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## Review

# COULOMB LIQUIDS ALONG PHASE BOUNDARIES

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Three areas in which Coulomb interactions are of obvious importance are considered in turn, namely liquid alkali metals, electron–hole droplets, and lithium halides. The focus in the first two areas is dominantly on critical point properties, whereas in the third area the solid–liquid transition is the predominant interest.

Keywords: Alkali metals; Electron–hole droplets; Lithium halides

### 1. BACKGROUND AND OUTLINE

In this review we will demonstrate some correlations between various properties characterizing phase boundaries of Coulomb liquids. We start in Section 2 with phenomenology of the liquid–vapor coexistence curve (LVCC) of the liquid metals Rb and Cs in comparison with some insulating liquids. Section 3 deals with the density dependence of the critical temperature of electron–hole droplets in relation to that of the liquid alkali metals. The dependence of the melting temperature on density in the Li halides is the subject of Section 4. We end in Section 5 with a brief summary.

### 2. EMPIRICAL CORRELATIONS FOR THE LIQUID–VAPOR COEXISTENCE CURVE OF THE ALKALI METALS

Much progress has come in understanding the behavior of the low-density fluid alkalis through the experiments of Jüngst  $et$  al. [1], both using neutron scattering to study structure and also by thermodynamic measurements. In this section, it is the latter that provides the focus, and especially the mapping experimentally of the liquid– vapor coexistence curves of Rb and Cs. These elements are especially interesting to

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examine the influence of long-ranged Coulomb interactions on properties near the critical point since their metal–insulator transition coincides to within experimental error with the liquid–vapor critical point. Note that the one-component plasma (OCP), which is an important reference system for many other properties, does not have a liquid–vapor transition.

One of the striking results of the measurements of Jüngst *et al*. was the demonstration of marked departures from the law of rectilinear diameters (see e.g. [2,3]). This law, which is one of the classic laws of the physics of liquids, was stated already over a hundred years ago and essentially says that the average of the liquid and vapor density varies linearly with temperature over the whole range of temperature values, up to the critical point. Although surprisingly simple, no deviations had ever been reported until the experiments of Jüngst  $et$  al. on the heavy liquid alkalis.

As discussed in the introduction, we shall here take a strictly phenomenological approach to characterize these deviations, motivated by the early work of Guggenheim [4], and later March et al. [5], on insulating fluids such as neon and ethylene. We start by defining an order parameter  $\eta$  for the first-order liquid–vapor phase transition through

$$
\eta = \frac{\rho_l - \rho_g}{\rho_c} \tag{1}
$$

where  $\rho_l$ ,  $\rho_g$  and  $\rho_c$  respectively denote the density in the liquid phase, the gas phase and at the critical point. Then, as one expects for an order parameter,  $\eta$  is zero above the critical point and different from zero below the critical point, where the ''ordered'' liquid phase is still present. The scaled average density is given by

$$
\zeta = \frac{\rho_l + \rho_g}{2\rho_c} \tag{2}
$$

which evidently means  $\zeta = 1$  at the critical point.

We will first determine how the observed breakdown of the law of rectilinear diameters manifests itself in the dependence of the average density  $\zeta$  on the order parameter  $\eta$ . From the work of Guggenheim, who fitted rational exponents to experimental data on the LVCC of several insulating liquids, March et al. [5] obtained the relation

$$
\zeta = 1 + \frac{6}{243} \eta^3 \tag{3}
$$

In Fig. 1 we have plotted  $\zeta$  vs  $\eta^3$  for Rb. Clearly the linearity obtained by Guggenheim for insulating liquids is not reproduced for metallic Rb and a similar conclusion was obtained for Cs. This has led us to generalize Eq. (3) to the form

$$
\zeta = 1 + \aleph \eta^{\Gamma}.\tag{4}
$$

Empirically, we found that  $\Gamma = 2$  is now the correct choice for Rb and Cs as is demonstrated also in Fig. 1 for Rb. Thus our first conclusion is that  $\Gamma$  is markedly different as



FIGURE 1 Shows experimental data of Jüngst *et al.* along the liquid–vapor coexistence curve for Rb in the form of relations between difference density  $\eta$  defined in Eq. (1) and average density  $\zeta$  in Eq. (2). It can be seen that the linear plot corresponds to  $\eta^2$  vs  $\zeta$ . Guggenheim's form Eq. (3), valid for example for neon and ethylene, is plainly not appropriate for the metallic fluid Rb.

we go from the insulating fluids, where  $\Gamma = 3$  works well, to the specific metallic cases Rb and Cs, where  $\Gamma = 2$  is appropriate.

Generalizing the work of March *et al.* [5] we next point out that both the results for the heavy alkali metals and the insulating liquids can be embraced by the differential equation

$$
\eta \frac{d^2 \eta}{d \zeta^2} + (\Gamma - 1) \left(\frac{d \eta}{d \zeta}\right)^2 = 0
$$
\n(5)

with the appropriate choice for  $\Gamma$ . Similar to the insulating case discussed by March et al., we expect this form to display at least some of the structure expected in a more definitive theory of the coexistence curve.

To explicitly obtain the temperature dependence of the average density  $\zeta$ , we first examine the exponent which determines how  $\eta$  tends to zero as the critical point is approached. Empirically, we find that the data is well described by

$$
\eta = \text{constant} \times \left(1 - \frac{T}{T_c}\right)^{1/3} \tag{6}
$$



FIGURE 2 Difference density  $\eta$  plotted vs  $(1 - T/T_c)^{1/3}$ , where  $T_c$  is the critical temperature, using the data of Jüngst *et al.* for metallic Cs. The slope of the straight line has a value of  $4.12 \pm 0.03$ .

as is demonstrated for Cs in Fig. 2. From experiments on insulating liquids an exponent  $(0.35 \pm 0.02)$  was found (see e.g. [6]) and so taking into account experimental error, the behavior of the order parameter is not, or only very little, changed when going from insulating liquids to the metallic systems Rb and Cs.

With  $\Gamma = 2$  in Eq. (4), as is appropriate for metallic Rb and Cs, one is then led to the prediction for  $\zeta$ 

$$
\zeta = 1 + \text{constant} \times \left(1 - \frac{T}{T_c}\right)^{2/3} \tag{7}
$$

while of course, the law of rectilinear diameters simply means that

$$
\zeta = 1 + \text{constant} \times \left(1 - \frac{T}{T_c}\right) \tag{8}
$$

Our second conclusion is thus that departures from the law of rectilinear diameters are, for Rb and Cs, quantitatively well described by the power law in  $(1 - (T/T_c))$  used in Eq. (7).

Note that our exponents describe the behavior of the considered properties over the whole range of temperatures available from experiment. Because of this, these



FIGURE 3 Shows quality of reconstruction of liquid–vapor coexistence curve for Rb. The circles represent the experimental data of Jüngst *et al.*, whereas the continuous curves are obtained using the  $1/3$  and  $2/3$  power laws in Eqs. (6) and (7) for the difference density  $\eta$  and the average density  $\zeta$  respectively.

exponents are strictly speaking not *critical* exponents, which describe the behavior only very close to the critical point.

To illustrate the overall quality of our proposed exponents and the range of their validity we plot in Fig. 3 the liquid–vapor coexistence curve for Rb, reconstructed from Eqs. (6) and (7), together with the experimental data. Very good agreement over the full range of available data, including very close to the critical point, is shown.

It seems natural to relate the breakdown of the law of rectilinear diameter to the range of the pair-wise interaction, which is of course very different for insulating liquids and liquid metals. As discussed in [6] we expect the pair-potential in liquid metals to be proportional to  $r^{-4}$  whereas the insulating liquids interact through a van der Waals  $r^{-6}$ potential. If the polarization contribution  $r^{-4}$  is indeed present, this should lead to an ionic structure factor having the small-k form

$$
S_{ii}(k) = S_{ii}(0) + a_1k + a_2k^2 + O(k^3)
$$
\n(9)

and it would be of clear interest if a small angle scattering experiment could confirm the expansion (9).

The relation between interatomic potentials and liquid–vapor critical points was recently studied by Okumura and Yonezawa [7] using molecular dynamics simulations. They found that the law of rectilinear diameters was fulfilled for all potentials considered but these only included van der Waals type potentials and potentials with even stronger decay at large distances. It would be very interesting to extend their

calculations to include pair-potentials with a weaker decay, and in particular potentials proportional to  $r^{-4}$ , to determine if and when the law of rectilinear diameters no longer holds and whether the exponent obtained above follows.

### 3. CRITICAL POINT PROPERTIES OF LIQUID ALKALIS COMPARED TO THOSE OF ELECTRON–HOLE DROPLETS

The focus in this section will be on properties at the critical point itself. Chapman and March [8] have shown that a relationship exists between the critical temperature  $T_c$  and critical number density  $\rho_c$  for the five fluid alkalis, namely

$$
T_c \rho_c^{-1/3} = \text{constant} \tag{10}
$$

Here we examine the relevance of this relation to electron–hole liquids (EHLs) in semiconductors. An EHL (or droplet) is a condensed phase of excitons which can form in semiconductors irradiated by light at sufficiently low temperatures provided the exciton density is high enough (see e.g. [9]).

This study is prompted by the very recent experiment of Shimano *et al.* [10], who reported on the formation of an EHL with a high-critical temperature  $T_c$ 165 K in diamond, by means of time-resolved luminescence measurements under an intense femtosecond photoexcitation above the band gap. Then, by time-resolved spectral shape analysis, a very high carrier density  $\rho_0 = 1.0 \times 10^{20} \text{ cm}^{-3}$  at  $T = 0$  is revealed, together with the high value of the critical temperature  $T_c$  already recorded above.

Equation (10) above provides a natural starting point, but unfortunately while  $T_c$  is known for at least 5 EHLs, the same is not true for the critical density  $\rho_c$ . However, prompted by the form (10), we have collected  $T_c$  and the zero temperature densities  $\rho_0$  for EHLs in 5 semiconductors in Table I. Due to the form (10) we have studied first the question as to whether  $T_c$  correlates with  $\rho_0$  in the 5 EHLs referred to in Table I. Figure 4 demonstrates beyond reasonable doubt that  $T_c$  and  $\rho_0$  indeed correlate strongly. There is power law behavior between  $T_c$  and  $\rho_0$ , although the 1/3 power in Eq. (10) must be modified empirically, to 1/2.

Kalt *et al.* [11] observed picosecond electron–hole droplet formation in the indirect gap material  $Al_xGa_{1-x}As$  and, in the course of their experimental work, they refer to a ''scaling law''

$$
\frac{\rho_c}{\rho_0} \cong 0.3\tag{11}
$$

TABLE I Values for the EHL number density  $\rho_0$  at  $T=0$ , the critical temperature  $T_c$  and the dielectric constant  $\varepsilon$  in five indirect-gap semiconductors (extracted from Table 1 of Shimano et al., 2002)

	$\rho_0$ (cm <sup>-3</sup> )	$T_c$ (K)	ε
Ge	$2.5 \times 10^{17}$	6.7	16.0
Si	$3.3 \times 10^{18}$	24.5	12
GaP	$6 \times 10^{18}$	40	9.1
3C-SiC	$7.8 \times 10^{18}$	41	9.72
Diamond	$1.0 \times 10^{20}$	165	5.7



FIGURE 4 Critical temperature  $T_c$  of EHL in the 5 semiconductors referred to in Table I versus the (zero temperature) number density  $\rho_0$ . The curve drawn is given by  $T_c \rho_0^{-1/2} = 16.3 \times 10^{-9} \text{ cm}^{3/2} \text{ K}$ .

Obviously, if we adopt this so-called ''scaling law'', then Eq. (10) and the result from Fig. 4 come into intimate contact, and the conclusion is that there is a different exponent, say generally denoted by  $\aleph$  for the relationship

$$
T_c \rho_c^{-8} = \text{constant} \tag{12}
$$

with  $\aleph = 1/3$  for the 5 fluid alkalis and  $\aleph = 1/2$  for the EHLs.

Surprisingly already more than 20 years ago, Reinecke and Ying [11] had anticipated a relation  $T_c \rho_c^{-1/2}$  = constant. This is remarkable because it can be seen from Fig. 4 that the 'diamond' point of Shimano et al. [10] is crucial to fitting the parabolic form, and allowing the above constant to be made wholly quantitative. They went on however to express doubts as to whether such a relation had any fundamental basis. This leads us to the contribution of Likal'ter [13].

Likal'ter studies what he emphasizes is the limiting situation of the EHL in which hole mass  $m_h \gg m_e$ , the electron mass. He then gave individual formulae for the critical constants  $T_c$  and  $\rho_c$  discussed above, and also for the critical pressure  $p_c$ . Our interest in Likal'ter's formulae is to expose relations between these critical constants which can then be compared and contrasted with known results for the fluid alkali metals.

The first  $T_c - \rho$  result which follows from Likal'ter's model is

$$
T_c = \text{constant} \times \frac{\rho_c^{1/3}}{\varepsilon} \tag{13}
$$



FIGURE 5 Critical temperature  $T_c$  of EHL of 5 semiconductors referred to in Table I versus  $n_0^{1/3}/\varepsilon$ , where  $n_0$  is the number density of the EHL at  $T = 0$  and  $\varepsilon$  the dielectric constant.

This formula differs qualitatively from the fluid metals finding of Chapman and March [8] by the dielectric constant  $\varepsilon$  of the semiconductors appearing in the denominator. In Fig. 5 we have used the data from Table I, including the experimental dielectric constant values recorded in the final column, to plot  $T_c$  vs  $(\rho_0^{1/3}/\epsilon)$ . There is clearly a linear relation as predicted by Likal'ter's model. Accepting the scaling relation (11), the parabolic fit of the data in Fig. 4 can be reconciled with the linearity of Fig. 5 provided  $\varepsilon$  and  $\rho_0$  are related, of course approximately, by a 1/6 power law. However, at present we have no fundamental justification for a relation  $\varepsilon \rho_0^{1/6} = \text{constant}$ , because, at least in principle, such a formula could contain the binding energy of excitons, or the effective electron mass.

Returning to the similarities and differences from the fluid metals, we have also employed Likal'ter's individual formulae for  $T_c$ ,  $\rho_c$  and  $p_c$  to calculate the so-called compressibility ratio  $Z_c$  defined as

$$
Z_c = \frac{p_c}{\rho_c k_B T_c}.\tag{14}
$$

The limiting formulae (for  $m_h \gg m_e$ ) then lead to the result

$$
Z_c = \frac{7}{24}.\tag{15}
$$

This relates to the value for the heavier alkalis, 0.217 for Rb and 0.203 for Cs, but there is a wide variation through the alkali fluids (see [14]), Li having a value of 0.064. The value in Eq. (15) is not so much smaller than the prediction from the van der Waals equation of state, namely 3/8 (see e.g. [6]).

The conclusion from the present section is firstly that there are some interesting similarities but also differences between the EHLs and the fluid alkali metals with regard to critical properties. In both cases  $T_c$  is related to  $\rho_c^{1/3}$ , which reflects the fingerprints of Coulomb interactions. For the EHL, however, this quantity  $\rho_c^{1/3}$  is divided by the dielectric constant  $\varepsilon$  of the semiconductors, high for Si and considerably reduced for diamond. Secondly, the limiting formulae of Likal'ter [13] for  $T_c$ ,  $\rho_c$  and  $p_c$  are shown to yield the constant critical compressibility ratio  $Z_c$  in Eq. (15), in contrast to the wide spread of values of  $Z_c$  in the fluid alkalis.

#### 4. MELTING OF THE LITHIUM HALIDES COMPARED TO THE OCP

Here we consider the question of the behavior of the melting temperatures  $T_m$  of the lithium halides LiF, LiCl, LiBr, LiI and we interpret also the hydride LiH as a halide. To this end, we list in Table II the measured values of  $T_m$  for these five crystalline materials, together with their dielectric constant  $\varepsilon$  and the Wigner–Seitz (WS) radius  $r_s$  defined as usual in terms of the average atomic density  $\rho$  through

$$
\rho = \frac{1}{(4/3)\pi r_s^3} \tag{16}
$$

From the OCP, we expect from the melting analogue of Eq. (10) that

$$
T_m = \text{constant} \times \frac{1}{r_s} \tag{17}
$$

However, similar to the comparison between the liquid alkalis and the EHL, the dielectric constant of the crystal is now expected to modify this relation.

Figure 6 shows  $T_m$  versus  $1/\varepsilon r_s$ . Though the horizontal errors in Fig. 6 are relatively large because of the errors in the dielectric constant  $\epsilon$ , it is plain that apart from LiI, a striking linear relation between  $T_m$  and  $1/\varepsilon r_s$  exists. Clearly because of the variation in the dielectric constant no such relation holds when only the density is taken as the independent variable as is the case for the OCP. Note however that a

TABLE II Experimental values for the melting temperature, the dielectric constant and the WS-radius  $r_{s}$ . Data are taken from [15–17]

	$T_m$ (K)	ε	$r_s$ (a.u.)
LiH	963	12	1.93
LiF	1113	9.2 and 9.0	1.90
LiCl	883	11.1 and 11.9	2.42
LiBr	823	12.1 and 13.0	2.60
LiI	740	11.0	2.84



FIGURE 6 Experimental melting points of the alkali halides versus  $1/\varepsilon r_s$ .

non-negligible intersection at infinitely low density is now present, which is absent for the OCP.

Lithium bromide and iodide are almost not separated by the accuracy of  $1/\epsilon r_s$ , while their melting temperatures differ substantially. It would therefore be of considerable interest if the static (or low-frequency) dielectric constants of these two materials could be re-measured with substantially improved accuracy. Here common expectations concerning the increase in anionic size from Cl to Br to I would suggest that the polarizabilities also increase from Cl to Br to I. Then one expects electron affinities to correspondingly decrease, as is in agreement with experiment, so that also one expects  $\varepsilon$ (LiCl) < $\varepsilon$ (LiBr) < $\varepsilon$ (LiI) which however is in disagreement with the reported measurements of Lowndes and Martin [16].

Further, data available for the Na and K halides (see [16,17]) does display our expected trend. The disagreement might most likely be due to an erroneously lowmeasured value for  $\varepsilon$ (LiI), and it may be noted that a modest increase toward our expectation for  $\varepsilon$ (LiI) would bring the plot into better form. A reasonable fit of the results in Fig. 6, neglecting LiI, is given by

$$
T_m = \frac{A}{\varepsilon r_s} + B \tag{18}
$$

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#### 5. CONCLUSION

In this review we have discussed some of the empirical correlations we exposed concerning Coulomb liquids along phase boundaries. In particular we considered liquid alkali metals and electron–hole droplets in 5 semiconductors near the critical point of their liquid–vapor coexistence curve and Li halides at melting point. For these systems, the density dependence of respectively the critical and the melting temperature was shown to be fully determined by the Coulomb interaction, modified when appropriate by the dielectric constant. The exponents involved in the breakdown of the law rectilinear diameters very close to the critical point in the heavy alkalis were studied empirically and related to the behavior of the liquid–vapor coexistence curve over a wide range of temperatures.

#### Acknowledgement

It is a pleasure to thank Professor M. Ausloos for valuable correspondence concerning the relation between  $T_m$  and  $T_c$  in the alkali metals.

#### Notes added in proof

- 1. In the present context it is noteworthy for liquid metallic Cs that a transport property, electrical resistivity R, along the liquid–vapour coexistence curve (compare Fig. 3 for Rb), can be related to a thermodynamic quantity, namely magnetic susceptibility X. Forming the quantity  $X^{-1} - X_c^{-1}$ , where  $X_c$  denotes the Curie susceptibility, extracted for Cs by W.W. Warren (1984) Phys. Rev., B29, 7012 (see also R.G. Chapman and N.H. March (1988) Phys. Rev., B38, 792) one of us (NHM: in Recent Developments in Liquid State Physics (1992) Adam Hilger, Bristol, p. 188) has demonstrated a functional relation between R and  $X^{-1} - X_c^{-1}$ . Subsequently, the present writers have given theoretical arguments for the approximate linearity between  $R^{-2/3}$  and  $X^{-1} - X_c^{-1}$  (unpublished work).
- 2. Relating to Section 3, the interested reader is referred for fuller details to F.F. Leys, N.H. March, G.G.N. Angilella and M.-L. Zhang (2002) Phys. Rev., B66, 073314.
- 3. It is of interest that the melting temperature  $T_m$  and the critical temperature  $T_c$  of the alkali metals obey a remarkably linear relation  $T_m = aT_c + b$ , where  $a \sim 1/8$  which is the inverse of the local coordination number of the body-centred-cubic lattice. Furthermore, though less data is available for the lithium halides, discussed in Section 4, for critical constants, available evidence points to a similar linearity, with  $a \sim 1/12$ .

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