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Scaling theory of the paranematic–nematic critical point

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A thermodymanic scaling analysis of the paranematic–nematic critical point is presented. Although presumed to be in the Ising universality class, general arguments suggest that the order parameter approaches the critical point in an asymmetrical manner in contrast to mean-field predictions.

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Upon lowering of the temperature in an orientationally isotropic liquid phase of nematic molecules, a first-order phase transition to an anisotropic liquid phase is possible. This isotropic–nematic phase transition involves an orientational symmetry change and as a result only an isolated critical point on the first-order transition line, known as the Landau point, is possible [1–3]. This multicritical point has been observed in certain lyotropic materials [4]. The application of a uniform electric (or magnetic) field to a uniaxial nematic fluid which has positive dielectric anisotropy (or positive diamagnetic anisotropy in the magnetic case) induces the thermally averaged local direction of the molecules to be parallel to the external field direction. The zero-field isotropic phase becomes paranematic, and a line of first-order paranematic–nematic transitions develops in the temperature, external field plane. Such a coexistence curve has been observed experimentally [5]. Since there is no symmetry change in going from a paranematic liquid to a nematic liquid, the possibility exists for gradual weakening of the first-order transition. Thus the coexistence curve in the temperature, external field plane may eventually terminate at an ordinary critical point of the liquid–gas type. Although such a scenario has yet to be achieved experimentally, there have been several theoretical investigations which predict the existence of a paranematic–nematic critical point (PNCP) within mean-field arguments [6–10]. Since it is known that critical fluctuations may drastically alter mean-field results, it is timely to discuss the PNCP from the general point of view that can be provided by the thermodynamic scaling theory of critical phenomena in anticipation of future experimental progress.

Thermodynamic scaling theory [11] for the problem of the PNCP starts with the assumption that near a critical point, the singular part of the free energy per unit volume is a homogeneous function of the appropriate scaling fields [12]. There are two relevant scaling fields since the PNCP is obtained by adjusting both the temperature T and the external field E to critical values T_c and E_c (we use E to generically denote electric or magnetic fields). An appropriate dimensionless measure of the thermal 'distance' from the PNCP is $t=(T-T_c)/T_c$. For the external field, the appropriate quantity is $h=\Delta\chi(E^2-E_c^2)/E_c^2$, where $\Delta\chi>0$ is the molecular anisotropy (dielectric or diamagnetic) of the nematic system [6]. The thermal scaling field t and external field scaling field $\mathbf{h}$ are analytic functions t and h in general, and the singular part of the free energy density takes the form

$$F_{\text{sing}}(s^{1/\nu}\mathbf{t}, s^{\Delta/\nu}\mathbf{h}) = s^{d} F_{\text{sing}}(\mathbf{t}, \mathbf{h}), \tag{1}$$

where s is an arbitrary dimensionless scale change, ν is the correlation length critical exponent, Δ is the external field critical exponent, and $d=3$ is the dimension of the system. Choosing s to be the correlation length (appropriately normalized) gives the condition, $s^{1/\nu}|\mathbf{t}|=1$. Hence, we obtain

$$F_{\text{sing}}(\mathbf{t}, \mathbf{h}) = |\mathbf{t}|^{2-\alpha}\Phi(\mathbf{h}|\mathbf{t}|^{-\Delta}), \tag{2}$$

where $d\nu = 2-\alpha$ (hyperscaling relation) defines the specific heat critical exponent, and $\Phi(x) \equiv F_{\text{sing}}(1, x)$ is a universal scaling function of $x = \mathbf{h}|\mathbf{t}|^{-\Delta}$.

In the vicinity of the PNCP, critical fluctuations occur in the *degree of anisotropy* of the paranematic and nematic phases which is quantified by a *scalar* order parameter, ψ. The direction of the anisotropy is parallel to the external field and is non-critical. This suggests the hypothesis that the critical exponents and scaling function of the PNCP belong to the $n=1$ universality class of the spin $-1/2$ Ising model [13]. A consequence of this hypothesis is that the scaling fields for a system in the Ising universality class have the important property that the coexistence curve is $\mathbf{h}=0$ for $T<T_c$.

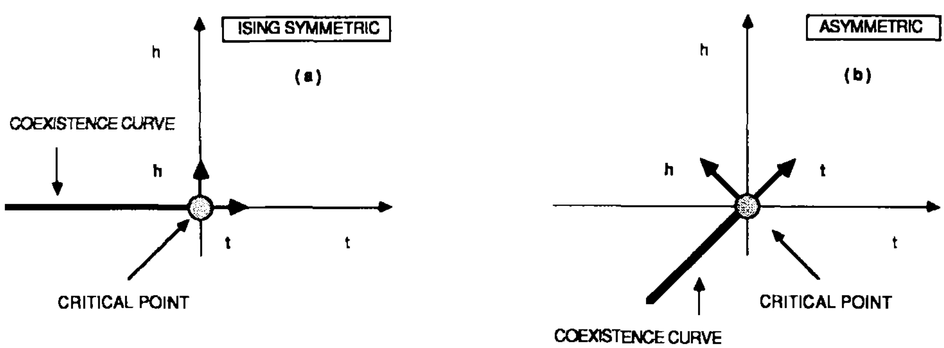
For example, a uniaxial ferromagnet (the canonical system described by the spin $-1/2$ Ising model) has $h=0$, $t<0$ as the coexistence curve in the (t, h) plane, where h is the (dimensionless) externally applied magnetic field. This is a consequence of the fact that the underlying microscopic hamiltonian is invariant under time reversal symmetry and thus is unchanged under reversal of the externally applied magnetic field, $h \to -h$. As a result, the scaling fields [12] are just $\mathbf{h}=h$, $\mathbf{t}=t$ (see the figure (a)). Hence, the thermodynamic conjugate to the external field, the magnetization (order parameter), on the coexistence curve is

$$m_{\pm} = -(\partial F_{\text{sing}}/\partial h)_{\mathbf{h}\to\pm 0}, \tag{3}$$

and we obtain

$$m_{\pm} = \pm|t|^{\beta}\Phi'(0), \tag{4}$$

where the order parameter critical exponent is $\beta = 2-\alpha-\Delta$, and $\Phi'(0) = (d\Phi/dx)_{x=0}$.



(a) Sketch of a coexistence curve terminating at a critical point in the reduced temperature (t) and reduced field (h) plane for an Ising symmetric system, such as a uniaxial ferromagnet. The bold symbols are the respective scaling fields, $\mathbf{t}$ and $\mathbf{h}$, which are identical to t and h in this case. (b) In the asymmetric case, the coexistence curve in the (t, h) plane has no particular symmetry, however, the directions of the scaling fields reflect the assumed underlying Ising universality near the paranematic–nematic critical point.

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On the other hand for the nematic system, the 'external field', which couples to the order parameter, is not proportional to the applied external electric or magnetic field, but rather its square. As a result, near the PNCP, the 'field' which couples to the order parameter is $h = \Delta\chi(E^2 - E_c^2)/E_c^2$ as previously stated. The PNCP has no symmetry between h and $-h$ since, for a given h, states with $-h$ are not possible for $\Delta\chi > 0$ and any reversal of the external field direction. As a consequence of this lack of symmetry between h and $-h$, the general form of the scaling fields near the critical point are linear combinations

$$\left.\begin{aligned} \mathbf{h} &= h + At, \\ \mathbf{t} &= t + Bh, \end{aligned}\right\} \tag{5}$$

where A and B are non-universal (system dependent) constants. See the figure (*b*). However, since the ***underlying Ising universality*** of the PNCP system requires that the coexistence curve is determined by setting the scaling field $\mathbf{h} = 0$, for $t < 0$, the paranematic–nematic order parameter,

$$\Psi_{\pm} = -(\partial F_{\rm sing}/\partial h)_{\mathbf{h} \to \pm 0}, \tag{6}$$

on the first-order line near the PNCP is

$$\Psi_{\pm} = \pm |(1-AB)t|^{\beta}\Phi'(0) + B(2-\alpha)|(1-AB)t|^{1-\alpha}\Phi(0). \tag{7}$$

Hence, the half-width of the coexistence region is

$$(\Psi_+ - \Psi_-)/2 = |(1-AB)t|^{\beta}\Phi'(0), \tag{8}$$

and the average value of the order parameter in the coexistence region is [14–16]

$$(\Psi_+ + \Psi_-)/2 = B(2-\alpha)|(1-AB)t|^{1-\alpha}\Phi(0). \tag{9}$$

This results for the half-width, equation (8), and the average value, equation (9), are consequences of fluctuations in the vicinity of the PNCP. Thus, the mean-field prediction [8–10]

$$(\Psi_+ + \Psi_-)/2 \equiv 0, \tag{10}$$

is lacking in this respect. Baring the accidental vanishing of the non-universal amplitude B, we reach the conclusion that the order parameter approaches the critical point from the paranematic and nematic phases in an asymmetrical manner and in addition develops a singularity in its average value (a diverging slope proportional to the heat capacity) at the PNCP. It is important to note that the thermodynamic scaling analysis can only suggest that there is no underlying symmetry argument requiring B to vanish identically. The existence of a non-zero B requires a detailed analysis of the interaction between the critical and non-critical degrees of freedom. Such a study will be reported in a future work.

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