# Coulombic Criticality in General Dimensions

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Debye-Hückel theory for spheres of diameter $a$ with charges $\pm q$ is extended to general dimension $d$. Explicit results include Bjerrum association of $+/-$ ions and the dipole-ionic solvation energy. For all $d > 2$ a critical point terminates conducting-liquid–conducting-vapor coexistence: The critical density $\rho_c^* = \rho_c a^d$ falls with $d \searrow 2$, but $T_c^* \equiv k_B T_c a^{d-2}/q^2$ *rises*. When $d = 2$, an *insulating* vapor appears for $T^* \leq \frac{1}{4}$ (the Kosterlitz-Thouless point), separated from conducting fluid by an infinite-order critical line ending at a *tricritical* point: $\rho_t^* = \rho_c^*(d \to 2) \simeq 0.004$, $T_t^* = T_c^*(d \to 2) \lesssim \frac{1}{4}$ for $\rho^* > \rho_t^*$ the transition is first order.

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Despite the pioneering work of Debye and Hückel (DH) [1] and Kosterlitz and Thouless (KT) [2] and many subsequent studies [3–6], a full elucidation of the phase transitions and critical behavior of a fluid, say of hard spheres, with pure Coulombic interactions remains both a theoretical [3–5] and a computational [6] challenge to statistical physics. In $d = 3$ dimensions renewed interest comes from recent experiments [7] suggesting that fluid-fluid criticality that is driven principally by Coulombic forces is of mean-field or van der Waals rather than of Ising character, or, at least, that the ultimate crossover to asymptotic Ising criticality occurs uncommonly close to the critical point and is unusually sharp [8]. In $d = 2$ dimensions, the fate of the KT insulator- or dielectric-to-conductor transition at higher ionic densities is of ongoing interest both for its own sake [5,6(a),6(b)] and for gaining insight into related magnetic, electronic, and superfluid systems [9]. Furthermore, the significance of the KT dipolar screening mechanism for $d \neq 2$ is also little explored.

At this point in the development of a full theory we believe there is a valuable role to be played by relatively simple approximate theories (as provided by mean-field analyses of magnetic transitions) that give the general structure of a phase diagram and, in particular, reveal and assess the dominant physical mechanisms. The latter should, in due course, be incorporated into more sophisticated but probably less transparent theories. In this spirit, the original DH theory [1] for the so-called *restricted primitive model* (RPM), namely, hard spherical ($d = 3$) ions of diameter $a$ in a medium of dielectric constant $D$, one-half carrying charges $+q$ the other $-q$, has recently been extended [10]. Bjerrum's concept (Bj) [3(a)] of the association of + and − ions into neutral dipolar pairs [11], which serves to correct the DH linearization of the Poisson-Boltzmann equation, was augmented by a calculation of the solvation free energy of a dipole in the screening ionic fluid (DI). The resulting DHBjDI theory [10] predicted a critical point at surprisingly low density and temperature: specifically, with further allowance for hard-core (HC) volume exclusion [10], at $\rho_c^* \simeq 0.026$ and $T_c^* \simeq 0.055$. (Here and below we take $\rho^* = \rho a^d$, where $\rho = N/V$ is the density of the *total* number of ions, associated or not, and $T^* = k_B T D a^{d-2}/q^2$.) These critical point predictions turn out to be remarkably close to recent Monte Carlo estimates [6(a),6(e),10] (although the simulations are still subject to various reservations and, in particular, say nothing about the critical exponents).

Apart from the intrinsic interest of studying Coulombic fluids for general $d$, a strong motivation is provided by the search for a suitable small parameter for the critical region, like $\epsilon$ in the $d = 4 - \epsilon$ renormalization group expansions for standard Ising systems and $1/N$ in the case of polymer solutions, see [4]. In fact, for Coulombic fluids it transpires that $d = 2$ is a marginal dimension. Furthermore, as we report here, $T_c^*(d)$ rises when $d$ falls, which suggests that the fluctuations are, in some way, playing a *reduced* role in low $d$ (the reverse of the usual situation) so that mean-field-like theories may work best in $d = 2 + \epsilon$ dimensions with $\epsilon$ small. Likewise, within *pure* DH theory we find that $\rho_c^*(d)$ decreases to *zero* as $d \to 2+$. At this stage, however, we have been unable to exploit these discoveries in a systematic way that treats all fluctuation effects.

To present our results for general $d = 2 + \epsilon$, we take Coulomb's law to be $F_{ij}(r) = q_i q_j / D r^{d-1}$ and write the pair interaction potential as [12]

$$\varphi_{ij}(r) = \frac{q_i q_j}{D\epsilon}\left(\frac{1}{r^\epsilon} - \frac{1}{a^\epsilon}\right). \tag{1}$$

When $d \to 2$, this yields $\varphi_{ij}(r) = -(q_i q_j / D)\ln(r/a)$, the expected logarithmic potential [2,5].

Now for $d = 2$ consider a neutral fluid of *pointlike* ions. (See, e.g., [11(b)], and references therein.) Via scaling, the logarithmic potential leads to the exact equation of state

$$p/\rho k_B T = 1 - T_0/T \quad \text{with} \quad T_0^* = \tfrac{1}{4}. \tag{2}$$

valid [13] for $T^* > \frac{1}{2}$, while $T_0$ is recognized as the KT temperature [2,5]. This result seems plausible asymptotically in the limit $\rho^* \equiv \rho a^d \to 0$ *even* for hard-core ions. But: "Is it a prediction of the DH theories [10] for $d = 2$?" The answer is "definitely yes!"

More generally, we follow [10] and compute the pressure $p(T, \mu_+, \mu_-, \mu_2)$ as a function of the chemical potentials $\mu_s$, for free + and − ions and bound dipolar pairs, by maximizing $f(T; \rho_+, \rho_-, \rho_2) + \sum_s \mu_s \rho_s$ over the corresponding densities $\rho_+$, $\rho_-$, and $\rho_2$. The Helmholtz free-energy density, $-f = F/V$, is constructed approximately as a sum of terms starting with

$$f^{\text{Ideal}} = k_B T \sum_s \rho_s [1 - \ln(\rho_s \Lambda_s^d / \zeta_s)], \qquad (3)$$

where $\Lambda_+ = \Lambda_- = \Lambda(T)$ is the thermal de Broglie wavelength and $\Lambda_2 = \Lambda^2$, while $\zeta_+ = \zeta_- = 1$ and $\zeta_2(T)$ represent the internal ionic and dipolar partition functions. Then a term $f^{\text{HC}}$ is added to represent the hard-core repulsions: a free-volume form [10] is sufficiently accurate at the low densities of most interest.

The DH excess ionic free energy $f^{\text{DH}}$ is calculated straightforwardly in the usual way [1,4,14,15] based on Poisson's equation written now for general $d$ as [14]

$$\nabla^2 \phi = -C_d \rho_q(\mathbf{r})/D, \quad C_d = 2\pi^{d/2}/\Gamma\left(\tfrac{1}{2}d\right). \qquad (4)$$

The standard arguments [1,4,15] yield the Debye screening length $\xi_D = 1/\kappa(T, \rho_1)$ via

$$\kappa^2 = C_d q^2 \rho_1 / D k_B T \quad \text{with} \quad \rho_1 = \rho_+ + \rho_-. \qquad (5)$$

Next, solution of the appropriate cylindrically symmetric general $d$ Helmholtz DH equations [4,10,14,16] using modified Bessel functions, $K_\nu(x)$ yields the electrostatic energy (within DH theory) of a typical ion *or* dipole [16]. Finally, the Debye charging process gives the purely ionic DH free energy [12] via

$$C_d a^d \beta f^{\text{DH}} = \ln[C_d (x/2\pi)^{d/2} K_{d/2}(x)], \qquad (6)$$

where $\beta = 1/k_B T$ and $x = \kappa a$: for $d = 3$, this reproduces the well-known result [1,4,10,15]. Likewise [10,14,16] with

$$\begin{aligned}\omega_2(x) &= \frac{d}{x^4} \ln\left[ C_{d+2} \left(\frac{x}{2\pi}\right)^{1+d/2} K_{1+d/2}(x) \right] + \frac{1}{2x^2} \\ &= \{1 - x^\epsilon [1 + O(\epsilon)]\}/4d\epsilon + O(x^2), \end{aligned} \qquad (7)$$

the dipole-ion (DI) contribution is found to be

$$\beta f^{\text{DI}} = \rho_2 (\kappa^2 a^{2+\epsilon} / a_2^\epsilon T^*) \omega_2(\kappa a_2) \sim \rho_2 \rho_1 / T^2, \qquad (8)$$

where [10,12,16] $a_2 \equiv (1 + s_2) a$ is the radius of the effective spherical cavity enclosing a dipolar pair and excluding the centers of other ions. As explained previously [10], we may estimate $a_2$ via an angular average which yields $s_2 \simeq 0.2525 - 0.1233\epsilon + 0.0328\epsilon^2$ for $0 \lesssim \epsilon \lesssim 1$ [14]. However, our principal results are not sensitive to $s_2$.

To allow for ion-pair formation we need the *association constant* $K(T) \equiv \zeta_2(T)/\zeta_+ \zeta_-$, see (3). Following, as before [10], Ebeling's careful analysis [3(c),17] for $K(T)$ when $d = 3$, we adopt Bjerrum's original proposal [3(a),10] which should be fully adequate at *low* $T^*$ ($< 1/d$): We thence derive [14]

$$K(T, d) = \frac{C_d a^d T^*}{1 - dT^*} \left[ 1 + \frac{\epsilon T^*}{(1 - dT^*)^2} + \cdots \right], \qquad (9)$$

the ellipsis being of order $(\epsilon T^*)^2$ and $(T^*)^{1/\epsilon T^*}$: In fact, the truncated expansion proves adequate in the critical region, to within 3%, even for $d = 3$ [14]. Equilibrium between ions and dipoles at an *overall* density $\rho = \rho_1 + 2\rho_2$ is ensured by $\mu_+ + \mu_- = \mu_2$, which leads to

$$\rho_2 = \tfrac{1}{4} \rho_1^2 K(T) \exp(2\beta \mu_1^{\text{ex}} - \beta \mu_2^{\text{ex}}), \qquad (10)$$

where the excess chemical potentials are $\mu_i^{\text{ex}}(\rho_1, \rho_2) = -\partial f^{\text{ex}}/\partial \rho_i$ ($i = 1, 2$), while $f^{\text{ex}} = f^{\text{DH}} + f^{\text{DI}} + f^{\text{HC}}$.

In summary, all thermodynamic properties follow directly from (3), (5)–(10). Figure 1 shows results for pure DH theory [supplemented, in (a), by hard-core terms]. As $d \to 2+$, the coexistence curve approaches a sharply cusped form in which the $d = 2$ vapor phase resides only at *zero* density and pressure, while the coexisting liquid density is described by

$$\rho_{1\text{liq}}^*(T) \approx \frac{t'(1 - t')}{\pi \ln^2 \{(t_0/t') \ln^2 [(t_0/t') \ln^2 (t_0/t')]\}}, \qquad (11)$$

where $t' = (T_c - T)/T_c$ and $t_0 = \frac{1}{2} \exp(\frac{1}{2} - 2\gamma_E) \simeq 0.26$. The critical temperature varies with $d$ as

$$T_c^* = \tfrac{1}{4} - \tfrac{1}{8} \epsilon \left( |\ln \tfrac{\epsilon}{2}| + 2 \ln |\ln \tfrac{\epsilon}{2}| - 2\gamma_E + \cdots \right), \qquad (12)$$

when $\epsilon \to 0$, while the critical density vanishes as

$$\rho_c^* = \epsilon / 4\pi \left[ \ln \frac{\epsilon}{2} - 2 \ln |\ln \tfrac{\epsilon}{2}| + 2\gamma_E + 1 + \cdots \right]^2, \qquad (13)$$

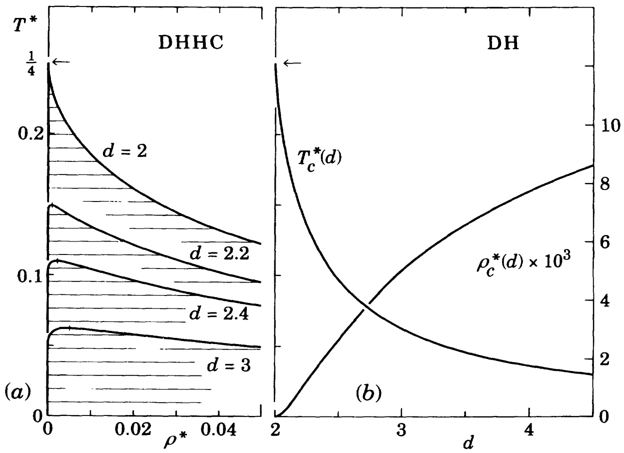

FIG. 1. (a) Coexistence curves in general $d$ for Debye-Hückel theory (with hard-core terms); (b) critical temperature and density (right- and left-hand scales) vs $d$ for pure DH theory.

see Fig. 1(b). For $d = 2$ one sees from the appropriate Landau-type expansion that the critical point is actually *tricritical* in nature (as in $N \rightarrow \infty$ polymer solution theory, see [4]); this is consistent also with the criticality of the line $\rho = 0$ (implying $\kappa \equiv 0$), for $T_c^* > \frac{1}{4}$. When $d < 2$ pure DH theories predict $T_c^* = \infty$ and zero-density vapor for all $T$. Of course, these results are surely incorrect for the RPM; to correct them ion pairing must be allowed for.

As in $d = 3$ dimensions [10], inclusion of Bjerrum pairing (both with and without dipole-ionic interactions) always leads to much larger and, in fact, more reliable values for $\rho_c^*(d)$, see Fig. 2. Furthermore, $\rho_c^*(d)$ no longer vanishes when $d \rightarrow 2$. Rather, (i) for pure DHBj theory one finds

$$\rho_c^* \rightarrow \rho_{\mathrm{tri}}^* = e^{-4\gamma_E}/8\pi \simeq 0.003\,954 \quad \text{as} \quad d \rightarrow 2, \tag{14}$$

while (12) remains valid for $T_c^*(d)$; similarly (ii) DHBjDI theory yields $\rho_{\mathrm{tri}}^* \simeq 0.003\,854$ but now $T_c^*(d) \rightarrow T_{\mathrm{tri}}^* \simeq 0.246\,936$ (see Fig. 2).

More remarkably, when $d = 2$, a completely new low-density vapor phase appears for $T^* < \frac{1}{4}$ as shown in Fig. 3. In this phase $\rho_1$, the density of free ions, vanishes *identically* leaving only bound dipolar pairs: This phase must thus be an *insulator* as first identified in KT theory [12]. Furthermore, $\rho_1 \equiv 0$ implies $p/k_BT \approx \rho_2 = \frac{1}{2}\rho$ and $\xi_D = \infty$. At all low densities, up to $\rho_{\mathrm{tri}}$, the insulating, unscreened dipolar vapor is separated from the conducting, screened ionic fluid, with $\rho_1 > 0$ by a line $T_\infty^*(\rho)$ of infinite-order critical points which terminates at a *tricritical point,* see Fig. 3. Indeed, the Debye correlation length diverges as

$$\xi_D \approx \tfrac{1}{2} a e^{c/t + \gamma_E + 1/2}, \quad c = \tfrac{1}{4}\,|\ln(\rho/\rho_{\mathrm{tri}})|\,, \tag{15}$$

for $\rho < \rho_{\mathrm{tri}}$, when $t = [T - T_\infty(\rho)]/T \rightarrow 0+$, while the singular part of the free energy vanishes as $\beta f_s \approx -\ln(\xi_D/a)/2\pi\xi_D^2$, which, except for the logarithmic factor yielding a coefficient $\sim 1/t$, is in accord with hyperscaling.

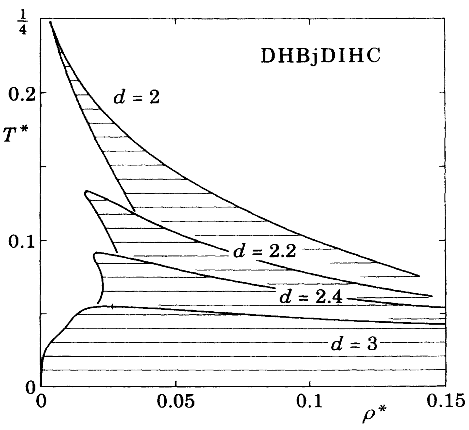

FIG. 2. Coexistence curves for DH theory with Bjerrum pairing plus dipole-ionic interactions (and hard-core terms).

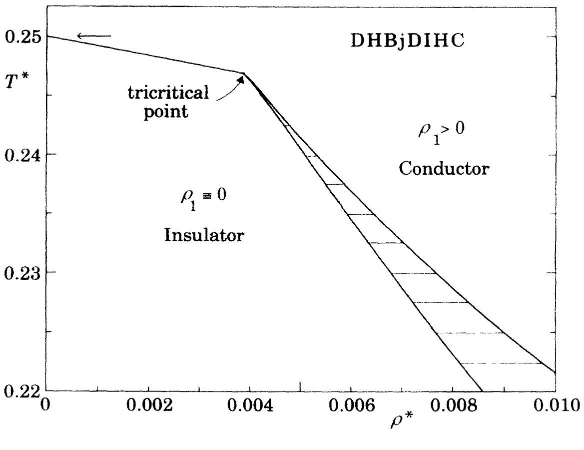

FIG. 3. Phase diagram for $d = 2$, according to DHBjDIHC theory (Fig. 2), showing an infinite-order conductor-insulator transition ending at a tricritical point.

In both DHBj and DHBjDI $d = 2$ theories, the coexistence curve between insulating vapor and conducting liquid satisfies $\Delta\rho(T) \equiv \rho_{\mathrm{liq}} - \rho_{\mathrm{vap}} \sim t/\ln|t|$ and so terminates in a cusp at $T_{\mathrm{tri}}$, see Fig. 3. Indeed, for DHBj theory $\Delta\rho = \rho_{1\mathrm{liq}}(T)$ is given by (11). Furthermore, both sides of the coexistence curve meet the critical line $T_\infty(\rho)$ *tangentially* at the tricritical point; however, since the degree of tangency is only logarithmic, this is hardly visible in Fig. 3. To be more explicit, note that in DHBj theory $T_\infty(\rho)$ is (artificially) constant: then the divergence of

$$\frac{\partial\rho_{\mathrm{vap}}^*}{\partial t} \approx \frac{\partial\rho_{\mathrm{liq}}^*}{\partial t} \approx -2\rho_{\mathrm{tri}}^* \ln\left[\frac{t_0}{t'}\ln^2\!\left(\frac{t_0}{t'}\right)\right], \tag{16}$$

when $t \rightarrow 0-$ ensures the tangency.

The mathematical origin of the surprising appearance of an insulating phase in DHBj theories lies in the law-of-mass-action expression (10), and the fact that when $d = 2$ one has $\beta\mu_1^{\mathrm{ex}} \approx -(\ln\rho_1^*)/4T^*$ and $\beta\mu_2^{\mathrm{ex}} \sim \rho_1^*(\ln\rho_1^*)/T^{*2}$ as $\rho_1^* \rightarrow 0$: see (6)–(8) when $\epsilon \rightarrow 0$. This leads to $\rho_2 \sim \rho_1^{2-1/2T^*}$. [Compare with [5(b),11(b)].] For $T^* > \frac{1}{4}$, this clearly allows both $\rho_2$ and $\rho_1 > 0$ to vanish continuously when $\rho \equiv \rho_1 + 2\rho_2 \rightarrow 0$, but no such possibility exists when $T^* < \frac{1}{4}$. Thus one is forced to a phase with $\rho_1 \equiv 0$ and $\rho = 2\rho_2$. This conclusion follows also in an analytically smooth way by examining the limit $d \rightarrow 2+$ with $T^* \lesssim \frac{1}{4}$. We remark that, unfortunately, this complex behavior when $\epsilon \rightarrow 0$ does not seem promising for a systematic renormalization group expansion in powers of $\epsilon$.

In as far as the DHBj theories yield a low-$T$ insulating phase when $d = 2$, they agree with the original Kosterlitz-Thouless theory [2], although this describes *only* the insulating phase. However, when KT theory is supplemented by RG considerations [5,18] or a full RG theory is used [19], the factor $e^{c/t}$ in (15) is replaced by $\exp(c/t^{\bar{\nu}})$ with

$\tilde{\nu} = \frac{1}{2}$ [20]. To achieve this it is clearly necessary to account for the dipole-dipole interactions which have so far been completely neglected: work to that end is under way. On the other hand, the KT and RG theories are valid only for low densities and give essentially no hint of the switch to a first-order transition which we find already at the strikingly low density $\rho_{\text{tri}}^* \equiv \rho_{\text{tri}} a^2 \simeq 4 \times 10^{-3}$. Such an abrupt change in the nature of the insulator-conductor transition has long been suspected [5(c),5(d),6(a),6(b),9] but, to our knowledge, no such explicit estimate of the threshold density has been exhibited. It must be mentioned, however, that on the basis of a somewhat *ad hoc* hybrid treatment of the KT recursion relations [5(c),5(d)] it has been suggested that the KT line terminates at a critical endpoint, associated, at a higher temperature, with a new type of $d = 2$ critical point. While this scenario might well be realized in some systems exhibiting KT-type transitions, we withhold further comment as regards Coulombic fluids until the dipole-dipole interactions have been incorporated into our DHBj-based theories.

In summary, we have presented compact closed expressions for the free energy of the basic hard-sphere ionic fluid in general dimensions within Debye-Hückel theory, extended by Bjerrum $+/-$ ion pairing and dipole-ionic interactions. The predicted low-density behavior appears correct for all $d$ ($\geq 2$). When $d$ falls $T_c^*(d)$ rises, suggesting that fluctuations may be of less importance in low $d$. Bjerrum pairing plays a crucial role when $d \to 2$ since, in its absence, condensation into an unrealistic *zero-density*, low-$T$ vapor occurs below the KT point $T_c^* = \frac{1}{4}$. However, inclusion of pairing leads instead to an insulating, unscreened dipolar phase with no free ions. The low-density transition from conducting to insulating fluid is critical but becomes first order at very low density via a tricritical point. On the critical line one has $\ln(\xi/a) \sim (T - T_c)^{-\tilde{\nu}}$ as in RG-KT analysis; however, the current DHBjDI theory yields $\tilde{\nu} = 1$ rather than $\frac{1}{2}$. This indicates a need for the inclusion of dipole-dipole interactions which, however, seems feasible within the same explicit and straightforward framework that, despite its approximate status, seems remarkably sound physically.

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[12] For $d = 3$, the term $1/a^{\epsilon}$ in (1) is normally dropped so that $\varphi_{ij} \to 0$ as $r \to \infty$. But the effect is merely to add a constant $u_0$ ($= \frac{1}{2}q^2/D\epsilon$ for the RPM) to the free energy per particle [see Eq. (6)] which has no significant consequences. Likewise, a factor $\exp(+1/T^*\epsilon)$ then appears in the result (9) for $K(T)$ but cancels in (10).

[13] Equation (2) follows from A. M. Salzberg and S. Prager [J. Chem. Phys. **38**, 2587 (1963)], but may be extended to general $d$ for $S$ logarithmically interacting species carrying charges of $G$ ($<S$) distinct types [14].

[14] X.-J. Li, Y. Levin, and M. E. Fisher (to be published). For the cylindrical Laplacian in general $d = 2 + \epsilon$ we have $r^{1-d}\partial_r(r^{d-1}\partial_r) + r^{-2}\sin^{-\epsilon}\theta\, \partial_\theta(\sin^{\epsilon}\theta\, \partial_\theta)$.

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