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Thermodynamic instability and the critical point of fine particle (dusty) plasmas: enhancement of density fluctuations and experimental conditions for observation

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Abstract
We analyze thermodynamics of fine particle (dusty) plasmas, regarding them as systems composed of charged particles with hard cores interacting via the repulsive Yukawa potential and the ambient plasma (of ions and electrons), taking the contribution of the latter properly into account. When the Coulomb coupling between fine particles becomes sufficiently strong, the isothermal compressibility of the whole system diverges and we have a phase separation and an associated critical point. The enhancement of long-wavelength density fluctuations near the critical point is shown. 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 dependence on ion species and other factors is discussed. These phenomena will be observed in the bulk three-dimensional system which may be realized on the ground by somehow canceling the effect of gravity on fine particles. Experiments under the microgravity environment are naturally expected to provide a chance of observation.

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(Some figures in this article are in colour only in the electronic version)

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 the 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 [2–4]. 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 Yukawa OCP generally diverges with the increase of the Coulomb coupling [5]. We emphasize that, 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. This instability is suppressed when the background is almost incompressible as in the usual case of systems modeled by OCP. In the system of Yukawa particles embedded in ambient plasma, the total isothermal compressibility possibly diverges when the coupling of Yukawa particles is sufficiently strong [5, 6]. (In [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 near the critical point [6]. We here give an example of the enhancement factor.

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. We discuss this correspondence [7] and analyze the effects of ion species and other factors related to experiments.

When fine particles have mutual attraction, the vapor–liquid transition and critical phenomena are expected [8]. The condition for the existence of such an attraction, however, seems to be not easy to realize. Though we have to take the ambient background plasma into account, the explicit attractive interactions are not assumed (nor excluded) in our analysis. Possible existence of attraction may make the requirement on characteristic parameters for critical point less severer.

2. The equation of state of Yukawa particulates and ambient background plasma

We consider the system in a volume V 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 $-Qe$, satisfying the charge neutrality condition for densities

$$en_i + (-e)n_e + (-Qe)n_p = 0, \tag{1}$$

where $n_i = N_i/V$, $n_e = N_e/V$ and $n_p = N_p/V$. We assume that $n_i, n_e \gg n_p$ and take the statistical average with respect to electrons and ions to have an expression for the Helmholtz free energy of the system. The effective interaction energy for fine particles is given by [4]

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

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

Here the charge density $\rho(\mathbf{r}) = \sum_{i=1}^{N_p}(-Qe)\delta(\mathbf{r}-\mathbf{r}_i) + Qen_p$ includes that of the background plasma Qen_p and $\lambda = (4\pi n_i e^2/k_BT_i + 4\pi n_e e^2/k_BT_e)^{-1/2}$. The term U_{sheath} is the (free) energy associated with the sheath around fine particles. Fine particles interact via the repulsive Yukawa interaction and *effectively confined* by the average charge density of background plasma composed of ions and electrons. It should be noted that *this effective confinement comes from the charge neutrality of the whole system.* We explicitly take into account the contribution of the background plasma to the equation of state. We also take the finite radius of fine particles into account assuming they are spheres of the same size [6].

Our system is characterized by four dimensionless parameters:

$$\Gamma = \frac{(Qe)^2}{ak_BT_p}, \qquad \xi = \frac{a}{\lambda}, \qquad \Gamma_0 = \frac{(Qe)^2}{r_pk_BT_p} = \Gamma\frac{a}{r_p}, \qquad A = \frac{n_ik_BT_i + n_ek_BT_e}{n_pk_BT_p} \gg 1, \tag{4}$$

where $a = (3/4\pi n_p)^{1/3}$ is the mean distance between particles and r_p is the radius of core of fine particles. We assume three components have different temperatures, T_p, T_i and T_e as usually observed in experiments.

Within some appropriate approximations, we obtain an expression for the Helmholtz free energy of our system and other thermodynamic quantities [6]. The total pressure is given by

$$\frac{p_{\text{tot}}}{n_pk_BT_p} \approx \frac{A}{1-\eta} + \frac{p_p}{n_pk_BT_p}, \tag{5}$$

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

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

$$\eta = \left(\frac{\Gamma}{\Gamma_0}\right)^3, \qquad \frac{\tilde{\Gamma}}{\Gamma} = \frac{\mathrm{e}^{2\tilde{r}_p}}{(1+\tilde{r}_p)^2}, \qquad \tilde{r}_p = \frac{r_p}{\lambda}. \tag{7}$$

3. Phase diagrams

The pressure of Yukawa particulates p_p takes on increasingly negative values with the increase of the Coulomb coupling Γ. The main contribution comes from the term with a_1 which is negative. Though the first term on the right-hand side of (5) 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. Since $\Gamma, \tilde{\Gamma} \propto Q^2$ and $Q \sim 10^3$–$10^4 \gg 1$, this is possible even when $n_i, n_e \gg n_p$.

We note that the inverse isothermal compressibility also decreases and eventually vanishes due to negative contribution from fine particles. The total isothermal compressibility thus diverges. We have vanishings of the pressure and the inverse isothermal compressibility separately but at similar strengths of coupling. The latter leads to important consequences for thermodynamic behavior of the system, a phase separation and related critical point [5, 6].

Example of phase diagrams are shown in figure 1. In the (Γ, ξ) plane, we have a domain where phases with higher and lower densities coexist. In the $(\Gamma/\xi^2, p_{\text{tot}})$ 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. The locus of critical point in the $(\Gamma/\Gamma_0, \xi)$ plane is shown in figure 2.

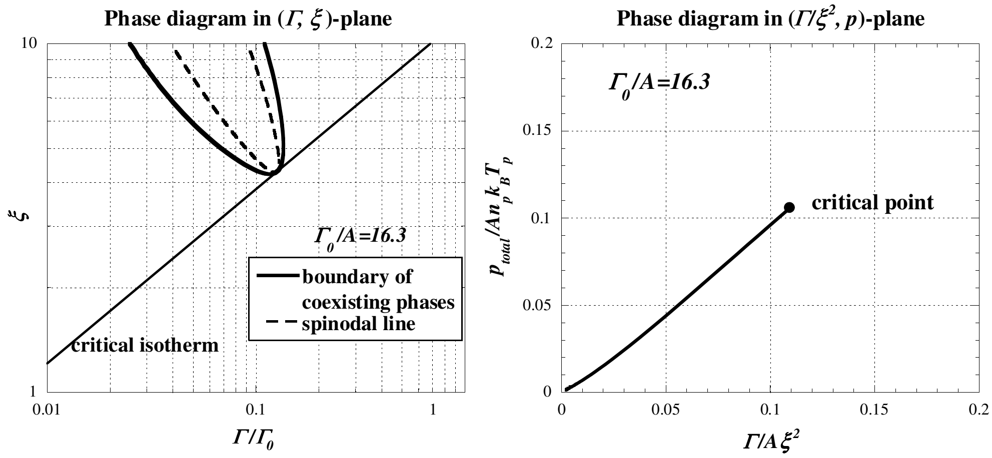

Figure 1. Examples of phase diagrams in $(\Gamma/\Gamma_0, \xi)$ plane and $(\Gamma/\xi^2, p_{\mathrm{tot}})$ plane.

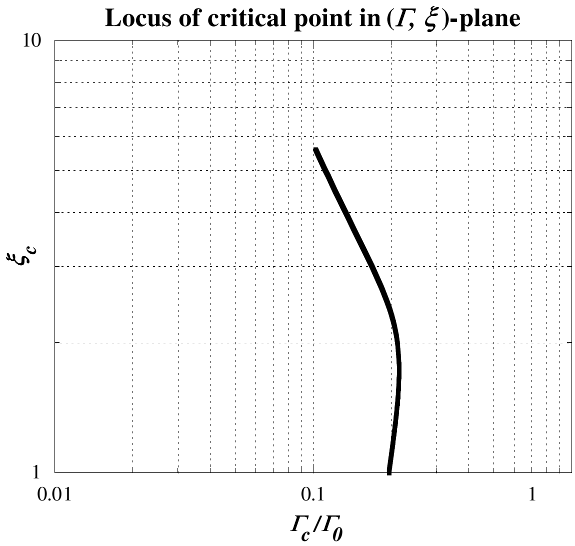

Figure 2. Locus of critical point.

4. Density fluctuations

The static form factor of Yukawa particles $S(k)$ is related to the dielectric response function $\varepsilon(k, \omega = 0)$ describing the response to external Yukawa particle density via the fluctuation–dissipation theorem as [6, 9]

$$S(k) = \frac{k^2 + 1/\lambda^2}{k_D^2}\left[1 - \frac{1}{\varepsilon(k, \omega = 0)}\right], \tag{8}$$

where $k_D^2 = 4\pi n_p (Qe)^2/k_B T_p$. In the limit of long wavelengths, the balance of the external force and the pressure gradient gives [6],

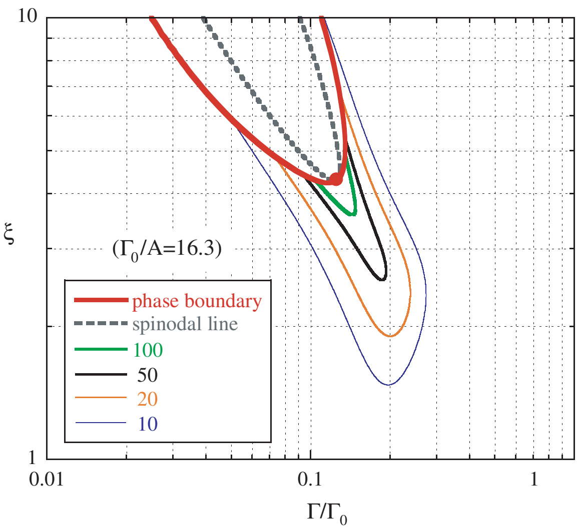

Figure 3. Density fluctuation enhancement. Numbers are enhancement factors in amplitude squared.

$$\frac{1}{S(k)} \sim -\frac{V}{n_p k_B T_p}\left(\frac{\partial p_{\text{tot}}}{\partial V}\right)_{T_i,T_e,T_p} + \mathcal{O}(k^2). \tag{9}$$

(We obtain not exactly the same but similar results from the thermodynamic perturbation [6].) Density fluctuations are thus enhanced near the critical point as shown in figure 3.

It should be noted that, within the treatment as Yukawa OCP, the response to the external field is related to the 'screened response' giving

$$\frac{1}{S_{\text{OCP}}(k)} \sim k_D^2\lambda^2 - \frac{V}{n_p k_B T_p}\left(\frac{\partial p_p}{\partial V}\right)_{T_p} + \mathcal{O}(k^2). \tag{10}$$

Therefore *the divergence of the isothermal compressibility of Yukawa OCP,* $-(\partial V/\partial p_p)_{T_p}$, *does not directly lead to the enhancement of the density fluctuation* [6, 9, 10]. As has been discussed in the case of Coulombic OCP, we need a freely deformable background in order for the divergence of the isothermal compressibility of Yukawa OCP to give such an enhancement [10].

5. Experimental conditions

In describing statistical properties of fine particle plasmas dimensionless characteristic parameters are used. 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 [7].

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 here assign different values for the temperatures of the components. In total, we have eight parameters for charged components of the system, $(r_p, n_p, T_p, Q, n_i, T_i, n_e, T_e)$.

Let us list the conditions which determine the above eight quantities. We assume that the charge neutrality condition (1) is satisfied. When characteristic parameters (Γ, ξ) are specified, we have two conditions. 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 $Q = f_Q k_B T_e/(e^2/r_p)$, we have

$$n_e \left(\frac{k_B T_e}{m_e}\right)^{1/2} \exp\left(-\frac{f_Q}{1+\tilde{r}_p}\right) - n_i \left(\frac{k_B T_i}{m_i}\right)^{1/2} \left(1 + \frac{f_Q}{1+\tilde{r}_p}\frac{T_e}{T_i}\right) = 0 \tag{11}$$

in the orbital-motion-limited (OML) theory. (The applicability of the OML theory has been discussed in [7].) We also take into account the effect of finite radius on the surface potential of fine particles.

The above four conditions, (1), Γ, ξ and (11), for eight experimental parameters leave four degrees of freedom. We represent them by r_p, Γ_0, $A = (n_e T_e + n_i T_i)/n_p T_p$, and the ratio of the ion and fine particle temperatures

$$\tau_{ip} = \frac{T_i}{T_p}. \tag{12}$$

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}.

We first note that $n_p, n_i/(A/\tau_{ip}), n_e/(A/\tau_{ip}), T_i/(A/\tau_{ip}) = \tau_{ip} T_p/(A/\tau_{ip})$, and $T_e/(A/\tau_{ip})$ are explicitly expressed in terms of $r_p, \Gamma/A, \xi, \Gamma/\Gamma_0$, and f_Q [7]. We also point out that, since $n_e \geqslant 0$, we have to satisfy the condition $(1/3)\xi^2/(\Gamma/A) \geqslant 1$ or

$$\xi^2 = \left(\frac{a}{\lambda}\right)^2 \geqslant 3\left(\frac{\Gamma}{A}\right). \tag{13}$$

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 [7].

Substituting expressions for $n_p, n_i/(A/\tau_{ip}), n_e/(A/\tau_{ip}), T_i/(A/\tau_{ip}) = \tau_{ip} T_p/(A/\tau_{ip})$ and $T_e/(A/\tau_{ip})$, we have an equation for f_Q which has to be solved numerically. We note that, since (11) 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 $E_0 = k_B T_0$ defined respectively by $n_0 = 3/4\pi r_p^3$ and $E_0 = k_B T_0 = e^2/r_p$. 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* $(r_p, A/\tau_{ip})$: 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 conditions for densities or temperatures.

5.1. Typical examples for 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 figure 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 figure 5. The condition on the densities seems to become easier for $r_p = 20\,\mu$m.

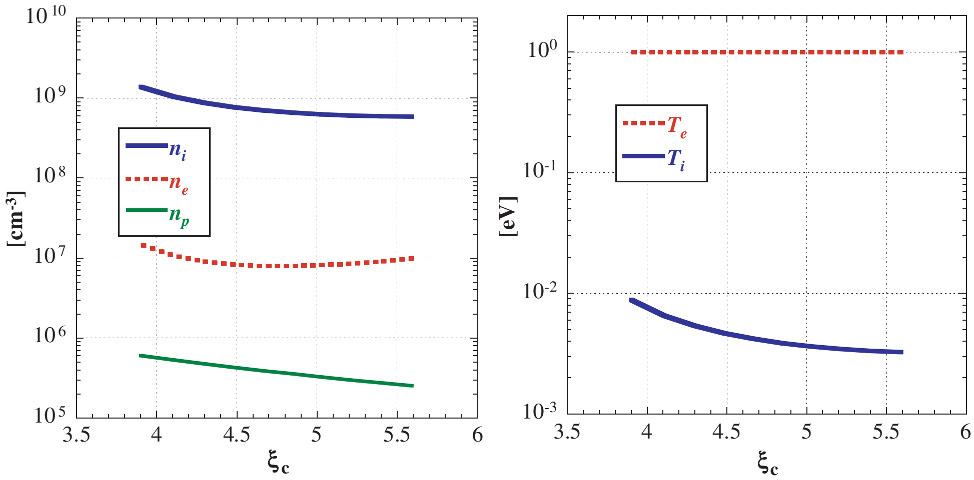

Figure 4. Ar with $r_p = 10\,\mu$m and $T_e = 1$ eV; n_i, n_e, n_p (left) and T_i, T_e (right).

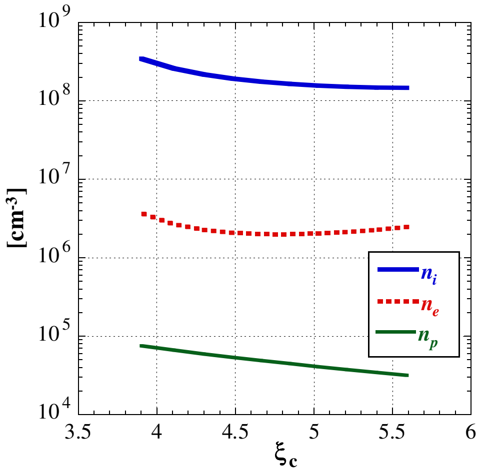

Figure 5. Ar, $r_p = 20\,\mu$m, $T_e = 1$ eV.

5.2. The 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 figures 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.

5.3. Optimal conditions for experiments

Comparing the results in figures 4–7, we may tentatively conclude that the experiments with larger particles, heavier gas and higher electron temperatures seem to be desirable to realize

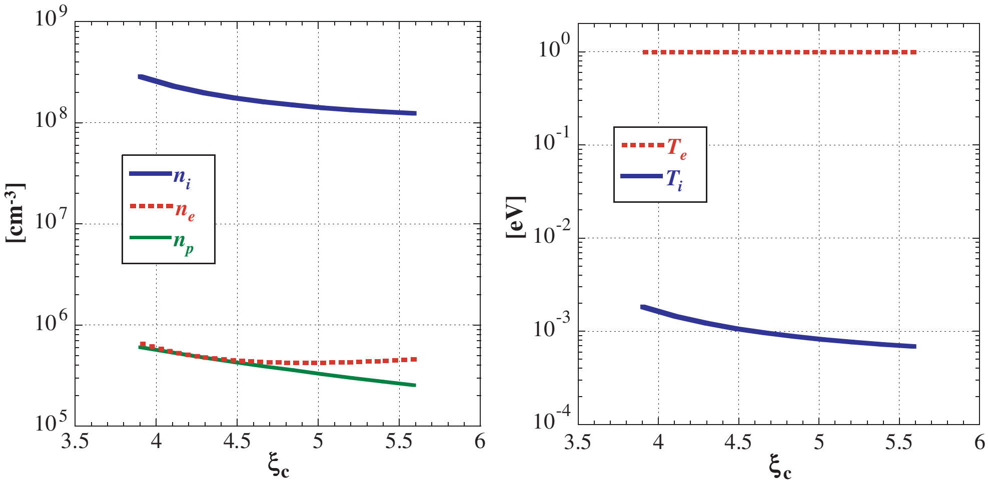

Figure 6. Xe with $r_p = 10\,\mu$m and $T_e = 1$ eV.

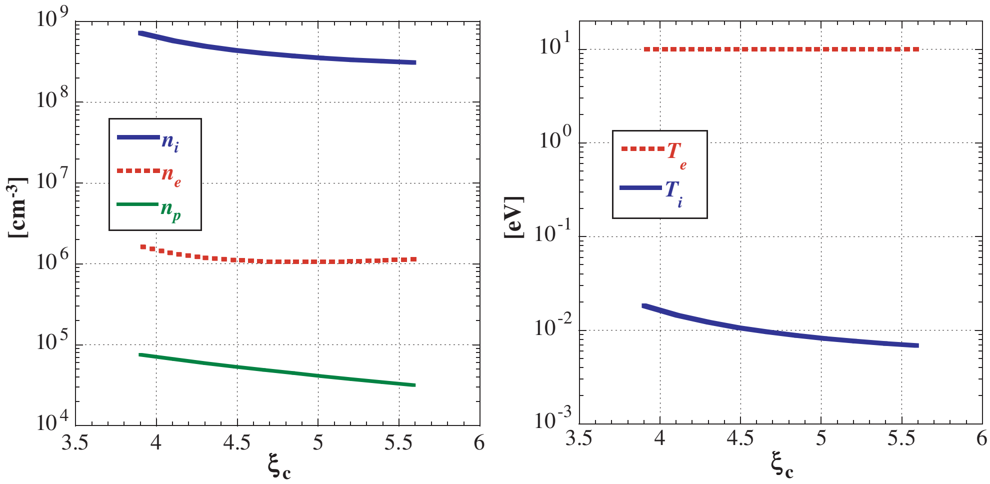

Figure 7. Xe with $r_p = 20\,\mu$m and $T_e = 10$ eV.

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 fine particle plasmas and given corresponding experimental conditions with discussions on various possibilities. To observe phenomena near the critical point, we need to have a bulk isotropic three-dimensional system of fine particle plasmas in the domain of very strong coupling. On the ground, we may apply thermal or other forces to cancel the effect of gravity. Experiments under microgravity are naturally expected to provide a chance of observation.

Acknowledgments

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