On melting density gap and non-congruence of phase transitions in models of dusty and colloid plasmas

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Abstract. Two simplified variants of a dusty, condensed dispersed phase and colloid plasmas models are considered as a thermodynamically equilibrium combination of classical Coulomb particles: a 2-component electroneutral system of macro- and micro-ions $(+Z, -1)$ and a 3-component electroneutral mixture of macro-ions and two kinds of micro-ions $(+Z, -1, +1)$. The base for a consideration is a phase diagram of dusty plasma by Hamaguchi *et al* (1997 *Phys. Rev.* E **56** 4671) for an equilibrium charged system with the Yukawa potential. Parameters of a splitting the one-dimensional melting boundaries of the Hamaguchi diagram (i.e. hypothetical melting density gap between separate freezing liquid line (liquidus) and melting crystal line (solidus)) are discussed. Estimation of a density gap value is made. Additional splitting of all phase boundaries in the three-component model because of so-called non-congruency of all phase transitions in this model is discussed also.

1. Introduction

Phase transitions in equilibrium highly asymmetric ensemble of macro- and micro-ions has been studied for a very long time. A number of simplified Coulomb models accompany these investigations. One example is a model of Charged Hard Spheres (CHS), which is used to describe strong asymmetric electrolytes (see e.g. [1] and references therein). One more example is so-called plasma "with condensed dispersed phase" (CDP) of 1980-s [2], and it has been the subject of study in the last 30 years. Dusty and colloid plasmas can be considered in the simplest ("primitive") approximation CDP-plasma and contemporary fully-equilibrium as an equilibrium electroneutral two-component highly asymmetric system of classical macro- and micro-ions with fixed charge numbers $+Z$ and -1 $(Z \gg 1)$. Such a two-component system (denoted below as $(+Z,-1)$) or a more complicated three-component electroneutral system with additional positive micro-ions $(+1)$ (denoted below as $(+Z,-1,+1)$) are considered as simplest dusty and colloid plasmas models [3] and so-called highly charged colloid systems [3, 4]. Both mentioned above models are well known in traditional electrolyte theory and could be successfully studied using analytical approximations such as the Debye-Hueckel app. and its improved versions (see e.g. [5]) as well as within exact computational modeling methods: Monte-Carlo, Molecular Dynamics etc. In the present paper we consider 3 systems:

A electrodischarge dusty plasma which is not fully equilibrium but all the system is trapped by a external field, the temperatures of macro- and microions are different $(Z \sim 10^2 - 10^3, T_e \neq T_i \neq T_z)$ [6];

B thermally ionized equilibrium isothermal CDP-plasma of fire gases or dense alkali metal wet steam ($Z \approx$ const, $Z \sim 10$–10^3, $T_e = T_z$) [2];

C fully equilibrium colloid plasma of highly charged macro-molecules in liquid, temperature is room ($T_e = T_z = T_i$) [7].

A base model for a comparison is

D OCP (one-component plasma) of macroions (charge numbers Z) with the Yukawa potential and a fixed screening length.

The next significant simplification is ordinary in dusty and colloid plasmas theories. Instead of considering 2 and 3-component Coulomb models (+Z,-1) and (+Z,-1,+1) with the long-range (Coulomb) interaction potential, depending on a distance only, we consider a *one-component* system of macro-ions (+Z) with the effective *screened short-range* (Debye) interaction potential of Yukawa type, with the parameter (screening length) depending on temperature and density of micro-ions.

$$V_{ij}(r) = Z_i Z_j / r, \tag{1}$$

$$V_{eff}(r) => \Phi(r \mid T_i, T_e, n_i, n_e) = \frac{(Ze)^2}{r} exp(-r/r_D), r_D = \left(\frac{4\pi e^2 n_e}{T_e} + \frac{4\pi q_i^2 n_i}{T_i} \right)^{-1/2}. \tag{2}$$

Here V_{ij} – Coulomb potential, Φ – Yukawa potential, r_D – Debye (screening) length, $T_e, T_i, T_z, n_e, n_i, n_z$ – micro- (electrons -1 and ions +1) and macroions (Z) temperatures and concentrations respectively.

Well-known phase diagram for this model in Γ – κ coordinates [8] was the starting point of the present paper.

$$\Gamma = \frac{(Ze)^2}{aT}, \kappa = \frac{a}{r_D}, a = (3/4\pi n_z)^{1/3}, \tag{3}$$

a – Wigner-Seits radius.

This diagram has been obtained by methods of exact computer simulations. There are three phase states of a Debye system with the potential (2): fluid, crystal bcc and crystal fcc. All the three boundaries are one-dimensional curves, while it is well-known that there is a first derivative discontinuity of thermodynamic potential, and thus there must be at least one gap (entropy, density) in a process of melting like in a process of a first-order transition. This density gap in a melting process is a finite for all systems except the Coulomb model of one-component plasma with the frozen neutralizing medium (OCP) and can be significant ($\sim$ 5–10%).

The purpose of this paper is, firstly, to plot an equivalent of the Hamaguchi phase diagram in "natural" coordinates temperature – density. And, secondly, it is to analyze character of splitting between different boundaries of fluid freezing and crystal melting. Also, the purpose of this paper is to analyze character of a noncongruence of phase transitions and compare this phenomenon with a congruent version of these boundaries.

2. Phase diagrams of the equilibrium Yukawa system in the natural coordinates

One can plot the Hamaguchi diagram in the "natural" coordinates in different ways. It depends on the exact definition of thermodynamic parameters of microions background. In a variety of papers ([8] and others) it is implicitly presumed that this background is passive one with constant parameters: N_m is density of microions and T_m is temperature of microions (the subscript "m" denotes "medium" as in [9]). In particular, temperature of microions T_m may be not the same as temperature of macroions. The density N_m may also be concerned with the density of macroions by a elektroneutrality condition. Finally, a thermodynamic role of this "medium" in a widely spread approach [8] is to hold the elektroneutrality condition and to guarantee Debye screening of a macroion interaction. At the same time a macroion radius of the

Debye screening $r_D(n_i, n_e, T_i, T_e)$ does not depend on a change of a phase state of a macroions subsystem. It can be consistent only in the case when the density of microions, their temperature T_e and the temperature of macroions T_i are constant during phase transitions of the system of macroions. The isochoric conditions are considered in the papers [8] and others, and density gaps of macro- and microions during phase transitions are not stipulated. Nevertheless, even taking into account this limitation, the Hamaguchi diagram [8] can be plotted in the natural coordinates (n_z, T_z). The authors consider completely equilibrium system with the same temperatures of macro- and microions $T_z = T_i = T_e = T$ (only for models B and C!) to make this supposal valid.

Let's examine the diagram in the coordinates $n_z - T_z$ ($n_z = n$, $T_z = T$) taking as an example the model $(+Z, -1)$. The key moment (the fact) is in the case (in addition to the equality of temperatures) densities n_z and n depend on each other linear because of the elektroneutrality condition ($Z n_z = n$). So, we have

$$\Gamma \propto (n/T^3)^{1/3}, \kappa \propto (n/T^3)^{1/6} => \Gamma = \frac{\kappa^2 Z}{3}. \tag{4}$$

In particular it means that if the value Z is fixed, all the variety of points on the *two-dimensional* plane n_z–T is equal to a *one-dimensional curve* $\Gamma \sim \kappa^2$ on the diagram $\Gamma - \kappa$ [8]. This curve is a line in the often using logarithmic presentation $\lg n$–$\lg T$ in a theory of Coulomb systems (figure 1).
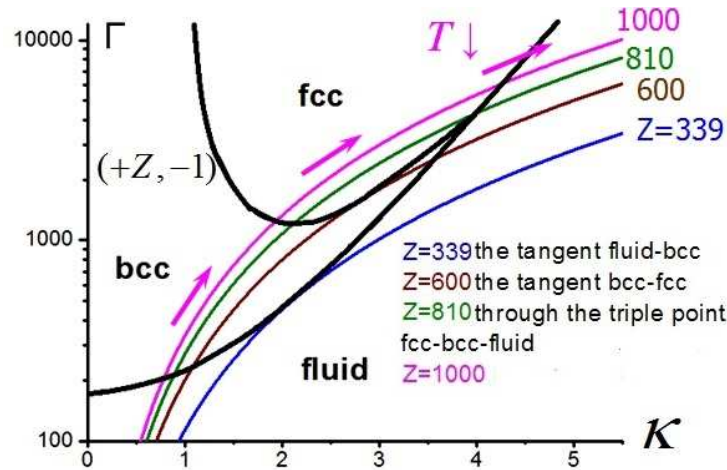

Figure 1. Phase diagram of a Debye system with the boundary lines $Z^* = const$.

Thus, there are 3 boundary values depending on a quantity of the charge of a macroion $Z = Z_1^*, Z_2^*, Z_3^*$. These values separate 4 types of phase diagram of the Yukawa model – the equivalent of the model (+Z,-1):

- $Z = Z_1^*$: – a line $\Gamma \sim \kappa^2$ is tangent to the melting boundary bcc-fluid;
- $Z = Z_2^*$: – a line $\Gamma \sim \kappa^2$ intersects the boundary bcc-fluid and is tangent to the boundary of the transition bcc-fcc;
- $Z = Z_3^*$: – a line $\Gamma \sim \kappa^2$ intersects the boundaries bcc-fluid, bcc-f and passes through the triple point.

Calculations of this paper gives the following values Z_1^*, Z_2^*, Z_3^*:
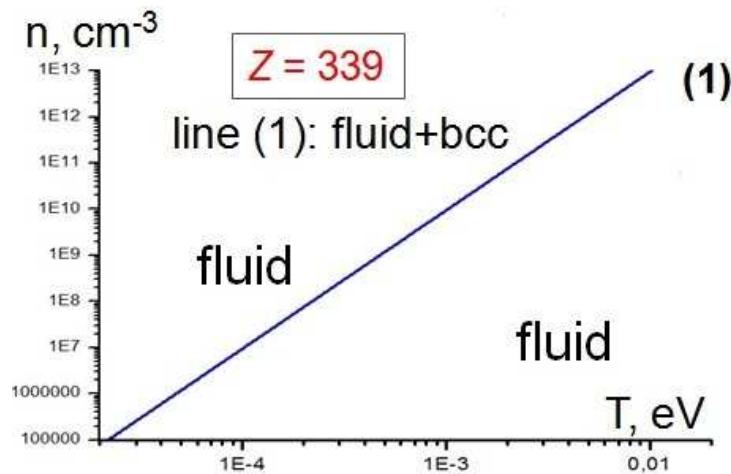
$$Z_1^* = 339, Z_2^* = 600, Z_3^* = 810.$$

Figure 2. Phase diagram of a Debye system for $Z^* = 339$. Line (1): 2 phases coexist – bcc+fluid.
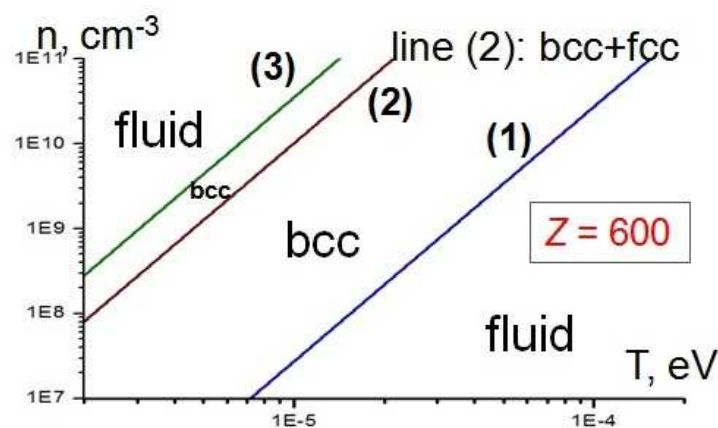

Figure 3. Phase diagram of a Debye system for $Z^* = 600$. Line (2): 2 phases coexist – bcc+fcc.

Accordingly, the considered equivalent of the diagram [8] is a set of stripes in the coordinates lg n–lg T, which is equal to three calculated in [8] phase states of the Yukawa model bcc-fcc-fluid. As it is seen from the figures 2–5, these sets are different for different Z.

To make it clear, for example, in the case of isochoric cooling it gives:

$Z < Z_1^*$ fluid;

$Z_1^* < Z < Z_2^*$ fluid $\Leftrightarrow$ bcc $\Leftrightarrow$ fluid;

$Z_2^* < Z < Z_3^*$ fluid $\Leftrightarrow$ bcc $\Leftrightarrow$ fcc $\Leftrightarrow$ bcc $\Leftrightarrow$ fluid;

$Z_3^* < Z$ fluid $\Leftrightarrow$ bcc $\Leftrightarrow$ fcc $\Leftrightarrow$ fluid.

It should be emphasized that fluid is always the stable final phase if one decreases temperature isochorically of equilibrium one-temperature model (+Z,-1) basing on the Yukawa model with the Debye screening. This is nominally correct even for $T = 0(!)$. And it appears to be a real artifact of this model that should be examined carefully. But there is nothing strange because r_D decreases if $T \to 0$ (considering the system (+Z,-1)).
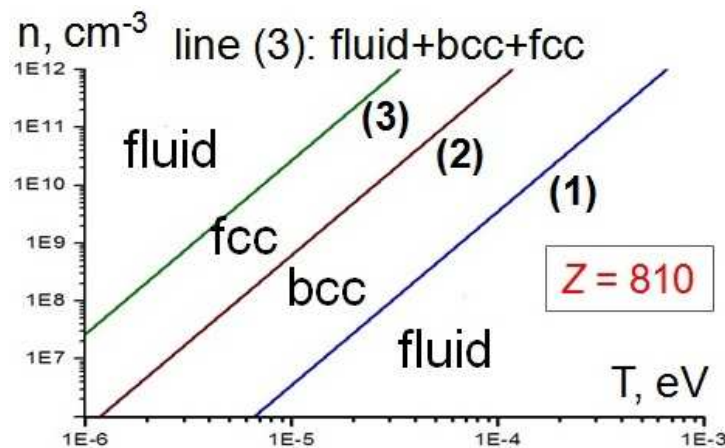
Figure 4. Phase diagram of a Debye system for $Z^* = 810$. Line (3): 3 phases coexist – fluid+bcc+fcc.
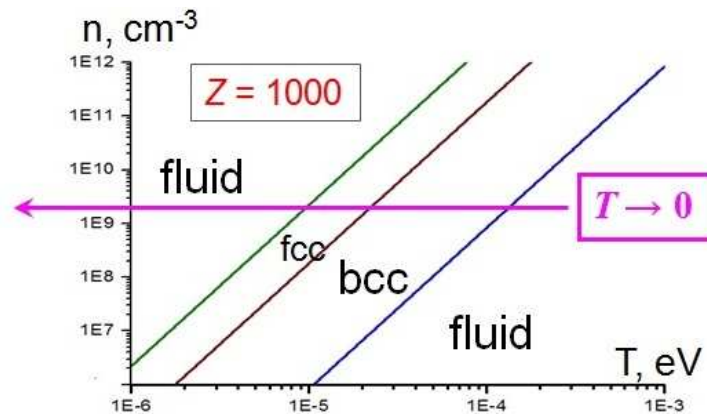

Figure 5. Phase diagram of a Debye system for $Z^* = 1000$.

3. Density gap of the model (+Z,-1)

By definition there is at least one first derivative discontinuity of thermodynamic potential speaking about of a first-order phase transition. For example, it may be the derivate of Helmholtz free energy $F(T, N, V)$ – density and entropy of coexistent phases. There is one situation when the first-order transition, melting, corresponds to the gap of only one derivate – entropy. This is the simple and well known model OCP – a one-component (classic or quantum) system of mobile charges (ions or electrons) in an *incompressible* homogeneous neutralizing medium of opposite sign charges ([10] and others). The background is rigid in the initial model (like in [5, 11] we call this model OCP(#)). Variations of volume are not determined. Well known negativity of formally appointed pressure and compressibility (when $\Gamma \gg 1$) does not mean a loss of thermodynamic stability. The only first-order phase transition in the system – crystallization – happens without a volume change and is accompanied only the entropy gap.

A more realistic version OCP($\sim$) [5, 11] takes into consideration a finite compressibility of the background, keeping the background itself *homogeneous*. This compressibility depends, for example, on a degree of degeneracy of electrons, which form the "passive" background for OCP($\sim$) of ions. There are all of three phase transitions in this system: melting, boiling and sublimation. All of them are accompanied by the finite density gap (see details in [5, 11]).

The considered in [8] and other papers model with the Yukawa potential and the fixed screening length r_D is short-range interactive. All the phase transitions in this model are accompanied by the density gap (this relates also to melting). It is known that there is a density gap in the process of melting in the model of soft spheres (SS – Soft Spheres, the other title is the model IPL, Inverse Power Low) is bigger, the harder a repulsion is. It grows from 0 for the Coulomb model (#) to $\sim 10\%$ for the model of Hard Spheres (HS). But there is no correct definition of admissible variations of volume of both the system and its subsystems in the Yukawa model from [8]. There is also no corresponding thermodynamic response to these variations (we mean pressure and compressibility). All of this leads to artificial isochoricity of all considered phase transitions fluid – bcc – fcc and to absence of the phase transition gas-fluid and gas-crystal. Meanwhile, it is well known that there are the transitions fluid-gas and crystal-gas in initial prototype of the Yukawa model [8] – asymmetric model of Charged Hard Spheres (CHS: + Z,-1). Moreover, there are finite gaps of specific volume in phase transitions in a condensed state (melting and polymorphic transitions). So, there is a question – how can one estimate values of these gaps considering the one-temperature Yukawa model with the screening length, which is defined definitely by parameters of macroions subsystem?

It is well-known, that a condition of "truncated" phase equilibrium is (besides a temperature equality) a condition of specific Helmholtz free energy of coexisting phases when one artificially imposes a condition of isochoricness, $f(T, \rho) = F(T, V, N)/N$ ($\rho = N/V$), (look, for example, [10]):

$$f'(T, \rho') = f''(T, \rho'').\tag{5}$$

It should be underlined that the equality of pressures is not obligatory in this truncated variant of phase equilibrium. Exactly this type of equilibrium has been calculated in the papers [8] and others. Thus, pressure is not considered in this approach. A general condition of phase equilibrium is (besides a temperature equality) also a condition of pressure equality and Gibbs free energy of both phases equality,

$$g(T, P) = G(T, P, N)/N = f(T, P) + p(T, \rho),$$
$$g'(T, P') = g''(T, P''), p'(T, \rho') = p''(T, \rho'').\tag{6}$$

A macroions Yukawa system response to a volume variation isn't determined in [8]. The same is valid for the thermodynamically equilibrium microion background. Because of these facts there is no definition of pressure, which has the single meaning in the Yukawa model. If the correct variant to determine pressure $p(T, \rho)$ has been chosen, a difference of specific volumes of coexisting phases can be estimated in a simplest way the changing "truncated" (isochoric) variant of phase transition [8] (5) on the complete isobaric variant of this transition (6). Supposing pressure and Gibbs specific energies disbalances small, $\Delta p = p'' - p'$ and $\Delta g = g'' - g'$, so that they agree to mentioned above isochoric variant of phase equilibrium [8], and considering for definiteness that that specific specific volumes of coexisting phases change (isochorically) so that their changes $d\nu' = -d\nu''$, we have after taking into account (5)

$$\Delta \rho \approx \Delta p / (\partial p / \partial \rho)_T.\tag{7}$$

It is expected that properties of the model with the Yukawa potential melting are similar to melting of well-known Soft Spheres model with the potential $\Phi(r) = 1/r^m$. It is also expected that if $m \to \infty$ melting parameters of the Yukawa system approach to melting parameters in Hard Sphere system. So, the density gap is anticipated to be $\frac{\Delta \rho}{\rho} \sim 10\%$! (look, for example, [12]). And if $m \to 1$ melting parameters of the Yukawa system approach to melting parameters of the model OCP(#) – $\frac{\Delta \rho}{\rho} \sim 0$ (figure 6).

The density gap is about 3% in the triple point in the Soft Sphere system [13].
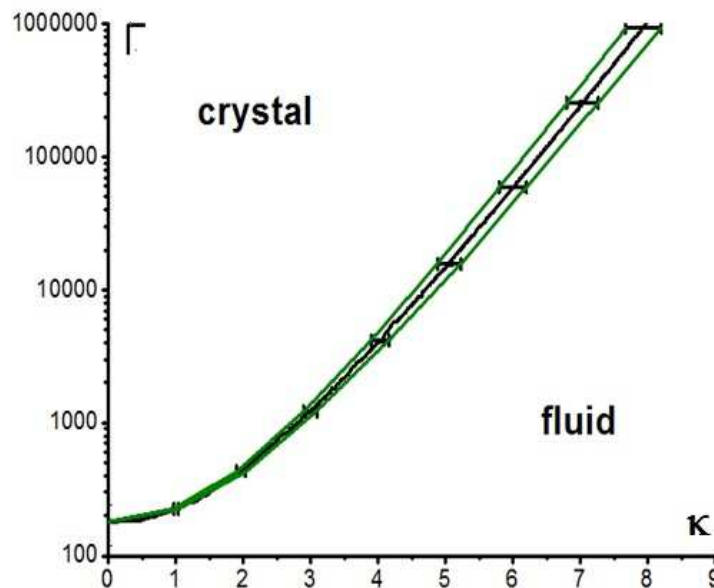
Figure 6. Estimation of the density gap in the model with the potential of the Yukawa form based on the systems' analogy with Soft Sphere.

As one needs to get total fluid and solid pressures we analyzed previous results [8,9] and found out that there is no single-valuedness of a thermodynamic background role exact definition.

$$P = -\Big(\frac{\partial F}{\partial V}\Big)_T, \tag{8}$$

$$P_{tot} = P_{id} + P_{ex} = nkT + n_e k T_e + P_{ex}. \tag{9}$$

In the figure 7, there are fluid excess pressures as functions of the parameter Γ, which were plotted in [9,14]. Notice that all 3 curves are different if $\kappa = 0$. Moreover, if κ increases slopes of the curves from [9] decreases while slopes of the curves from [14] increases.

In this case, there is nothing strange in a following situation. But there are 2 different curves if one plots fluid excess pressure on the melting curve as a function of the parameter Γ (figure 8)!

It is easy to notice that as Γ on the melting curve increases P_{tot} decreases and, finally, becomes negative! This becomes valid for compressibility $(\partial n/\partial P)_T$. According to [8] for the (electroneutral) system (+Z,-1) and $T = T_e$

$$p_{fluid} = 1 + Z + p_{ex,fluid}, \tag{10}$$

$$p_{solid} = Z + p_{ex,solid}, \tag{11}$$

we have that on the melting curve

as $Z = 1000$ $p < 0$ as $\Gamma \approx 1295$, $\kappa \approx 3,05$;

as $Z = 1000$ $(\partial P/\partial n)_T < 0$ as $\Gamma \approx 967$, $\kappa \approx 2,76$;

as $Z = 339$ $p < 0$ as $\Gamma \approx 590$, $\kappa \approx 2,28$;

as $Z = 339$ $(\partial P/\partial n)_T < 0$ as $\Gamma \approx 435$, $\kappa \approx 1,97$.

Thus, we suppose that there is no single-valuedness of a thermodynamic background role exact definition. First, background can be supposed incompressible like in [9] but, thus, there are no volume variations. In this case pressure $p = -(\partial f/\partial n)_T$ is formal quantity and has no
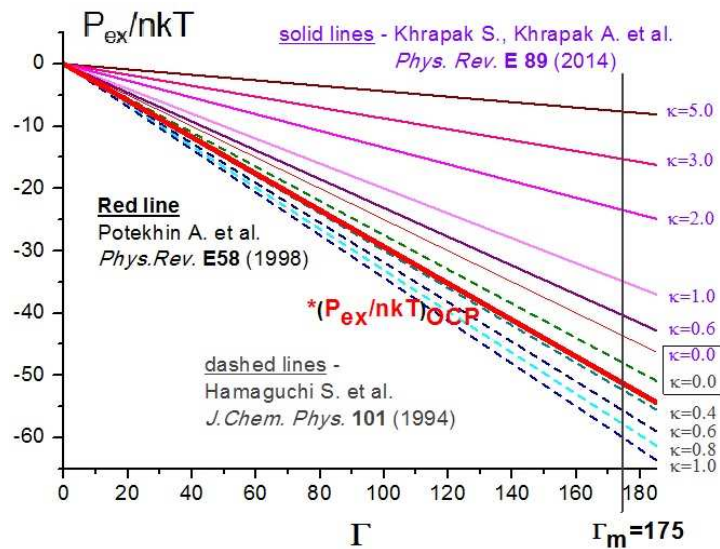
Figure 7. Fluid excess pressure as a function of the parameter Γ plotted by Hamaguchi et al. [14], Khrapak S.A. et al. [9] and this dependence for the model OCP [15].
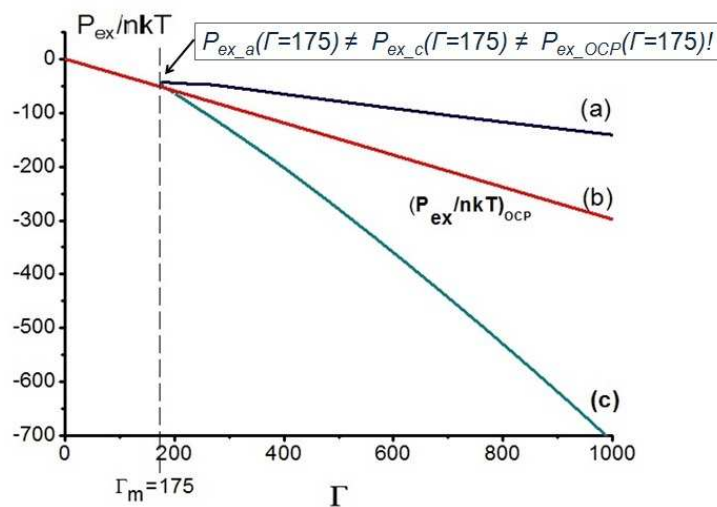

Figure 8. Comparison fluid excess pressures on the melting curve as functions of the parameter Γ based on [8, 9, 14] and (as $\kappa = 0$) excess pressure as a function of Γ for OCP [15]. (a) is based on [14] (Khrapak S.A. et al.), (b) is based on [15] (Potekhin et al.), (c) is based on [8, 14] (Hamaguchi et al.).

physical meaning. Second, medium can be supposed to be compressible like in [8]. But in such a way a system should collapse if pressure becomes negative and there is no phase immiscibility if compressibility becomes negative.

The density gap is less than 1% as $\kappa \leq 2$.

4. On non-congruence of phase transitions of the model CHS (+Z,-1,+1). Galvani potential

Non-congruent (or incongruent) phase transition (NCPT) is the most common form of first-order phase transitions in the equilibrium systems that consist from two or more chemical elements, e.g. in mixtures or compounds (for example, look [16, 17]). This type of phase transitions is general and is widely realized in various situations, including exotic forms of nuclear and quark-hadron transitions fluid-fluid in superdense matter of extreme parameters (for example, look [18, 19]). One of distinctive properties of non-congruent phase transitions is the fact that all the interphase boundaries in *intensive thermodynamic* variables, for example, $P(T)$ must be not one-dimentional curves, like in the Van-der-Waals transition gas-fluid or in melting, but two-dimensional zones for a non-congruent transition.

All the above mentioned also fairs to plasma phase transitions with macroions (dusty, colloidal, electrolytic and others right up to so-called "pasta plasma" (look, for example, [20]). The key condition to realize a non-congruent situation of one of phase transitions in a system (besides "usual" forced-congruent situation, e.g. equilibrium (according to Maxwell's thermodynamic relations)) is its two or higher thermodynamic dimension. It means that there should be two or more "charges". In the case of chemically reacting plasma it is equal to presence two or more chemical elements in co-existence phases. A long-range action of Coulomb forces in Coulomb systems and, as a result of it, the electroneutrality condition in each of co-existing macroscopic phases lowers per unit the mentioned above thermodynamic dimension of the phase transition. It means that considering in this paper phase transitions are always congruent in the more simple (not Coulomb) Yukawa model because a system is one-component.

Transitions in two-component (Coulomb) model CHS $(+Z, -1)$ are also congruent because a system is one-component in view of the electroneutrality condition. The system $(+Z, -1, +1)$ is thermodynamically two-dimensional because of extra degree of freedom (free parameter) ratio of concentrations of microions $(+1, -1)$ (or ions and electrons in complex plasmas).

A specify of this system is in the fact that its non-congruence is explicitly linked with another important property of interphase surfaces in the Coulomb systems — a presence of a stationary gap of a middle electrostatic potential in co-existing phases on these interphase boundaries. This potential is called the Galvani potential $\Delta\varphi$ (look [5, 17, 21] and references therein). As for the model CHS $(+Z, -1, +1)$, the phase equilibrium condition in many-component plasmas (the Gibbs-Guggenheim condition, look, for example, [18]), is

$$T' = T'', P' = P'', \tag{12}$$

$$\mu'_z = \mu''_z + Ze\Delta\varphi, \mu'_i = \mu''_i + e\Delta\varphi, \mu'_e = \mu''_e - e\Delta\varphi. \tag{13}$$

We underline that pressure and all "local" (usual) chemical potentials of macro- and microions (μ_z, μ_e, μ_i) are functions from temperature and all concentrations $\mu = \mu(T, n_z, n_i, n_e)$.

Non-congruence of phase equilibriums in the model $(+Z, -1, +1)$ manifests itself in an inequality of a connection $x \equiv n_i/n_e$ in co-existing phases. Here it is an analogue of chemical "composition" in mixtures. Because of the electroneutrality condition $(Zn_z + n_i = n_e)$:

$$x' \neq x'' \Rightarrow (n_i/n_e)' \neq (n_i/n_e)''. \tag{14}$$

Correct calculation of parameters of a non-congruent (full) phase transition in a many-component system is a more complicated issue than calculations of a congruent (partial) equilibrium in this system (look, for example, [17, 22]). To make a simple estimation of a sign and a value of hypothetical non-congruence of melting in the model $(+Z, -1, +1)$ it is used maximum of possible simplifications.

It is assumed that a shift to a non-congruent melting is a small deviation from the isochoric melting calculated in [8]. It means it can be also assumed that macroions densities in crystal

and fluid are the same and corresponding to calculated in [8]. We suppose that a little shift to a non-congruent equilibrium happens only because of an exchange of microions among phases (relatively equal concentrations n_i and n_e, which are implied because of the isochoric conditions [8]). Moreover, it means that results of calculations of free energy of both phases (look [8]) can be used to estimate a difference of the chemical potentials of macroions n_z in crystal and fluid. And while one calculates these chemical potentials they can differentiate with respect only to an explicit dependence $F(T, n_z, r_D(T, n_i, n_e))$ from n_z and neglect an implicit dependence in the Debye radius. Thus, because of the first of equations (13) it gives an estimation of the Galvani potential value:

$$\Delta\varphi(T, n, r_D) = Z^{-1}\{(dF'(T, n, r_D)/dn)_{T, r_D} - (dF''(T, n, r_D)/dn)_{T, r_D}\}. \tag{15}$$

The equations (13) and (14) give the value of difference (local) chemical potentials of microions:

$$\mu_i' - \mu_i'' = \mu_e'' - \mu_e' = Z^{-1}(\mu_z' - \mu_z'') = Z^{-1}(\mu_i' - \mu_i'')_{[8]} = e\Delta\varphi. \tag{16}$$

Isochoric equilibrium is used as a zero approximation. The concentrations of microions n_{io} and n_{eo} of co-existing phases (also both in crystal and fluid) are implied the same ($n_{i0}' = n_{i0}''$; $n_{e0}' = n_{e0}''$) in calculations of the isochoric equilibrium. The condition (13) can be realized only if there is a shift of n_i and n_e. It is important to make the following conditions (15) valid. It is taken into account a dominating quasi-ideal dependence chemical potential $_{i,e}$ from $n_{i,e}$ only in order to make a simple estimation of the stated shift. Small (Coulomb) corrections are also neglected because of non-ideality:

$$\mu_{i,e}(T, n_i, n_e, n) \approx kT \ln(n_{i,e} r_{i,e}^3). \tag{17}$$

The equations (16) and (17) give the sought shifts:

$$n_i'/n_i'' \approx exp(e\Delta\varphi/kT), n_e'/n_e'' \approx exp(-e\Delta\varphi/kT). \tag{18}$$

Finally, we have for the shift of the parameter $x \equiv n_i/n_e$ during the non-congruent melting (NCPT) compared to congruent [8], where:

$$(x'/x'')_{NCPT} \approx exp(2e\Delta\varphi/kT) = exp(2\Delta\mu_z/kT). \tag{19}$$

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