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Experimental Conditions for Observation of Thermodynamic Instability and Critical Point of Fine Particle (Dusty) Plasmas

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When the Coulomb coupling between fine particles becomes sufficiently strong in fine particle plasmas, the isothermal compressibility of the whole system diverges and we have a phase separation and an associated critical point. Experimental conditions of fine particle plasmas, densities and temperatures of components and the fine particle size, are obtained corresponding to characteristic parameters around the critical point and the dependency on ion species and other factors is discussed.

1. INTRODUCTION

Fine particle plasmas (dusty plasmas) are charge-neutral mixtures of macroscopic fine particles (dust particles, particulates), ions, and electrons. They have been investigated as one of typical and important examples of strongly coupled plasmas where each particle can be directly observed enabling kinetic analyses of various phenomena in strongly coupled systems[1].

Fine particles in these plasmas can be approximately modeled as Yukawa particles with hard cores embedded in the ambient plasma composed of ions and electrons[2, 3, 4]. When the latter is regarded as an inert background, we call the system of Yukawa particles the Yukawa one-component plasma (OCP).

The isothermal compressibility of OCP generally diverges at some not-so-large value of the Coulomb coupling in both classical and quantum cases. The isothermal compressibility of Yukawa OCP which is characterized by the strength of Coulomb coupling and the strength of screening also diverges with the increase of the Coulomb coupling[5].

In ordinary systems, the divergence of the isothermal compressibility means a thermodynamic instability which leads to phase separations and possible critical points. In the case of OCP, however, the compressibility is defined with the assumption that the background automatically follows the deformation. Therefore, in most systems modeled as OCP, this instability is suppressed by the real background which is almost incompressible. In order to explore the possibility of observing phenomena related to this instability, it is essential to take the background into account as a real entity to the system.

In the system of Yukawa particles embedded in ambient plasma, there is a possibility that the total isothermal compressibility diverges when the coupling of Yukawa particles is sufficiently strong[5, 6]. (In Ref.[6], the applicability of our analysis has also been discussed in relation to properties of fine particle plasmas which are not simply represented by the Yukawa system.) We have a phase separation with a critical point and largely enhanced density fluctuations are expected when we approach the critical point[6]. The enhancement of the same nature was first predicted for Coulombic OCP in a deformable background[7].

From the analysis of thermodynamic functions, phase diagrams are given in terms of dimensionless characteristic parameters. In order to make observations, it is necessary to interpret characteristic parameters into experimental conditions. In this article, we mainly discuss this correspondence[8] and analyze the effects of ion species and other factors related to experiments, focusing our attention to the critical point in fine particle plasmas.

2. MODEL

We consider the system composed of N_i ions (i) with the charge e, N_e electrons (e) with the charge $-e$, and N_p fine particles (p) with the charge $-Qe$ in a volume V, satisfying the charge neutrality condition for densities

$$en_i + (-e)n_e + (-Qe)n_p = 0, \qquad (1)$$

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where $n_i = N_i/V$, $n_e = N_e/V$, and $n_p = N_p/V$. We assume three components have different temperatures, T_p, T_i, and T_e as usually observed in experiments.

Based on the fact that $n_i,\ n_e \gg n_p$, we take the statistical average with respect to the coordinates of electrons and ions and obtain an expression for the Helmholtz free energy of the system[2, 3]. The effective interaction energy for fine particles is given by[4]

$$U_p = U_{coh} + U_{sheath}, \tag{2}$$

$$U_{coh} = \frac{1}{2}\int\int d\mathbf{r}d\mathbf{r}'\frac{e^{-|\mathbf{r}-\mathbf{r}'|/\lambda}}{|\mathbf{r}-\mathbf{r}'|}\rho(\mathbf{r})\rho(\mathbf{r}') - (\text{self-interactions}). \tag{3}$$

Here the charge density $\rho(\mathbf{r})$ is defined by

$$\rho(\mathbf{r}) = \sum_{i=1}^{N_p}(-Qe)\delta(\mathbf{r}-\mathbf{r}_i) + Qen_p \tag{4}$$

including the average charge density the background plasma Qen_p, and λ is the screening length in the background plasma given by

$$\lambda = \left(\frac{4\pi n_i e^2}{k_B T_i} + \frac{4\pi n_e e^2}{k_B T_e}\right)^{-1/2}. \tag{5}$$

The term U_{sheath} is the (free) energy associated with the sheath around fine particles and does not depend on their mutual configuration. Fine particles thus interact via the repulsive Yukawa interaction

$$\frac{(Qe)^2}{r}\exp(-r/\lambda) \tag{6}$$

and, at the same time, are *effectively confined by the average background charge density*. We note that this confinement comes from the condition of the charge neutrality.

In calculating thermodynamic quantities of our system, we use the results of numerical simulations on the Yukawa OCP and explicitly take into account the contribution of the background plasma. We also take the finite radius of fine particles into account assuming they are spheres of the same size[6].

In our model, the statistical properties are described in terms of four dimensionless characteristic parameters,

$$\Gamma = \frac{(Qe)^2}{ak_BT_p}, \tag{7}$$

$$\xi = \frac{a}{\lambda}, \tag{8}$$

$$\Gamma_0 = \frac{(Qe)^2}{r_pk_BT_p} = \Gamma\frac{a}{r_p}, \tag{9}$$

and

$$A = \frac{n_ik_BT_i + n_ek_BT_e}{n_pk_BT_p} \gg 1. \tag{10}$$

Here $a = (3/4\pi n_p)^{1/3}$ is the mean distance between particles and r_p is the radius of fine particle cores. Since ion and electron densities are much larger than that of fine particles in usual cases, we have the inequality in (10).

Within some appropriate approximations, we obtain an expression for the Helmholtz free energy of our system and other thermodynamic quantities[6]. Due to the inequality (10), thermodynamic quantities have the the functional form as shown below for the (total) pressure p_{tot}:

$$\frac{p_{tot}}{n_pk_BT_p} \approx \frac{A}{1-\eta} + \frac{p_p}{n_pk_BT_p}, \tag{11}$$

$$\begin{aligned}\frac{p_p}{n_pk_BT_p} \approx\ & \frac{1+\eta+\eta^2-\eta^3}{(1-\eta)^3} \\ & + a_1\tilde{\Gamma}e^{a_2\xi}\left(\frac{1}{3}+\frac{1}{6}a_2\xi+\frac{\tilde{r}_p^2}{1+\tilde{r}_p}\right) \\ & + a_3\tilde{\Gamma}^{1/4}e^{a_4\xi}\left(\frac{1}{3}+\frac{2}{3}a_4\xi+\frac{\tilde{r}_p^2}{1+\tilde{r}_p}\right) \\ & + \frac{3}{2}\tilde{\Gamma}\xi^{-2}\frac{\tilde{r}_p^2}{1+\tilde{r}_p}(1+e^{-2\tilde{r}_p}) \\ & - \frac{1}{4}\tilde{\Gamma}\xi e^{-2\tilde{r}_p},\end{aligned} \tag{12}$$

where $a_1 = -0.896$, $a_2 = -0.588$, $a_3 = 0.72$, $a_4 = -0.22$,

$$\eta = \left(\frac{\Gamma}{\Gamma_0}\right)^3, \tag{13}$$

$$\frac{\tilde{\Gamma}}{\Gamma} = \frac{e^{2\tilde{r}_p}}{(1+\tilde{r}_p)^2}, \tag{14}$$

and

$$\tilde{r}_p = \frac{r_p}{\lambda}. \tag{15}$$

In order to have thermodynamic instability of our system, it is necessary to have

$$\Gamma_0 > \Gamma \geq A \tag{16}$$

and combined with the inequality $A \gg 1$, we have $\Gamma \gg \Gamma^{1/4}$. In this case, $(p_{tot}/n_pk_BT_p)/A$ and other properly normalized thermodynamic quantities are approximately expressed as functions of $(\Gamma/A,\ \xi,\ \Gamma_0/A)$.

3. PHASE DIAGRAM

The pressure of Yukawa particulates p_p takes on increasingly negative values with the increase of the Coulomb coupling Γ, the main contribution coming from the negative term with a_1. Though the first term due to the background plasma on the right-hand side of (11) is positive and large, the total pressure p_{tot} becomes negative, when the coupling becomes sufficiently strong and p_p overcomes the positive contribution from ambient plasma of ions and electrons.

The inverse isothermal compressibility also decreases and eventually vanishes due to negative contribution from fine particles. The total isothermal compressibility thus diverges. In principle, the zero pressure and the zero inverse isothermal compressibility are independent phenomena. As a matter of fact, however, we have these zeros at similar strengths of coupling. The latter leads to a phase separation and related critical point[5, 6]. Near the critical point, we expect enhanced density fluctuations[6].

This is the same phenomenon that was predicted for the case of OCP some years ago by the author[7]: While its observation is almost impossible in simple OCP's, it is difficult but possible in fine particle plasmas.

Example of phase diagrams in the $(\Gamma/\Gamma_0,\ \xi)$-plane and the $(p_{tot}, \Gamma/\xi^2)$-plane are shown in Fig.1. In the $(\Gamma,\ \xi)$ plane, we have a domain where phases with higher and lower densities coexist. In the $(\Gamma/\xi^2,\ p_{tot})$-plane, we have a line where two phases coexist, terminating at the critical point. The former is analogous to the density-temperature diagram and the latter, to the pressure-temperature diagram for the usual gas-liquid transition. The locus of critical point is also shown in Fig.1.

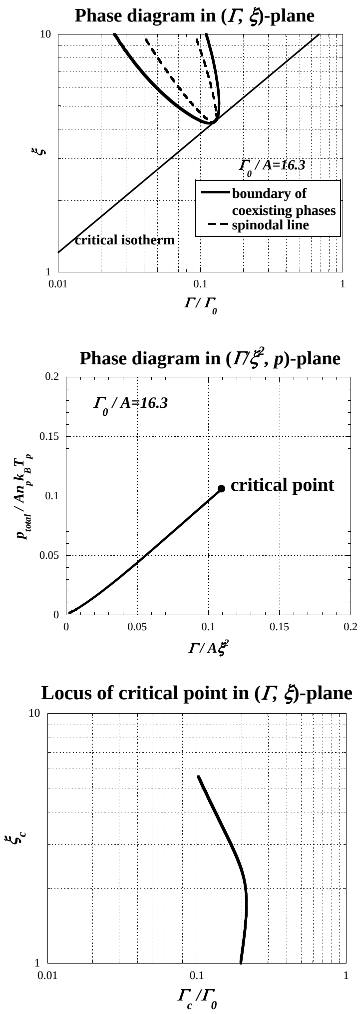

Fig. 1 Phase diagrams and locus of critical point.

4. CORRESPONDENCE BETWEEN EXPERIMENTAL CONDITIONS AND CHARACTERISTIC PARAMETERS: GENERAL PROPERTIES

In order to perform experiments, we have to interpret characteristic parameters into experimental conditions. Characteristic parameters are readily calculated from experimental conditions by definitions but the reverse needs at least some manipulation and numerical solution of equations[8].

In most experiments, it is observed that $T_e > T_i$. As for T_i and T_p, usually $T_i \sim T_p$ is implicitly assumed but there seem to exist the cases where they are different. We have therefore assigned different values for the temperatures of the components in our model. In total, we have eight parameters for charged components of the system, $(r_p,\ n_p,\ T_p,\ Q,\ n_i,\ T_i,\ n_e,\ T_e)$.

The above experimental conditions are related to dimensionless characteristic parameters of our system. Let us list the conditions which determine the above eight quantities.
(1) We assume that the charge neutrality condition (1) is satisfied.
(2) When characteristic parameters $(\Gamma,\ \xi)$ are specified, we have two conditions.
(3) We specify values of r_p, Γ_0, $A = (n_eT_e + n_iT_i)/n_pT_p$.
(4) The charge on a fine particle $-Qe$ is determined by the balance between the fluxes of ions and electrons onto the surface of fine particles. Introducing f_Q by

$$Q = f_Q \frac{k_BT_e}{e^2/r_p}, \tag{17}$$

we have

$$\begin{aligned} n_e \left(\frac{k_BT_e}{m_e}\right)^{1/2} \exp\left(-\frac{f_Q}{1+\tilde{r}_p}\right) \\ -n_i \left(\frac{k_BT_i}{m_i}\right)^{1/2} \left(1 + \frac{f_Q}{1+\tilde{r}_p}\frac{T_e}{T_i}\right) \\ = 0 \end{aligned} \tag{18}$$

in the orbital-motion-limited (OML) theory. (The applicability of the OML theory has been discussed in [8].) We also take into account the effect of finite radius on the surface potential of fine particles.

The above conditions give seven relations for eight experimental parameters. As the last condition, we take the ratio of the ion and fine particle temperatures

$$\tau_{ip} = \frac{T_i}{T_p}. \tag{19}$$

In principle, τ_{ip} is determined by other parameters through the energy relaxation processes which include neutral atoms. We here treat this ratio as an externally determined parameter instead of giving other conditions to determine τ_{ip}.

4.1 General relations

We first express n_p, $n_i/(A/\tau_{ip})$, $n_e/(A/\tau_{ip})$, $T_i/(A/\tau_{ip}) = \tau_{ip}T_p/(A/\tau_{ip})$, and $T_e/(A/\tau_{ip})$ explicitly in terms of r_p, Γ/A, ξ, Γ/Γ_0, and f_Q[8]. Substituting these expressions into (18), we have an equation for f_Q which has to be solved numerically. We note that, since (18) includes only the ratios n_e/n_i and T_e/T_i which are independent of r_p or A/τ_{ip}, the value of f_Q is determined self-consistently when the set of values $(\Gamma/A,\ \xi,\ \Gamma/\Gamma_0)$ is specified.

Let us now introduce n_0 and $E_0 = k_BT_0$ defined respectively by $n_0 = 3/4\pi r_p^3$ and $E_0 = k_BT_0 = e^2/r_p$. We also define $A' = A/\tau_{ip}$. Then the values of $(f_Q,\ n_p/n_0,\ (n_i/n_0)/A',\ (n_e/n_0)/A',\ (T_i/T_0)/A' = \tau_{ip}(T_p/T_0)/A',\ (T_e/T_0)A')$ are determined by the set of values $(\Gamma/A,\ \xi,\ \Gamma/\Gamma_0)$ *irrespective of the values of* $(r_p,\ A' = A/\tau_{ip})$: We may thus regard r_p, A, and τ_{ip} as a kind of *adjustable* parameters which can be chosen so as to satisfy the experimental requirement for densities or temperatures.

4.1.1 effect of fine particle radius

When the values of $(\Gamma/A,\ \xi,\ \Gamma/\Gamma_0)$ are specified and the temperatures are given, the dependency on the particle radius is

$$n_p \propto r_p^3, \tag{20}$$

$$n_i,\ n_e \propto r_p^2, \tag{21}$$

$$\frac{A}{\tau_{ip}} \propto r_p, \tag{22}$$

$$\frac{\Gamma}{\tau_{ip}},\ \frac{\Gamma}{\tau_{ip}} \propto r_p. \tag{23}$$

4.1.2 effect of ion mass and electron temperature

The mass of ions influences the charging of particles determined by (18) through the thermal velocity of ions. The ratio n_e/n_i corresponding to given values of f_Q and T_e/T_i becomes smaller with the increase of the ion mass. Since the rare gas is used as the neutral gas which fills the chamber, the mass of ions takes the values 6.64×10^{-24}g (He), 3.35×10^{-23}g (Ne), 6.63×10^{-23}g (Ar), 1.392×10^{-22}g (Kr), 2.18×10^{-22}g (Xe), 3.69×10^{-22}g (Rn).

4.2 Domain of characteristic parameters in fine particle plasmas

When we express $n_e/(A/\tau_{ip})$ in terms of r_p, Γ/A, ξ, Γ/Γ_0, and f_Q, we note that, since $n_e \geq 0$, we have to satisfy the condition $(1/3)\xi^2/(\Gamma/A) \geq 1$ or

$$\xi^2 = \left(\frac{a}{\lambda}\right)^2 \geq 3\left(\frac{\Gamma}{A}\right). \tag{24}$$

Thus the lower limit of realizable ξ is determined by Γ. Though some restriction is naturally expected from the condition of charge neutrality, the explicit expression in terms of characteristic parameters has been given for the first time in Ref. [8]. In the case where Γ is not so large, this inequality is not effective in restricting the domain of characteristic parameters. In the case of very strong coupling where $\Gamma \sim A$, on the contrary, the values of characteristic parameters are limited by the condition (24) as will be shown in what follows.

5. EXPERIMENTAL CONDITIONS AT CRITICAL POINT

5.1 The case of Argon

Let us now assume that the gas is Ar, ions are Ar^+, $r_p = 10\mu$m, and the electron temperature is $T_e = 1$eV. Along the locus of the critical point, we obtain experimental conditions by the above mentioned procedure. We show some examples of experimental parameters at the critical point in Fig.4. Due to the condition (1), the experiments of the critical point with fine particle plasmas are possible on the lines plotted in these figures.

When we increase the radius of fine particles, the densities of fine particles, ions and electrons changes as $n_p \propto r_p^{-3}$, $n_i, n_e \propto r_p^{-2}$. We plot the values for $r_p = 20\mu$m and $T_e = 1$eV in Fig.5. The condition on the densities seems to become easier for $r_p = 20\mu$m.

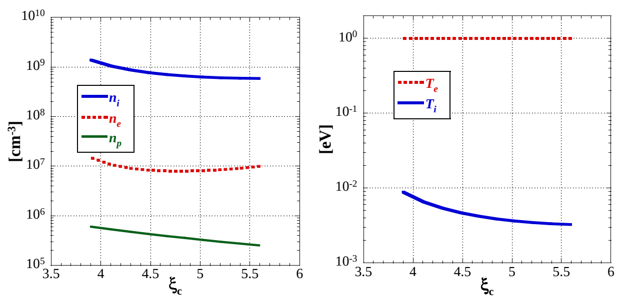

Fig. 2 Ar with $r_p = 10\mu$m and $T_e = 1$eV. Densities (left) and temperatures (right).

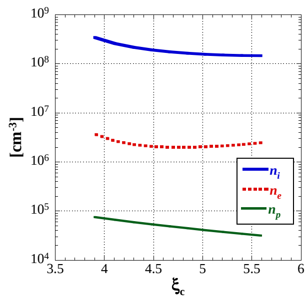

Fig. 3 Ar, $r_p = 20\mu$m, $T_e = 1$eV.

5.2 Effect of ion mass and electron temperature

We now analyze the effect of the mass of ion on experimental conditions by changing the gas from Ar to Xe and ions from Ar^+ to Xe^+. The results are shown in Figs.6 and 7. We observe that the condition on the ion and electron densities become easier. On the other hand, the ion temperature becomes too low and we may have difficulty to satisfy the condition.

The increase of the electron temperature also increases the ion temperature but, at the same time, the ion and electron densities also increase. We here give an example where both the fine particle radius and the electron temperature are increased.

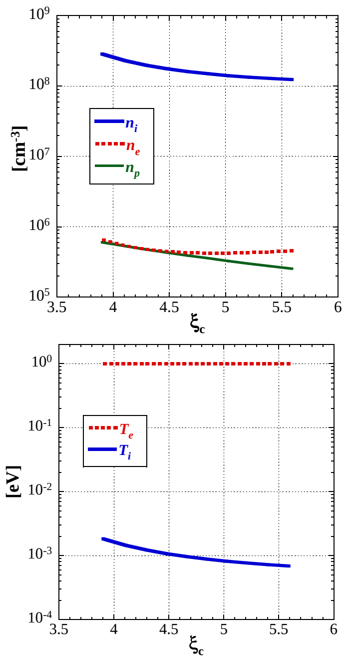

Fig. 4 Xe with $r_p = 10\mu$m and $T_e = 1$eV.

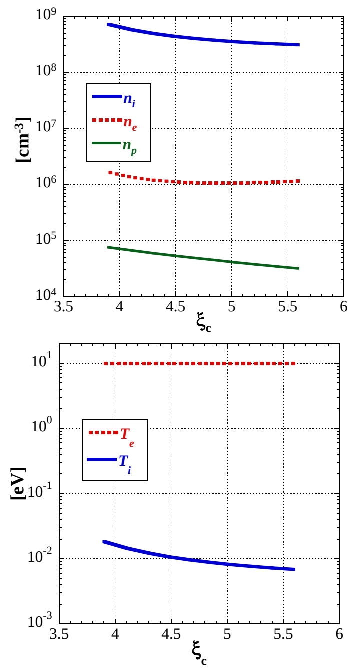

Fig. 5 Xe with $r_p = 20\mu$m and $T_e = 10$eV.

5.3 Optimal conditions for experiments

Comparing the results in Figs.4-7, we may tentatively conclude that the experiments with larger particles, heavier gas, and higher electron temperatures seem to be desirable to realize characteristic parameters around the critical point. In order to have large particles afloat in the background plasma, we have to compensate the effect of gravity. We hope the experiments under microgravity or the compensation by the temperature gradient may provide such an environment.

6. CONCLUSION

We have shown that the intrinsic thermodynamic instability of OCP and related critical phenomena can possibly be realized in experiments with fine particle plasmas and have given corresponding experimental conditions with discussions on various possibilities. In order to observe phenomena near the critical point, it is necessary to have a bulk isotropic three-dimensional system of fine particle plasmas in the domain of very strong coupling. Though it is difficult to realize such a system on the ground due to gravity on fine particles, we expect the experiment under microgravity may provide a chance of observation.

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