Ensemble inequivalence in the structure of a Coulomb liquid

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The ensemble inequivalence in a (unscreened) Coulomb liquid has been known since the years 1960s. The static structure described by the canonical ensemble and the grand canonical ensemble are not equivalent in these cases. This occurrence is reviewed in the larger setting of the ensemble inequivalence in long range fluids. We suggest that this inequivalence should continue to hold even in correspondence of a phase transition where one may have a "sum rules melting".

Keywords: Long range fluids; Coulomb liquids; static structure factor; canonical ensemble; grand canonical ensemble; ensemble inequivalence; sum rules; phase transitions

I. INTRODUCTION

Systems in d dimensions with a pairwise interaction potential which decays at large distances as $v(r) \simeq b/r^{d+\sigma}$ as $r \to \infty$ with $-d < \sigma \leq 0$, are referred to as nonintegrable, or systems with long range interactions. Such systems have an ill defined thermodynamic limit [1]. This may be correctly restored by applying the Kac prescription [2], within which the potential is rescaled by an appropriate factor which vanishes as the range of the interaction diverges. Or, for Coulomb systems [3–7], this problem is solved by making the liquid globally neutral. In a one component plasma one uses a uniform neutralizing background of opposite charge than the total charge of the particles and in a two component plasma the two components will have opposite charges so as to result in a net zero total charge of the whole system. Non quantum Coulomb systems offer some exact analytic solutions in one dimension [3, 4], in two dimensions [8], and at a special value of the coupling constant in two dimensions [5] on various surfaces: The plane [5], the cylinder [9], the sphere [10], the pseudosphere [11], the Flamm's paraboloid [12, 13]. However the energy remains non additive, i.e. the system cannot be divided into independent macroscopic parts, as is usually the case for short range interactions. Ensemble inequivalence, i.e. the possibility of observing different thermodynamic or structural properties depending on the statistical ensemble which describes the system, is one of the hallmarks of long range physics, which has been demonstrated in numerous classical systems [14–18].

In introductory books of statistical mechanics [19] we find that a statistical system can be described equally well by the four statistical ensembles: The microcanonical ensemble in which nothing fluctuates and it is completely constrained, the canonical ensemble where only the entropy S fluctuates, the grand canonical ensemble where S, N fluctuate, N being the number of particles, and the isothermal isobaric ensemble where S, V fluctuate, V being the volume of the system. In a strategy of reduction of the extensive variables (S, V, N) in favor of the intensive ones (temperature T, pressure P, chemical potential μ) which gives rise to the thermodynamic potentials in the various ensembles, starting from the internal energy E of the microcanonical, it is also natural to consider the completely unconstrained ensemble [15] as the one where all three of the extensive variables S, V, N fluctuate. Clearly, for additive systems the thermodynamic potentials are linear homogeneous functions of the extensive variables so that the thermodynamic potential of the unconstrained ensemble only makes sense for non additive systems such as the long range ones [15]. Non additivity is also responsible for the loss of convexity/concavity of thermodynamic potentials which in turn may result, from a Legendre transfrom, to non differentiable points and phase transitions [20].

The thermodynamic properties of a fluid can be extracted by its (static) structure, i.e. the knowledge of its many body correlation functions [21] and not vice versa. In this brief work we will show how the structure of long range many body systems displays an inequivalence between its canonical and grand canonical descriptions. This fact, which affects computer experiments on these kind of models as well, should be stressed clearly because it is often overlooked. Therefore, when dealing with long range systems, one should always specify which kind of ensemble he is using to extract the properties of the fluid he is studying. This becomes very important in numerical experiments as outlined in Section III.

The oldest, more important, and best studied of all long range systems is the many body Coulomb system. When talking about Coulomb systems we should have in mind its prototypical and simplest model, the *Jellium*. This is a system of pointwise electrons of charge e and number density n in the three dimensional Euclidean space filled with an uniform neutralizing background of charge density $-en$. The zero temperature, ground-sate, properties of

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the statistical mechanical system thus depends just on the electronic density n, or the Wigner-Seitz radius $r_s = (3/4\pi n)^{1/3}/a_0$ where a_0 is Bohr radius, or the Coulomb coupling parameter $\Gamma = e^2/r_s a_0$.

The model can be used for example as a first approximation to describe free electrons in metallic elements [22] ($2 \lesssim r_s \lesssim 4$) or the interior of a white dwarf [23] ($r_s \simeq 0.01$). More generally it is an essential ingredient for the study of ionic liquids (see Ref. [21] Chapter 10 and 11): *molten-salts*, *liquid-metals*, and *ionic-solutions*. In molten alkali halides the masses of the cation and the anion are comparable whereas in liquid metals the anions are replaced by electrons from the valence or conduction bands. The very small mass of the electron leads to a pronounced disymmetry between the two species present in the metal. In particular, whereas the behavior of the cations can be discussed in the framework of classical statistical mechanics, the electron form a degenerate Fermi gas for which a quantum-mechanical treatment is required. Restricting to the class of simple metals in which the electronic valence states are well separated in energy from the tightly-bound core states; their properties are reasonably well described by the nearly-free-electron model. Metals that are classified in this sense include the alkali metals, magnesium, zinc, mercury, gallium, and alluminium. Other liquid metals (noble and transition metals, alkaline earths, lanthanides, and actinides) have more complicated electronic structures, and the theory of such systems is correspondingly less well advanced. Molten-salt solutions are mixtures of liquid metals and molten salts. Ionic-solutions are liquids consisting of a solvent formed by neutral, polar molecules, and a solute that dissociates into positive and negative ions. They vary widely in complexity: in the classic electrolyte solutions, the cations and anions are comparable in size and absolute charge, whereas macromolecular ionic solutions contain both macroions (charged polymers chains or coils, micelles, charged colloidal particles, etc.) and microscopic counterions.

Whereas the finite, non zero, temperature T properties of Jellium depend additionally on the electron degeneracy parameter $\Theta = T/T_F$, where T_F is the Fermi temperature. Apart from its purely speculative interest, the temperature dependence of the Jellium properties are certainly of great astrophysical relevance. Examples are dense plasmas in the interior of giant planets [24] and brown dwarfs atmospheres. Other uses could be in highly compressed laboratory plasmas, such as laser plasmas [25], inertial confinement fusion plasmas [26], and pressure-induced modifications of solids, such as insulator-metal transitions [27]. These examples justify the growing interest recently emerged in matter under extreme conditions in the warm-dense regime [28].

II. ENSEMBLE INEQUIVALENCE

Given a fluid of N particles of mass m in a volume V with a density $\rho = N/V$ at an absolute temperature T in the canonical ensemble, the n-particle density of the many body system is defined as [21]

$$\rho_N^{(n)}(\mathbf{r}^n) = \frac{N!}{(N-n)!} \frac{\int\int \exp[-\beta \mathcal{H}_N(\mathbf{r}^N, \mathbf{p}^N)]\, d\mathbf{r}^{N-n} d\mathbf{p}^N}{Q_N(V,T)} \tag{2.1}$$

$$= \frac{N!}{(N-n)!} \frac{\int \exp[-\beta \mathcal{V}_N(\mathbf{r}^N)]\, d\mathbf{r}^{N-n}}{Z_N(V,T)}, \tag{2.2}$$

where $\mathbf{r}^n = (\mathbf{r}_1, \ldots, \mathbf{r}_n)$ and $\mathbf{p}^n = (\mathbf{p}_1, \ldots, \mathbf{p}_n)$ are respectively the coordinates and momenta of the n particles, we denote with $d\mathbf{r}^n = d\mathbf{r}_1 d\mathbf{r}_2 \cdots d\mathbf{r}_n$, $\beta = 1/k_B T$, $\mathcal{H}_N(\mathbf{r}^N, \mathbf{p}^N) = \mathcal{K}_N(\mathbf{p}^N) + \mathcal{V}_N(\mathbf{r}^N)$ is the Hamiltonian of the many body N particle fluid, $Q_N = \int\int \exp[-\beta \mathcal{H}_N(\mathbf{r}^N, \mathbf{p}^N)]\, d\mathbf{r}^N d\mathbf{p}^N$ is the canonical partition function, and $Z_N = \int \exp[-\beta \mathcal{V}_N(\mathbf{r}^N)]\, d\mathbf{r}^N$. So that the $\rho_N^{(n)}$ are normalized as $\int \rho_N^{(n)}(\mathbf{r})\, d\mathbf{r}^n = N!/(N-n)!$.

The n particles distribution functions are

$$g_N^{(n)}(\mathbf{r}^n) = \rho_N^{(n)}(\mathbf{r}^n) / \prod_{i=1}^{n} \rho_N^{(1)}(\mathbf{r}_i). \tag{2.3}$$

From the normalization condition for $\rho_N^{(n)}$, it follows immediately that if the system is homogeneous, i.e. the $\rho_N^{(1)}$ are constant and equal to $\rho = N/V$,

$$\begin{aligned} S_N^c(\mathbf{0}) &= 1 + \rho \int [g_N^{(2)}(\mathbf{r}) - 1]\, d\mathbf{r} \\ &= 1 + \rho \int \left[\frac{\rho_N^{(2)}(\mathbf{r}, \mathbf{0})}{\rho^2} - 1 \right] d\mathbf{r} \\ &= 1 + \int \frac{\rho_N^{(2)}(\mathbf{r}, \mathbf{0})}{\rho}\, d\mathbf{r} - N \\ &= 1 + \frac{N(N-1)}{V\rho} - N = 0. \end{aligned} \tag{2.4}$$

where $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$ and $S_N(\mathbf{k}) = 1 + \rho \int [g_N^{(2)}(\mathbf{r}) - 1] \exp(-i\mathbf{k}\cdot\mathbf{r})\, d\mathbf{r}$ is the static structure factor of the fluid, with $\mathbf{k}$ the wave vector of the beam scattering with the fluid.

Eq. (2.4) is correct for Coulomb systems even in the thermodynamic limit [29] where it is also known as the *charge sum rule*, the first of a hierarchy of sum rules called multipolar [6] imposed by electrostatics and the hypothesis of exponential clustering, i.e. the asymptotic exponential decay of correlation functions as two groups of particles are largely separated. Systems for which Eq. (2.4) holds in the thermodynamic limit are called *hyperunifrom* [30]. For long range non Coulombic potentials, $\sigma \neq -2$, (even if, instead of considering the Coulomb potential as the solution of the Poisson's equation, one could alternatively define it as $v(r) \propto 1/r$ in any dimension) the Fourier transform of the pair potential $\tilde{v}(k) \simeq a_d b k^\sigma$ as $k \to 0$, and an analysis of the equilibrium equations of the Born-Green-Yvon hierarchy shows (see section II.B.3 of Ref. [6]) that the Fourier transform of the static structure factor, in the thermodynamic limit, $S^c(k) = (\beta a_d b)k^{-\sigma} + g(k)$ where $g(k)$ depends on the three points correlation functions and is such that $g(k) \to 0$ as $k \to 0$ due to clustering. When $\sigma \neq -2$ the first term is not analytic in $\mathbf{k}$ at $\mathbf{k} = 0$ and this singularity introduces an algebraic term of order $r^{-(d-\sigma)}$ in the asymptotic, large r, development of $S^c(r) = \int S^c(k) \exp(-i\mathbf{k}\cdot\mathbf{r})\, d\mathbf{k}/(2\pi)^d$ [31]. Therefore among all possible long range potentials, it is only in the Coulomb case that a decay law of correlations faster than any inverse power is compatible with the structure of equilibrium equations.

For short range fluids, on the other hand, we know [32] that, in the thermodynamic limit,

$$\lim_{r\to\infty} g^{(2)}(\mathbf{r}) = g_\infty^{(2)} = 1 - \frac{\chi_T}{\chi_T^0}\frac{1}{N} + o(1/N), \tag{2.5}$$

where $r = |\mathbf{r}|$ is the modulus of the relative vector $\mathbf{r}$, $\chi_T = [\rho(\partial P/\partial\rho)_{N,T}]^{-1}$ is the isothermal compressibility of the fluid, and $\chi_T^0 = \beta/\rho$ is the isothermal compressibility of the ideal gas. Eq. (2.5) assumes the existence of a finite correlation length in a liquid [32]. It will break down in a neighborhood of a phase transition when correlation lengths diverge at criticality.

Then from Eq. (2.5) follows that, in the thermodynamic limit,

$$S(\mathbf{0}) = 1 + \rho \int [g^{(2)}(\mathbf{r}) - 1]\, d\mathbf{r} \to \frac{\chi_T}{\chi_T^0}, \tag{2.6}$$

in fact the integral can be split in two pieces: one over a volume V in a neighborhood of the origin and one over the remaining volume V' outside of V where to a good approximation $g^{(2)} \approx g_\infty^{(2)}$. But we know that the first piece must tend to -1 due to the property (2.4) whereas in the second piece we can replace $g^{(2)}$ with the $g_\infty^{(2)}$ of Eq. (2.5) and notice that in the thermodynamic limit $V'/V \to 1$. Eq. (2.6) shows how for short range systems in the thermodynamic limit both the canonical and grand canonical ensembles give the same result (2.6) and (2.14). This is not generally true anymore for long range systems as is proven by the counterexample of a Coulomb system where Eq. (2.6) breaks down since due to electrostatics the *charge sum rule* must hold also in the thermodynamic limit.

On the other hand, in the grand canonical ensemble, where $P(N)$ is the probability that the fluid contains N particles irrespective of their coordinates and momenta, we define instead

$$\rho^{(n)}(\mathbf{r}^n) = \sum_{N\geq n} P(N)\rho_N^{(n)}(\mathbf{r}^n) \tag{2.7}$$

$$= \frac{1}{\Theta}\sum_{N\geq n} \frac{z^N}{(N-n)!}\int \exp[-\beta\mathcal{V}_N(\mathbf{r}^N)]\, d\mathbf{r}^{N-n}, \tag{2.8}$$

where $z = \Lambda^{-3}\exp(\beta\mu)$ is the activity, with μ the chemical potential and $\Lambda = \sqrt{2\pi\beta\hbar^2/m}$ the de Broglie thermal wavelength, and $\Theta = \exp[-\beta\Omega(\mu, V, T)] = \exp(\beta PV) = \sum_{N=0}^\infty \exp(N\beta\mu)Q_N$, with Ω the grand potential and P the pressure. Then the $\rho^{(n)}$ are normalized as $\int \rho^{(n)}(\mathbf{r}^n)\, d\mathbf{r}^n = \langle N!/(N-n)!\rangle$, where $\langle\ldots\rangle$ denotes an average with respect to $P(N) = \exp(N\beta\mu)Q_N/\Theta$.

The average number of particles in the system is

$$\langle N\rangle = \sum_{N=0}^\infty NP(N) = \frac{\partial \ln\Theta}{\partial \ln z}, \tag{2.9}$$

so that

$$\frac{\partial\langle N\rangle}{\partial\beta\mu} = \langle N^2\rangle - \langle N\rangle^2. \tag{2.10}$$

It follows then

$$0 \leq \frac{\langle N^2\rangle - \langle N\rangle^2}{\langle N\rangle} = \frac{1}{\langle N\rangle}\frac{\partial\langle N\rangle}{\partial\beta\mu}, \tag{2.11}$$

where this intensive ratio is related to the isothermal compressibility. In fact, for an infinitesimal isothermal change it follows that $VdP = Nd\mu$, where P is the pressure. If the change also takes place at constant volume, both dP and $d\mu$ are proportional to dN: $dP = (\partial P/\partial N)_{V,T}dN$ and $d\mu = (\partial\mu/\partial N)_{V,T}dN$. So that $N(\partial\mu/\partial N)_{V,T} = V(\partial P/\partial N)_{V,T} = (\partial P/\partial\rho)_{N,T} = 1/\rho\chi_T$, with χ_T the isothermal compressibility. At equilibrium N may be replaced by $\langle N\rangle$ so that [21]

$$\frac{\langle N^2\rangle - \langle N\rangle^2}{\langle N\rangle} = \frac{\rho\chi_T}{\beta} = \frac{\chi_T}{\chi_T^0}, \tag{2.12}$$

with χ_T^0 the isothermal compressibility of the ideal gas.

The n particles distribution functions are

$$g^{(n)}(\mathbf{r}^n) = \rho^{(n)}(\mathbf{r}^n)/\prod_{i=1}^{n}\rho^{(1)}(\mathbf{r}_i). \tag{2.13}$$

From the normalization condition for $\rho^{(n)}$ and the thermodynamic condition of Eq. (2.11), it follows immediately that, if the system is homogeneous, i.e. $\rho^{(1)}$ is constant and equal to $\rho = \langle N\rangle/V$,

$$\begin{aligned} S^{gc}(\mathbf{0}) &= 1 + \rho\int[g^{(2)}(\mathbf{r}) - 1]\,d\mathbf{r} \\ &= 1 + \rho\int\left[\frac{\rho^{(2)}(\mathbf{r},\mathbf{0})}{\rho^2} - 1\right]d\mathbf{r} \\ &= 1 + \frac{\langle N(N-1)\rangle}{V\rho} - \langle N\rangle \\ &= \frac{\langle N^2\rangle}{\langle N\rangle} - \langle N\rangle = \frac{\chi_T}{\chi_T^0}, \end{aligned} \tag{2.14}$$

which agrees with the calculation in the canonical ensemble in the thermodynamic limit (2.6), but only for short range systems, since generally $\chi_T \neq 0$.

Comparing Eq. (2.14) with Eq. (2.4) we then see that the structure predicted by the canonical ensemble does not agree with the one predicted by the grand canonical ensemble for long range (unscreened) Coulomb fluids but they agree only for short range fluids for which Eq. (2.6) holds. They do give rise to ensemble inequivalence. Here we are thinking of unscreened charged liquids, i.e. liquids whose constituents particles are (not completely screened) charges. Not liquids like water or even like ionic screened two component liquids where one should rather look at the Bhatia-Thornton structure factors as done in Refs. [33–35]. In particular the work of Aqua and Fisher [35] suggests to look for ensemble inequivalence in ionic liquids near the critical point when there is a *ionic asymmetry* which couples charge and density fluctuations in a direct manner: The charge correlation length then diverges precisely as the density correlation length. For short range or long range non Coulomb systems one should study ensemble inequivalence case by case.

III. PRACTICAL IMPLICATIONS

The ensemble inequivalence in Coulomb fluids has practical implications in many numerical experiment that can be found in the literature. For example in the canonical ensemble the quantum one component electron plasma, the *Jellium*, has been studied by various authors using path integral Monte Carlo methods (see for example the works [36–38] and references therein). We clearly see from these studies that $S(0) = 0$. While those studies are on a plasma of pointwise charged fermions in the canonicalensemble, the authors of Ref. [39] study a plasma of pointwise charged bosons in the grand canonical ensemble (with the worm algorithm). The difference between these studies illustrates the role played by the different statistics used to describe the same quantum Coulomb system of identical charged particles. But it should not be overlooked the role played by the here discussed ensemble inequivalence when analyzing their results. In the two dimensional two component plasma of Ref. [40] the ensemble inequivalence is found for what they call the "underscreened" Coulomb fluid. At low temperature below the Kosterlitz-Thouless transition the ensemble inequivalence emerges in the insulator phase.

In classical Coulomb unscreened charged liquids one can consider for example the case of the two component plasma. Here one should rather look at the Bhatia-Thornton structure factors as done in Refs. [33–35]. The work of Aqua and Fisher [35] suggests to look for ensemble inequivalence in ionic liquids near the critical point when there is a *ionic asymmetry* which couples charge and density fluctuations: The charge correlation length then diverges precisely as the density correlation length giving rise to the breaking of the exponential clustering. Nonetheless the arguments outlined in the previous section will continue to hold since they are just normalization conditions for the correlation functions. So we still expect inequivalence between the canonical and the grand canonical ensembles.

The structure factor of a fluid can be obtained experimentally from measurements of cross-section for scattering of neutrons or X-rays as a function of the scattering angle. Since the structure factor $[S(\mathbf{k})-1]/\rho$ is the Fourier transform of the total correlation function $h(\mathbf{r}) = g(\mathbf{r}) - 1$, and viceversa, the large r asymptotic behavior of h determines the small $\mathbf{k}$ asymptotic behavior of S [31]. On the other hand the small r behavior of h determines the large k behavior of S. So that, for example, an integrable h that is analytic, i.e. infinitely derivable, produces a decay to zero of $S-1$ faster than any inverse power of k. Non analyticities will give rise to oscillations. In a computer experiment where one only has finite simulation cells, and mimics the thermodynamic limit by choosing periodic boundary conditions on it, Eqs. (2.4) and (2.14) will be subject to finite size errors.

IV. CONCLUSIONS

We showed that it has long been known about the ensemble inequivalence in Coulomb liquids which manifests itself for example comparing the structure of the liquid in the canonical and grand canonical ensembles. For example we can compare the canonical path integral Monte Carlo simulation of the Fermi one component plasma (the Jellium) of Ref. [36] where we clearly see from their Fig. 2 that $S(0) = 0$ with the grand canonical (worm algorithm) path integral Monte Carlo simulation of the Bose one component plasma of Ref. [39] where $S(0) \neq 0$. Recently there has been a revival of interest in this fact also for what concerns the inequivalence between the micro canonical and canonical ensembles of some long range fluids [14] or the inequivalence between the unconstrained and isothermal isobaric ensembles of the modified Thirring model [15–18]. In this short work we showed that long range fluids may or may not be inequivalent in the static structure described by the canonical and the grand canonical ensembles.

On approaching a phase transition [35, 41–43] some sum rules will merge and some will cease to hold. In particular, once a description in terms of an order parameter is found, we may end up with a unified treatment of sum rules and upon the divergence of certain correlation lengths some other may fail.

We will call this process the "melting or evaporation of the sum rules". An example of failure is given by the second and fourth moment sum rules for ionic liquids at their vapor-liquid phase transition [43]. In this case the divergence of the correlation length produces the exponential clustering breaking and the consequent failure or violation of multipolar sum rules *except* for the very first one, the charge sum rule, the zeroth moment sum rule, which, being just a consequence of the electrostatic charge neutrality, should always hold

The universality property of the renormalization group [44] theory of critical phenomena is a consequence of the existence of a fixed point for the scaling symmetry on which expand the smooth Hamiltonian space [45–51] on its tangent space. A different universality holds for the *mathematical* validity of a sum rule in the theory of many body systems which holds in different *physical* liquids like for example in the *Jellium problem* [3, 4, 6, 7, 52–54]. From this perspective it is rather amusing to notice how one often experiences a breakdown of sum rules at criticality.

The ensemble inequivalence should always be kept in mind when studying or simulating these kinds of long range many body systems.

AUTHOR DECLARATIONS

Conflict of interest

The author has no conflicts to disclose.

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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