\alpha + \tilde{\gamma} S_F + \tilde{\delta} S_F^2\Big), \quad \psi = 0 \text{ or } \pi/2 \tag{5}$$

where S_F is defined by

$$-\frac{2}{3}\frac{\alpha\tilde{\gamma}}{\beta} + 2A^* S_F - B^* S_F^2 + 3C^* S_F^3 = 0 \tag{6}$$

with the abbreviations $A^* = A - (2/3\beta)\tilde{\delta}\alpha - (1/3\beta)\tilde{\gamma}^2$, $B^* = B + (2/\beta)\tilde{\delta}\tilde{\gamma}$ and $C^* = C - (4/9\beta)\tilde{\delta}^2$. The new coefficients $\tilde{\delta}$ and $\tilde{\gamma}$ depend on the angle ψ and are

$$\tilde{\delta} = \begin{cases} \frac{3}{2}\delta_1 + \delta_2 \\ \frac{3}{2}\delta_1 + \frac{1}{4}\delta_2 \end{cases} \qquad \tilde{\gamma} = \begin{cases} \gamma & \text{for} \quad \psi = 0 \\ -\frac{1}{2}\gamma & \text{for} \quad \psi = \pi/2 \end{cases} \tag{7}$$

There is no extremum of (1) with $S = 0$ and $M \neq 0$, i.e. no ferromagnetic phase without nematic ordering. This can easily be seen directly from the coupling term $\sim \gamma$ in (1), where a finite M^2 acts as an external "field" on the nematic order, thus inducing a non-zero S.

Necessary conditions for the different phases to be stable are

$$\frac{\partial^2 f}{\partial S^2} > 0, \qquad \frac{\partial^2 f}{\partial M^2} > 0, \qquad \frac{\partial^2 f}{\partial \psi^2} > 0, \qquad \frac{\partial^2 f}{\partial S^2}\frac{\partial^2 f}{\partial M^2} - \Big(\frac{\partial^2 f}{\partial S\,\partial M}\Big)^2 > 0 \tag{8}$$

The derivatives in (8) have to be taken at the values (3-5) for the appropriate phases. For the isotropic ferrofluid phase the stability conditions are simply $\alpha > 0$ and $A > 0$. The paramagnetic nematic phase is stable, if $\alpha + \tilde{\gamma}S + \tilde{\delta}S^2 > 0$ and $24AC < B^2$.

The possible orientation between nematic and magnetic ordering in the ferromagnetic phase, either parallel ($\psi = 0$) or orthogonal ($\psi = \pi/2$), is fixed by the third stability condition in (8), where the former (latter) orientation is obtained for $\delta_2 S_F + 2\gamma < 0$ (> 0). Thus, a negative (positive) γ favors parallel (orthogonal) orientation. In the parallel case the phase has uniaxial $D_{\infty h}$ symmetry (as conventional nematics), while in the orthogonal case the phase is biaxial with orthorhombic D_{2h} symmetry. For ψ a value different from zero or ninety degrees could only be obtained by taking into account terms higher than forth order in the Landau expansion, a procedure, for which there is no a priori reason. The other stability conditions for the ferromagnetic case read

$$A - BS_F + \frac{9}{2}CS_F^2 + \frac{2}{3}\tilde{\delta}M^2 > 0 \tag{9}$$

$$\alpha + \tilde{\gamma}S_F + \tilde{\delta}S_F^2 < 0 \tag{10}$$

$$A^* - B^*S_F + \frac{9}{2}C^*S_F^2 > 0 \tag{11}$$

ensuring S_F and M^2 to be positive quantities. Obviously, a negative $\tilde{\delta}$ enhances the stability of the ferromagnetic phase. These stability conditions determine the existence ranges (in terms of temperature) of the different phases rather implicitly. The existence ranges of all three phases generally overlap. That phase with the lowest free energy is the stable one. A (first order) transition takes place, when 2 free energies are identical. The isotropic ferrofluid to paramagnetic nematic transition, thus, takes place, when the right hand side of (2) is zero taking for M and S the values (4). This happens for $A = B^2/27C$ leading to a transition temperature $T_{NI} = T^*_{NI} + B^2/27CA_0$ somewhat larger than the critical transition temperature of the hypothetical second order transition.

The transition from the paramagnetic nematic phase (with finite S_N) to the ferromagnetic one is then described by (2), which takes the form

$$f - f_N = \frac{1}{2}\tilde{\alpha}M^2 + \frac{1}{4}\beta M^4 \tag{12}$$

where F_N is the free energy of the paramagnetic nematic and $\tilde{\alpha} = \alpha + \tilde{\gamma}S_N + \tilde{\delta}S_N^2$. Obviously that transition is of second order and takes place at the temperature $T = T^*_{FP} - (1/\alpha_0)(\tilde{\gamma}S_N + \tilde{\delta}S_N^2)$, which is shifted from the critical para- to ferromagnetic transition temperature due to the finite nematic order. This shift can be negative as well as positive, i.e. the existing nematic order can favor or disfavor an additional ferromagnetic ordering. Again, a negative $\tilde{\delta}$ favors the ferromagnetic ordering.

Of course, there is the competing possibility of a direct transition from the isotropic ferrofluid to the ferromagnetic nematic phase. Substituting the solution (5) for the magnetic order parameter $M \neq 0$ and for ψ into the free energy (2) we get the free energy density for the ferromagnetic phase as a function of S_F alone, which can be written as

$$f - f_0 = -\frac{\alpha^2}{4\beta} - \frac{\alpha\tilde{\gamma}}{2\beta}S_F + \frac{3}{4}A^*S_F^2 - \frac{1}{4}B^*S_F^3 + \frac{9}{16}C^*S_F^4 \tag{13}$$

where the starred coefficients are defined after (6). Of the 3 possible extrema ($\frac{\partial f}{\partial S} = 0$ leading to a real solution for S_F) only those are relevant that exist within

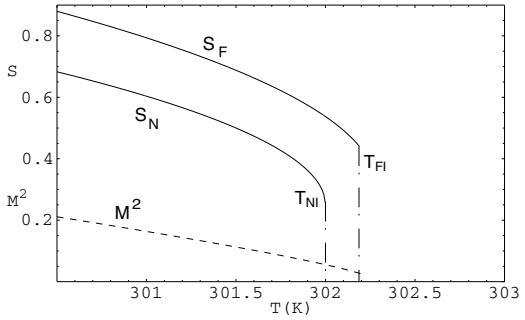

Figure 1: S and M^2 versus temperature. The upper (lower) solid line represents the orientational order parameter S_F (S_N), while the dashed line represents the (dimensionless) magnetic order parameter M^2 in the ferromagnetic phase. The values of the parameters in eq.(2) were taken to be $\alpha_0 = .1K^{-1}$, $A_0 = .1K^{-1}$, $T^*_{NI} = 301.7K$, $T^*_{FP} = 302K$, $B = .8$, $C = .53$, $\beta = 8$, $\tilde{\delta} = -.89$ (for the FI transition), $\tilde{\delta} = .16$ (for the NI transition), and $\tilde{\gamma} = -.05$ using a dimensionless free energy density f.

the stability range of the ferromagnetic phase. Although the solutions are involved, one important qualitative feature can be extracted immediately. It is obvious that a continuous IF transition ($S_F = 0 = M$ at the transition temperature) is not possible, except for the very special case $B = 0 = \gamma$ (involving $B^* = 0 = \tilde{\gamma}$) and $T^*_{NI} = T^*_{FP}$. In the general case there is a jump in S_F and M at some temperature T_{FI}. At that temperature the right hand side of (13) is zero for S_F given by eq.(6). Of course, T_{FI} has to be larger than T_{NI} for the direct transition to happen. Such a case is shown in Fig. 1 by choosing an appropriate set of parameters and numerically solving (6). Fig.1 shows that both order parameters, S_F and M jump simultaneously at the FI transition point. We also see that orientational order in the ferromagnetic phase is much higher than in the (superparamagnetic) nematic phase.

It is interesting to note that this phase transition is isomorphic to the direct isotropic to smectic A transition for ordinary liquid crystals, where the role of the magnetic order is played by the smectic order [23].

3. Phase Transitions in an External Field An external field induces a finite magnetization in the ferrofluid by orienting the magnetic particles. In addition it also orients the mesogens due to the diamagnetic anisotropy effect. Thus, any phase has a finite S and a finite M due to the external field. This is also obvious from the Landau free energy including the external field $\boldsymbol{H}$

$$f_H = f - \boldsymbol{M}\cdot\boldsymbol{H} - \frac{\chi_S}{2} H_i H_j Q_{ij} + \frac{\delta_3}{2}\boldsymbol{H}^2 Q_{ij}Q_{ij} + \frac{\delta_4}{2} H_i H_k Q_{ij} Q_{kj} \qquad (14)$$

with f the field-free energy (1). The angle between the magnetization and the field is dominated by the ferromagnetic coupling $-\boldsymbol{M}\cdot\boldsymbol{H}$ with $\boldsymbol{M}$ parallel to $\boldsymbol{H}$ being the ground state. Thus, the angle ψ between $\boldsymbol{M}$ and $\hat{\boldsymbol{n}}$ is also the angle between $\boldsymbol{H}$ and $\hat{\boldsymbol{n}}$. Then the diamagnetic coupling ($\sim \chi_S$) and the higher coupling term ($\sim \delta_4$) have the same ψ-dependence as the γ- and δ_2 terms in (2) leading again to the free energy extrema at $\psi = 0$ or $\pi/2$. The former (latter) is the stable minimum for $[(\delta_2 S + 2\gamma)M^2 + (\delta_4 S - 2\chi_S)H^2]S < 0$ (> 0). The latter case seems to be frequent in discotic lyotropic [19] and rod-like thermotropic nematic systems [20].

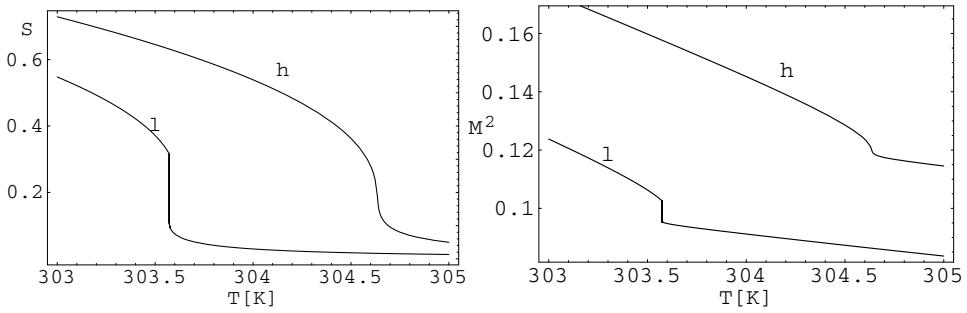

Figure 2: $S(T)$ and $M^2(T)$ according to eqs. (15,16) for two different values of the external magnetic field with $\hat{\boldsymbol{n}}$ and $\boldsymbol{M}$ parallel. The transition to the ferromagnetic state ($\tilde{\delta} = -.89$) is discontinuous for (l) lower fields ($H = .28$) and becomes smooth for (h) higher fields ($H = .41$). We have chosen $\chi_S = .05$ and $\delta_3 + (2/3)\delta_4 = -.89$; all other parameters are as in Fig.1.

Minimizing then (14) with respect to S and M leads to the coupled equations for $S(H)$ and $M(H)$

$$0 = -\tilde{\chi}_S H^2 + 3\tilde{A}S - \frac{3}{2}BS^2 + \frac{9}{2}CS^3 + 2\tilde{\delta}M^2 S + \tilde{\gamma}M^2 \tag{15}$$

$$0 = -H + \alpha M + \beta M^3 + \tilde{\delta}MS^2 + \tilde{\gamma}MS \tag{16}$$

with $\tilde{A} = A + H^2(\delta_3 + (2/3)\delta_4)$ and $\tilde{\chi}_S = \chi_S$ for $\psi = 0$, and $\tilde{A} = A + H^2(\delta_3 + (1/6)\delta_4)$ and $\tilde{\chi}_S = (-1/2)\chi_S$ for $\psi = \pi/2$. Actually there is a third possibility for the orientation of $\hat{\boldsymbol{n}}$ and $\boldsymbol{M}$ with respect to the external field $\boldsymbol{H}$: None is parallel to another one, but all three lie in a common plane separated by the angles ψ and ϕ (between $\boldsymbol{M}$ and $\boldsymbol{H}$). These angles are determined by the relations $HM\sin\phi = (3/8)SM^2(\delta_2 S + 2\gamma)\sin 2\psi = (3/8)SH^2(\delta_4 S - 2\chi_S)\sin 2(\phi - \psi)$. Again, $S(H)$ and $M(H)$ follow from equations of the form (15,16), where the coefficients, however, are very complicated. Since such a case has not been found in experiment, we will not discuss it further.

Obviously there is no solution of (15,16) with $S = 0$ and/or $M = 0$ for finite magnetic field. Instead, even the high temperature phase shows a small but finite nematic as well as magnetic order both induced by the external field. For the transition to the ferromagnetic nematic phase, where M aquires a spontaneous contribution in addition to the field-induced one, both jumps (in M and S) are reduced (and shifted to higher temperatures) and eventually, above some strong external fields, are replaced by smooth passages from low to high values (Fig. 2). This field dependence of S resembles the isotropic-nematic transition in the presence of an external *electrical* field [24]. In conventional nematics this behavior under an external magnetic field has never been observed, since the strong fields necessary are outside the experimental reach, while in ferronematics the expectation is that the necessary magnetic fields are much smaller and thus this effect is detectable.

The transition to the superparamagnetic nematic state (for positive coupling coefficients $\delta_{1,2,3,4}$) is again characterized by a jump from small S to a larger value of S, where the jump decreases with increasing field and eventually vanishes leading to a smooth passage from a low S to a high S state. For large coupling coefficients the transition temperature is shifted to lower values and at the transition the field-induced value of M is reduced due to the coupling to S, either discontinuously (low fields) or smoothly (large fields). For smaller coupling coefficients the transition

temperature is almost field-independent and M hardly affected by the change of S.

In an external magnetic field the superparamagnetic and the ferromagnetic nematic phases are identical in their symmetry properties and differ only quantitatively in the value of M. Thus, a clear distinction is best made at vanishing external field.

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