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Liquid Crystals

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713926090>

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Online publication date: 06 August 2010

To cite this Article Teixeira, P. I. C.(1998) 'An old model for magnetic nematics', *Liquid Crystals*, 25: 6, 721 – 726

To link to this Article: DOI: 10.1080/026782998205732

URL: <http://dx.doi.org/10.1080/026782998205732>

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An old model for magnetic nematics

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(Received 12 May 1998; accepted 31 July 1998)

A mean-field treatment is given of the off-lattice Krieger–James model of ordered fluids, which reduces to the more familiar Maier–Saupe liquid crystal (Heisenberg fluid) in the absence of ferromagnetic (nematic) interactions. As in the lattice version, isotropic, nematic and ferromagnetic nematic phases are found, but the nematic–ferromagnetic transition can either change order at a tricritical point, or terminate at a critical end point on the ferromagnetic–isotropic coexistence curve. In addition it is argued that the sequence of phase diagram topologies, as a function of the relative weights of ferromagnetic and nematic contributions to the free energy, should be similar to that obtained on varying the elongation of dipolar spheroids.

1. Introduction

It is often the case in the history of science that a model or theory should be formulated more than once, but named after one of its later, rather than original, creators [1]. Thus it was with Krieger and James [2]. They introduced a simple model of continuously-rotating spins on a lattice, with only nearest-neighbour interactions of the form

$$E(\theta_{ij}) = -AP_1(\cos \theta_{ij}) - BP_2(\cos \theta_{ij}) \quad (1)$$

where A and B are positive constants, θ_{ij} is the angle between the spin directions at sites i and j , and $P_n(x)$ is the n th Legendre polynomial. This was designed to describe orientational transitions in solid molecular crystals, but to the post-1970s liquid crystal (LC) theorist it is immediately recognizable as a generalization of the Lebwohl–Lasher (LL) model of a nematic LC, to which it reduces in the absence of ferromagnetic coupling ($A = 0$) [3]. In sharp contrast to the attention lavished on LL by simulators [3–8] and theorists [9, 10] alike, the Krieger–James (KJ) model has languished in relative obscurity. In their original paper, Krieger and James computed its phase diagram, latent heats and specific heats using the mean-field (MF) approximation; it is straightforward to identify their ‘even’ phase as a nematic (N) and their ‘mixed’ phase as a polar nematic—or ferromagnetic nematic (FN), as I shall call it in the remainder of this paper. They found that the transitions from the isotropic (I) or N phases into the FN phase could be first order or continuous, depending on the ratio B/A (the I–N transition being always first order). Estimates of the crossover temperatures between these

different regimes were refined by means of a variant of the Bethe approximation. Subsequently, Madhusudana and Chandrasekhar [11] again applied the MF and Bethe approximations to the same system to study the effect of dipolar forces (mimicked by $A < 0$) on the N phase; these authors were mostly concerned with characterizing local and global orientational order, and did not calculate the phase diagram. Lei [12] proposed equation (1) as a suitable model potential for bowtie LCs (where columnar structures with up–down asymmetry form due to molecular shape), but he merely quoted, and did not expand on, Krieger and James’s results. The renormalization analyses of Margaritis *et al.* [13] and Schoenmaker and Ruijgrok [14] yielded a continuous I–N transition in three dimensions, which casts some doubt on their reliability. More recently, Zannoni and co-workers [15, 16] mapped out the phase diagram of the KJ lattice fluid by Monte Carlo simulation and two-site cluster theory, thereby confirming the general topology of Krieger and James’s paper. Finally, the same three phases occur in a two-dimensional version of the KJ model introduced by Korshunov [17] and by Lee and Grinstein [18]: here the I–N and I–F transitions are Kosterlitz–Thouless, whereas N–F is of the Ising type. Sluckin and Ziman [19] mapped this problem onto a quantum spin chain and solved it using the transfer matrix formalism, finding behaviour consistent with the predicted phase diagram.

In this paper I consider the *off-lattice* KJ model, i.e. where an additional order parameter is introduced by allowing the density to vary. The motivation is twofold. Firstly, this is the natural continuation of a series of theoretical and simulation papers on the ferromagnetic

Heisenberg fluid (FHF) [20–26] and the Maier–Saupe LC (MSLC) [26–28], which are special cases of KJ with $B = 0$ or $A = 0$, respectively. It thus ties together the two strands, of lattice studies of the full model on the one hand, and of continuum, but less general, models on the other. Because both the FHF and the MSLC exhibit rich phase behaviour including critical behaviour, the same is *a fortiori* to be expected of the KJ fluid. Secondly, it is hoped hereby to provide a simple, but *microscopic*, description of the ‘ferronematics’ proposed by de Gennes [29] and since fabricated [30, 31], which consist of elongated magnetic particles dispersed in a nematic medium. In this first approach, treatment is restricted to the ‘one-component’ case: experimentally, this corresponds to the limit where the average nematic orientation follows that of the magnetic grains and there is no fractionation between coexisting phases. Furthermore, it is assumed that ferromagnetism is due to short ranged (exchange) interactions (i.e. that polarity is weak). In the spirit of the FHF, these are taken to have a much simpler orientational dependence than the long ranged dipolar forces; complex domain structures such as those obtained in, e.g. [32] will therefore be absent.

This paper is organized as follows: in §2 a MF expression is derived for the free energy density of the orientationally ordered (but positionally disordered) KJ fluids. Results are presented in §3 and compared with those for the lattice system. Finally in §4 the findings are summarized and their relation to the phase behaviour of dipolar spheroids, which exhibit similar phase diagram topologies, discussed.

2. Theory

Let us endow the KJ fluid with the pair potential

$$\phi_{\text{KJ}}(\mathbf{r}_{12}, \omega_1, \omega_2) = \begin{cases} +\infty & \text{if } r_{12} \leq \sigma \\ -A \left(\frac{\sigma}{r_{12}} \right)^6 P_1(\cos \theta_{12}) & \\ -B \left(\frac{\sigma}{r_{12}} \right)^6 P_2(\cos \theta_{12}) & \text{if } r_{12} > \sigma \end{cases} \quad (2)$$

where \mathbf{r}_{12} is the intermolecular vector of length r_{12} , $\omega_i = (\phi_i, \theta_i, \chi_i)$ is the set of orientational coordinates (Euler angles) of molecule i , and σ is the diameter of the (spherical) hard core. In this paper we shall take $A > 0$, $B > 0$, which are appropriate to a ferromagnetic nematic. As we shall work within the MF approximation, the detailed r_{12} -dependence of $\phi_{\text{KJ}}(\mathbf{r}_{12}, \omega_1, \omega_2)$ is immaterial, provided it is short ranged; the above choice is made for consistency with earlier work (see [26] and references therein). It is important to note at this stage that the

present theory is cast in thermotropic language, although most experimentally studied ferronematics [30, 31] are lyotropics (thermotropics tend to phase separate when doped [33]). In this context B should be interpreted as a measure of molecular shape anisotropy, see [27] and further discussion in §4.

In the spirit of perturbation theory, the intermolecular potential is written as

$$\phi_{\text{KJ}}(\mathbf{r}_{12}, \omega_1, \omega_2) = \phi_{\text{ref}}(\mathbf{r}_{12}) + \phi_{\text{p}}(\mathbf{r}_{12}, \omega_1, \omega_2) \quad (3)$$

where $\phi_{\text{ref}}(\mathbf{r}_{12})$ is the hard sphere (HS) ‘reference’ part, and $\phi_{\text{p}}(\mathbf{r}_{12}, \omega_1, \omega_2)$ the longer ranged ‘tail’. The MF Helmholtz free energy is then

$$\begin{aligned} \mathcal{F}[\rho(\mathbf{r}, \omega)] &= \mathcal{F}_{\text{ref}}[\rho(\mathbf{r}, \omega)] + \frac{1}{2} \int d\mathbf{r}_1 d\omega_1 d\mathbf{r}_2 d\omega_2 \\ &\times \rho(\mathbf{r}_1, \omega_1) \phi_{\text{p}}(\mathbf{r}_{12}, \omega_1, \omega_2) \rho(\mathbf{r}_2, \omega_2) \end{aligned} \quad (4)$$

where $\rho(\mathbf{r}, \omega)$ is the density–orientational profile. The free energy of the reference system, $\mathcal{F}_{\text{ref}}[\rho(\mathbf{r}, \omega)]$, consists of ideal-gas and excess contributions, the latter due to the excluded volume interaction:

$$\begin{aligned} \mathcal{F}_{\text{ref}}[\rho(\mathbf{r}, \omega)] &= \beta^{-1} \int d\mathbf{r} d\omega \rho(\mathbf{r}, \omega) \{ \log[\Lambda^3 \rho(\mathbf{r}, \omega)] - 1 \} \\ &+ \mathcal{F}_{\text{HS}}^{\text{exc}}[\rho(\mathbf{r})] \end{aligned} \quad (5)$$

where $\beta = 1/k_{\text{B}}T$, Λ is the thermal de Broglie wavelength, $\rho(\mathbf{r}) = \int d\omega \rho(\mathbf{r}, \omega)$, and the orientational entropy is described in the random-mixing approximation. In a uniform fluid, the excess of free energy can be treated in a local approximation:

$$\mathcal{F}_{\text{HS}}^{\text{exc}}[\rho(\mathbf{r})] = \int d\mathbf{r} \rho(\mathbf{r}) \Psi[\rho(\mathbf{r})] \quad (6)$$

with $\Psi(\rho)$ an excess HS free energy per particle, to be specified later.

The free energy density (FED) of an orientationally ordered, but positionally disordered, phase of density ρ , is now†

$$\begin{aligned} f(\rho, \eta) &= f_{\text{hs}}(\rho) + \rho \beta^{-1} \langle \log[4\pi \hat{f}(\omega)] \rangle \\ &- \frac{1}{2} A_{\text{int}} \rho^2 \eta_1^2 - \frac{1}{2} B_{\text{int}} \rho^2 \eta_2^2 \end{aligned} \quad (7)$$

where $f_{\text{hs}}(\rho) = \rho \beta^{-1} [\log(\Lambda^3 \rho) - 1] + \rho \Psi(\rho)$, $\hat{f}(\omega) = \rho(\omega)/\rho$ is the orientational distribution function (ODF), $\langle \cdot \rangle = \int d\omega \mathcal{A} \hat{f}(\omega)$, and $A_{\text{int}} = 4\pi\sigma^3 A/3$, $B_{\text{int}} = 4\pi\sigma^3 B/3$ are the integrated strengths of the ferromagnetic and nematic parts of the potential, respectively. In equation (7), η_1 and η_2 are, respectively, the polar and nematic orientational

† See [22, 27] for details, as our $\phi_{\text{p}}(\mathbf{r}_{12}, \omega_1, \omega_2)$ is a superposition of those used in either reference.

order parameters, defined as

$$\eta_1 = \int P_1(\cos \theta) \hat{f}(\omega) d\omega \quad (8)$$

$$\eta_2 = \int P_2(\cos \theta) \hat{f}(\omega) d\omega. \quad (9)$$

Again for consistency with previous work [27, 28], we take for $\Psi(\rho)$ the Percus–Yevick (compressibility) expression [34, 35]:

$$\beta\Psi(\rho) = -\log(1 - \xi) + \frac{6\xi - 9\xi^2 + 3\xi^3}{2(1 - \xi)^3} \quad (10)$$

where $\xi = \pi\rho\sigma^3/6$ is the packing fraction. Minimizing the FED with respect to the ODF (see, e.g. [27]), we obtain

$$\hat{f}(\omega) = Z^{-1} \exp \{ \beta\rho [A_{\text{int}}\eta_1 P_1(\cos \theta) + B_{\text{int}}\eta_2 P_2(\cos \theta)] \} \quad (11)$$

$$Z = \int \exp \{ \beta\rho [A_{\text{int}}\eta_1 P_1(\cos \theta) + B_{\text{int}}\eta_2 P_2(\cos \theta)] \} d\omega. \quad (12)$$

The equilibrium pressure p and chemical potential μ are likewise given by

$$p = p_{\text{hs}} - \frac{1}{2}\rho^2(A_{\text{int}}\eta_1^2 + B_{\text{int}}\eta_2^2) \quad (13)$$

$$\mu = \mu_{\text{hs}} + \beta^{-1} \log \left(\frac{4\pi}{Z} \right) \quad (14)$$

p_{hs} , μ_{hs} being, respectively, the pressure and chemical potential of the HS system. In the I phase $\eta_1 = \eta_2 = 0$, $Z = 4\pi$ and equations (13) and (14) reduce to $p = p_{\text{hs}}$, $\mu = \mu_{\text{hs}}$.

Because ferromagnetic ordering can set in via a continuous transition [2], it is important to look at the stability of the I or N phases with respect to FN fluctuations. This is done by searching for the lowest densities at which equation (8) acquires a non-trivial (i.e. $\eta_1 \neq 0$) solution. Substitution of equation (11) into (8) yields, by straightforward algebra:

$$\eta_1 = \frac{2}{3\varepsilon_B} \frac{\exp \left[\frac{3}{2}\varepsilon_B(\lambda^2 + 1) \right] \sinh \varepsilon_A}{\mathcal{J} \left(\frac{3}{2}\varepsilon_B, 1 + \lambda \right) + \mathcal{J} \left(\frac{3}{2}\varepsilon_B, 1 - \lambda \right)} - \lambda \quad (15)$$

where $\varepsilon_A = \beta\rho A_{\text{int}}\eta_1$, $\varepsilon_B = \beta\rho B_{\text{int}}\eta_2$, $\lambda = \varepsilon_A/(3\varepsilon_B)$, $\mathcal{J}(x, y) = \exp(xy^2)\mathcal{J}(y\sqrt{x})/\sqrt{x}$, and $\mathcal{J}(x) = \exp(-x^2)\int_0^x \exp(t^2)dt$ is Dawson's integral. Expanding the right hand side of equation (15) about $\eta_1 = 0$, we find the implicit equation

for the ferromagnetic instability density ρ_{inst} :

$$\beta\rho_{\text{inst}}A_{\text{int}} \left[\eta_2(\rho_{\text{inst}}) + \frac{1}{2} \right] = \frac{3}{2} \quad (16)$$

which reduces to the usual Curie line if $\eta_2 = 0$.

3. Results

Phase diagrams have been calculated by equating pressures and chemical potentials of the ordered (N or FN) and disordered (I) phases, equations (13) and (14). All results are given in terms of the reduced density $\rho^* = \rho\sigma^3$, the reduced temperature $T^* = k_B T/(A + B)$, and the ratio $R = B_{\text{int}}/A_{\text{int}} = B/A$ of nematic to ferromagnetic interaction strengths. Figures 1–4 illustrate the six different topologies obtained.

The same four types of transition have been identified as in Krieger and James's original study of the *lattice* model [2]: a continuous order–disorder transition between FN and I phases; continuous or first order magnetic ordering transitions between the N and FN phases; and a first order transition between I and FN phases. At small R , corresponding to an interaction potential with a large ferromagnetic admixture, only two phases, I and FN, are present: the I–FN transition is first order at low temperatures and becomes continuous at a tricritical point (tcp), see figure 1; this corresponds to range I in [2]. As R is increased, the tcp moves to higher and higher densities and temperatures, until the transition

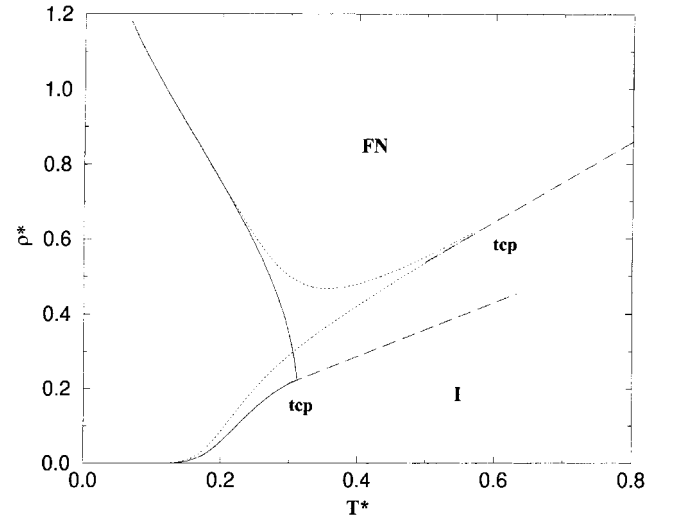


Figure 1. Phase diagram of the KJ fluid for $R = B/A = 0$ (solid lines) and $R = 0.5$ (dotted lines); the dashed lines are the Curie lines (loci of continuous order–disorder transitions). $\rho^* = \rho\sigma^3$ and $T^* = k_B T/(A + B)$ are the reduced temperature and reduced density, respectively. I=isotropic phase; FN=ferromagnetic nematic phase; tcp=tricritical point. Increasing R destabilizes the FN phase and derives the tcp to higher (reduced) temperatures and densities.

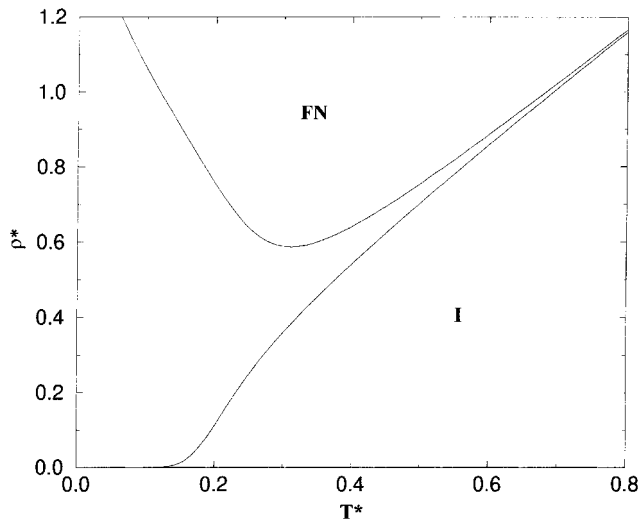
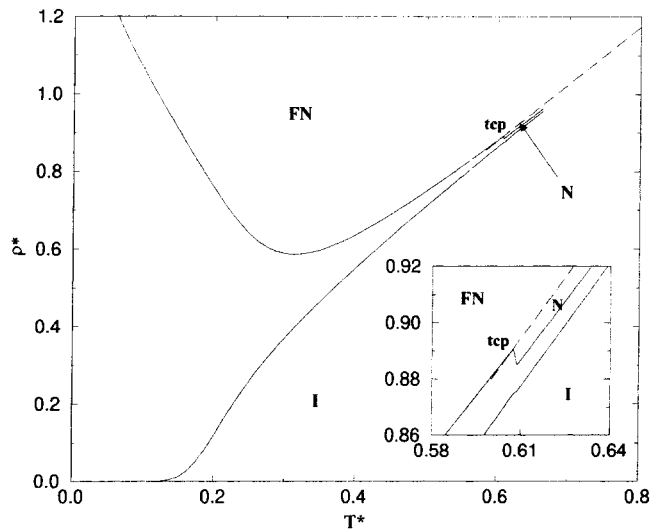
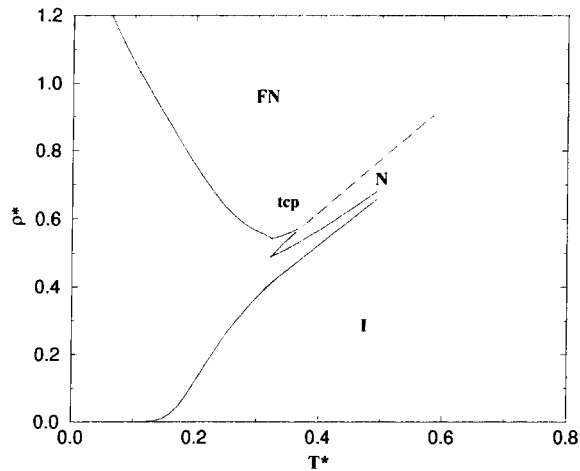


Figure 2. Phase diagram of the KJ fluid for $R = B/A = 2$. $\rho^* = \rho\sigma^3$ and $T^* = k_B T / (A + B)$ are the reduced temperature and reduced density, respectively. I = isotropic phase; FN = ferromagnetic nematic phase. The tricritical point (tcp) has disappeared, and consequently the I–N transition is everywhere first order (i.e. it is driven by nematic, rather than ferromagnetic, ordering).

is first order everywhere (or at least as far out as the coexistence equations could be solved, which was well beyond the normal range of liquid densities), see figure 2. Thus for $0.58 \lesssim R \lesssim 2.9$ we find ourselves in ranges II–III of [2]. If $R \lesssim 2.9$, a N phase appears at high temperatures, where on raising the density we first go from I to N via a first order transition, and then from N to FN via a continuous transition, see figure 3; at lower temperatures there is a direct (first order) transition from I to FN (range IV in [2]). The N–FN transition line terminates at a different tcp, below which there extends a narrow N–FN coexistence region (leading to an I–N–FN triple point). This tcp moves to lower temperatures and densities on increasing R further, until at $R \sim 6.93$ it transforms into a critical endpoint (cep) on the I–FN coexistence curve (see figure 4). For $R \lesssim 6.93$ we are in range V of [2], where increasing R moves the cep, and consequently the FN phase it bounds, to ever higher densities and ever lower temperatures, until at $R = \infty$ we are left with I and N phases only. The crossover values of R are collected, and contrasted with those of [2], in the table. Agreement is almost perfect for small R , which probably reflects the fact that the tcp occurs at high densities, where a continuum and a lattice model differ little. Note, however, that some of the action might be pre-empted by the formation of solid phases [24], which have not been considered here; on the basis of previous work, we expect the FN liquid to become unstable relative to the orientationally ordered solid for $\rho^* \sim 0.7$ [26].



(a)



(b)

Figure 3. Phase diagram of the KJ fluid for (a) $R = B/A = 3$ and (b) $R = 3.5$; the dashed lines are the Curie lines (loci of continuous order–disorder transitions). $\rho^* = \rho\sigma^3$ and $T^* = k_B T / (A + B)$ are the reduced temperature and reduced density, respectively. I = Isotropic phase; N = nematic phase; FN = ferromagnetic nematic phase; tcp = tricritical point. A new tcp has appeared (see inset of (a) for detail), where now the N–FN transition (as opposed to I–FN in figure 1) changes order; notice the (very narrow) regions of N–FN coexistence between the tcp and the I–N–FN triple point. For $R = 3.5$ the tcp is well within the range of typical liquid densities.

4. Discussion and conclusions

The phase diagram of the KJ fluid has been computed for different ratios R of the strength of nematic to ferromagnetic forces. The same topologies have been found as for the lattice model [2], but in addition we have been able to identify two types of critical points.

Table. Topology of the phase diagram of the KJ fluid from MF theory. I = isotropic; N = nematic; FN = ferromagnetic nematic; tcp = tricritical point; cep = critical end point. The extreme left and extreme right columns are Krieger and James's [2] nomenclature and results for the lattice version of the model: agreement is excellent at large R , corresponding to high tcp densities. At lower densities, a fluid contains much more entropy than a lattice, driving up the cross over R between ranges IV and V.

Range [2]	Ordering transition(s)	This work	Krieger and James [2]
I	I to FN, changes order at tcp	$0 < R < 0.58$	$0 < R < 0.56$
II, III	I to FN, first order	$0.58 < R < 2.9$	$0.56 < R < 2.89$
IV	$\left\{ \begin{array}{l} \text{I to N, first order;} \\ \text{N to FN, changes order at tcp;} \\ \text{I to FN, first order} \end{array} \right\}$	$2.9 < R < 6.93$	$2.89 < R < 3.21$
V	$\left\{ \begin{array}{l} \text{I to N, first order;} \\ \text{N to FN, continuous, ends at cep;} \\ \text{I to FN, first order} \end{array} \right\}$	$R > 6.93$	$R > 3.21$

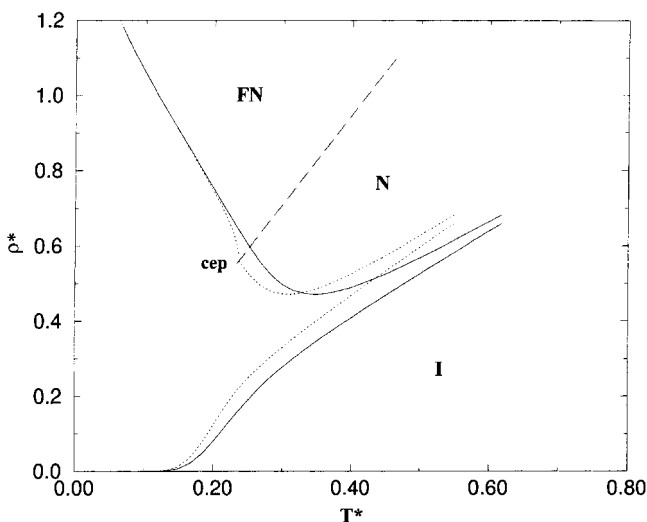


Figure 4. Phase diagram of the KJ fluid for $R = B/A = \infty$ (solid lines) and $R = 8$ (dotted lines); the dashed lines are the Curie lines (loci of continuous order-disorder transitions). $\rho^* = \rho\sigma^3$ and $T^* = k_B T / (A + B)$ are the reduced temperature and reduced density, respectively. I = isotropic phase; N = nematic phase; FN = ferromagnetic nematic phase; cep = critical end point. As R is increased, the cep moves up the FN branch of the I-FN coexistence to ever higher densities and lower temperatures, until the FN phase disappears at $R = \infty$ (in practice it would be pre-empted by a solid phase before that).

In this preliminary study treatment has been restricted to the simplest MF approximation, which has been shown [15, 16] correctly to describe the general features of the phase behaviour of the *lattice* KJ model. One expects, however, that fluctuations will carry more weight in the *continuum* fluid, hence simulations of this system would be desirable. Furthermore, no attempt has been made to calculate solid phase boundaries; results of earlier work [26] suggest nevertheless that all transitions predicted here, as well as the cross over from

tcp to cep criticality, should be (at least partially) within the liquid range.

One final point concerns the similarity between the above results and those reported by Groh and Dietrich for two fluids of dipolar non-spherical particles [36]. In their paper the effect of increasing particle elongation κ has almost the same effect on the phase diagram as we obtain by increasing R . Differences are the presence in their work of *two* tcp's and a re-entrant N phase at low temperatures (although the latter could be an artifact of the approximations used). Moreover it is not clear whether they see a cep or not. Let us address this issue by examining the stability of the I phase. According to Stecki and Kloczkowski [37], the condition of absolute stability of the I phase with respect to orientational fluctuations of symmetry l (where $l = 1$ corresponds to a ferromagnetic and $l = 2$ to a nematic) is

$$1 - \rho(2l + 1)^{-1/2} \int_0^{+\infty} d\mathbf{r}_{12} c^{ll_0}(r_{12}) = 0 \quad (17)$$

where $c^{ll_0}(r_{12})$ is a coefficient in the rotational invariant expansion of the direct correlation function (DCF), $c(\mathbf{r}_{12}, \omega_1, \omega_2)$. Within our MF theory, $c(\mathbf{r}_{12}, \omega_1, \omega_2) = -\beta\phi_{\text{KJ}}(\mathbf{r}_{12}, \omega_1, \omega_2)$ and it is immediately seen that increasing R lowers the instability limit with respect to the N phase, relative to that with respect to the FN phase. Likewise, although Groh and Dietrich use a different approximation for the DCF, their equation (24) has the same form as our equation (7), and the above discussion also applies. When κ is raised or lowered from 1, u_2 (playing the role of our B_{int}) must grow relative to u_1 (playing the role of our A_{int}) [37, equation (25)] as the molecule becomes less and less spherical, thus favouring the (non-polar) N instability of the I phase. In this sense, we can interpret our B as a measure of molecular asphericity. Crucially, the bulk phase behaviour (i.e. involving macroscopically homogeneous

phases only) is determined by the short range parts of the intermolecular interactions, while the long range contributions, if present, are responsible for domain formation.

I thank J. M. Tavares for a critical reading of the manuscript, and T. J. Sluckin for bringing refs [17–19] to my attention. Financial support from the EPSRC (UK) is gratefully acknowledged.

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