J. Plasma Fusion Res. SERIES, Vol. 8 (2009)

Phase Diagram of Strongly Coupled Yukawa Particulates in Deformable Background and Application to Fine Particle (Dusty) Plasmas

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(Received: 1 September 2008 / Accepted: 19 January 2009)

We analyze thermodynamics of the system 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 particles becomes sufficiently strong, the isothermal compressibility of the whole system diverges and we have a phase separation and associated critical point. When appropriate conditions are satisfied, the critical point can be in the domain of solid phase of Yukawa particles. An enhancement of the long wavelength density fluctuations near the critical point is expected. We investigate the applicability of these results to fine particle (dusty) plasmas. Experimental conditions of fine particle plasmas, densities and temperatures of components and the fine particle size, are explicitly expressed in terms of dimensionless characteristic parameters corresponding to the critical point. Though it might be difficult to realize such a system on the ground due to large influence of the gravity on fine particles, we expect the experiment under microgravity may provide a chance of observation.

Keywords: Yukawa particle, phase separation, critical point, experimental conditions, fine particle plasma

1. Introduction

Fine particle plasmas (dusty plasmas) are charge neutral mixtures of macroscopic fine particles (dust particles, particulates), ions, and electrons. Fine particle plasma is one of important examples of strongly coupled plasmas. In particular, each fine particle can be directly observed by CCD cameras or even by naked eyes enabling kinetic analyses of various phenomena in strongly coupled systems.

Fine particles in fine particle plasmas can be approximately modeled as Yukawa particles with hard cores embedded in the ambient plasma of ions and electrons. When the latter serves only as an inert background, we call the system of Yukawa particles the Yukawa one-component plasma (OCP).

The isothermal compressibility of OCP generally diverges with the increase of the Coulomb coupling and this holds also for the Yukawa OCP. In order to observe this thermodynamic instability, however, it is necessary to take the background into account as a real physical entry to the system. In other words, we have to take the deformation of the background into account. This instability is suppressed when the background is almost incompressible as in the usual case of systems modeled by OCP and this is the reason why this long-known instability has never been observed in experiments.

In the system of Yukawa particles embedded in ambient plasma, there is a possibility of deformation of the background and it is shown that the total isothermal compressibility diverges when the coupling of Yukawa particles is sufficiently strong[1, 2]. We have a phase separation and related critical point. When we approach the critical point, the density fluctuations of the system are largely enhanced.

In order to realize such an instability in experiments, it is necessary to interpret dimensionless characteristic parameters into experimental conditions. We discuss the correspondence between experimental conditions and characteristic parameters[3] and give examples of possible combinations of experimental conditions at the critical point.

Fine particle plasmas are not completely described by our model. We discuss some of their important aspects which are not included in our treatment and confirm the applicability of our analyses to fine particle plasmas

2. 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

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

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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]

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

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

The charge density $\rho(\mathbf{r}) = \sum_{i=1}^{N_p}(-Qe)\delta(\mathbf{r}-\mathbf{r}_i)+Qen_p$ includes that of background plasma Qen_p and

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

The term U_{sheath} is the (free) energy of the sheath around fine particles. Fine particles are *effectively* confined by background plasma, mutually interacting via the repulsive Yukawa potential. This effective confinement comes from *the charge neutrality of the whole system*: The charge $-Qe$ on fine particles has *attractive* Yukawa interaction with the background charge density Qen_p. 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[2].

Our system is characterized by four parameters:

$$\Gamma = \frac{(Qe)^2}{ak_BT_p},\ \xi = \frac{a}{\lambda},\ \Gamma_0 = \frac{(Qe)^2}{r_pk_BT_p} = \Gamma\frac{a}{r_p},$$
$$A = \frac{n_ek_BT_e + n_ik_BT_i}{n_pk_BT_p} \gg 1, \tag{5}$$

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.

We obtain an approximate expression for the Helmholtz free energy of our system and other thermodynamic quantities[2]. With explicit consideration of the deformation of background, the total pressure is given by

$$\frac{p_{tot}}{n_pk_BT_p} \approx \frac{A}{1-\eta} + \frac{p_p}{n_pk_BT_p}, \tag{6}$$

$$\frac{p_p}{n_pk_BT_p} \approx \frac{1+\eta+\eta^2-\eta^3}{(1-\eta)^3} + a_1\tilde{\Gamma}e^{a_2\xi}\left(\frac{1}{3}+\frac{1}{6}a_2\xi+\frac{\tilde{r}_p^2}{1+\tilde{r}_p}\right) + a_3\tilde{\Gamma}^{1/4}e^{a_4\xi}\left(\frac{1}{3}+\frac{2}{3}a_4\xi+\frac{\tilde{r}_p^2}{1+\tilde{r}_p}\right) + \frac{3}{2}\tilde{\Gamma}\xi^{-2}\frac{\tilde{r}_p^2}{1+\tilde{r}_p}(1+e^{-2\tilde{r}_p}) - \frac{1}{4}\tilde{\Gamma}\xi e^{-2\tilde{r}_p}, \tag{7}$$

where

$$\eta = \left(\frac{\Gamma}{\Gamma_0}\right)^3,\ \frac{\tilde{\Gamma}}{\Gamma} = \frac{e^{2\tilde{r}_p}}{(1+\tilde{r}_p)^2},\ \tilde{r}_p = \frac{r_p}{\lambda} \tag{8}$$

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

3. Phase Diagrams

Since a_1 is negative, the pressure of Yukawa particulates p_p takes on increasingly negative values when the Coulomb coupling Γ increases. For sufficiently large Γ, the total pressure vanishes and even becomes negative: The negative contribution of fine particles overcomes the positive contribution from ambient plasma of ions and electrons. More importantly, the inverse isothermal compressibility vanishes due to negative contribution of fine particles. (The vanishings of the pressure and the inverse isothermal compressibility occur separately but at similar strengths of coupling.) We thus have a separation into phases with high and low densities and related critical point[1, 2].

Examples of phase diagrams are shown in Fig.1. In $(\Gamma/\Gamma_0,\ \xi)$-plane, high and low density phases coexist inside of the thick solid line, the broken line being the spinodal. In $(\Gamma/\xi^2,\ p_{tot})$-plane, the domain of coexistence shrinks to a single line terminating at the critical point. These diagrams correspond to the density-temperature and the pressure-temperature diagrams in usual liquid-gas transitions. The locus of critical point is shown in Fig.2.

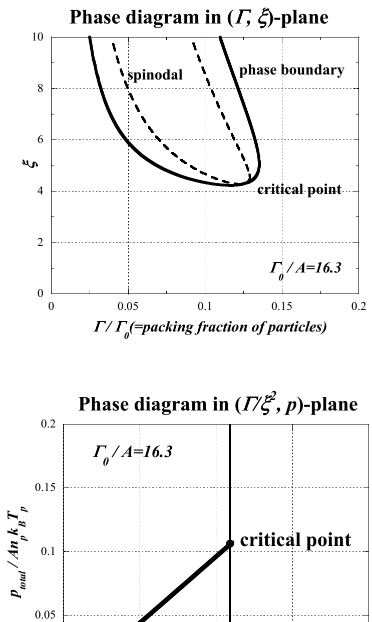

Fig. 1 Examples of phase diagrams. The critical isotherm is also shown in $(\Gamma/\Gamma_0,\ \xi)$-plane (top).

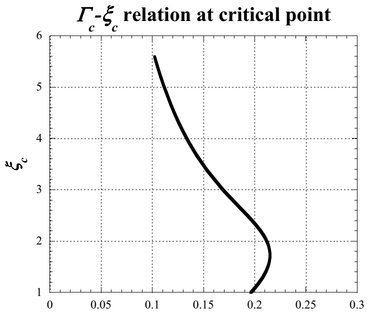

Fig. 2 Relation between Γ and ξ at the critical point.

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

$$S(k) = \frac{k^2 + 1/\lambda^2}{k_D^2}\left[1 - \frac{1}{\varepsilon(k, \omega = 0)}\right], \qquad (9)$$

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

$$\frac{1}{S(k)} \sim -\frac{V}{n_p k_B T_p}\left(\frac{\partial p_{tot}}{\partial V}\right)_{T_i, T_e, T_p} + \mathcal{O}\left(k^2\right). (10)$$

Density fluctuations are thus enhanced near the critical point as shown in Fig.3. We obtain not exactly the same but similar results from the thermodynamic perturbation[2].

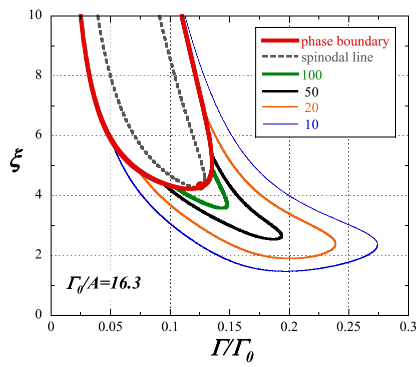

Fig. 3 Density fluctuation enhancement. Numbers are enhancement in amplitude squared.

Within the Yukawa OCP, the response to the external field is related to the 'screened response' and we have

$$\frac{1}{S_{OCP}(k)} \sim k_D^2 \lambda^2 - \frac{V}{n_p k_B T_p}\left(\frac{\partial p_p}{\partial V}\right)_{T_p} + \mathcal{O}\left(k^2\right). (11)$$

The divergence of the isothermal compressibility of Yukawa OCP $-(\partial V/\partial p_p)_{T_p}$ is not directly related to the enhancement of the density fluctuation[5].

5. Experimental Conditions

Thermodynamics of fine particles plasmas are characterized by theoretical parameters and it is necessary to interpret them into experimental parameters. Characteristic parameters are readily calculated from experimental parameters by definitions but the reverse needs at least some manipulation and numerical solution of equations[3].

Since it is believed that $T_e > T_i (\sim T_p)$ in most experiments, we assign different values for the temperatures of the components. We thus have eight parameters for charged components of the system, namely, $(r_p,\ n_p,\ T_p,\ Q,\ n_i,\ T_i,\ n_e,\ T_e)$.

We assume that the charge neutrality condition (1) is satisfied. When characteristic parameters $(\Gamma,\ \xi)$ 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. When we write Q in the form

$$Q = f_Q \frac{k_B T_e}{e^2/r_p} \qquad (12)$$

introducing f_Q, the condition is given by

$$n_e \left(\frac{k_B T_e}{m_e}\right)^{1/2} \exp\left(-\frac{f_Q}{1+\tilde{r}_p}\right) - n_i \left(\frac{k_B T_i}{m_i}\right)^{1/2}\left(1 + \frac{f_Q}{1+\tilde{r}_p}\frac{T_e}{T_i}\right) = 0 \qquad (13)$$

in the orbital-motion-limited (OML) theory. Here the effect of reduction of the electron density is reflected through (1). We take into account the effect of finite radius on the surface potential of the fine particles. The applicability of the OML theory is discussed in [3].

Since we have imposed four conditions, (1), Γ, ξ, and (13) for eight experimental parameters, we are left with four degrees of freedom. As these four, we take the radius 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

$$\tau_{ip} = \frac{T_i}{T_p}. \qquad (14)$$

In principle, the ratio τ_{ip} is determined by other parameters through the energy relaxation processes which include neutral atoms. In most experiments, however, $\tau_{ip} \sim 1$ is implicitly assumed without detailed analysis of the latter processes or temperature measurements. In this paper we treat this ratio as an externally determined parameter expecting to be around unity, 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, Γ/A, ξ, Γ/Γ_0, and f_Q[3]. We also point out that, since $n_e \geq 0$, we have to satisfy the condition $(1/3)\xi^2/(\Gamma/A) \geq 1$ or

$$\xi^2 = \left(\frac{a}{\lambda}\right)^2 \geq 3\left(\frac{\Gamma}{A}\right). \tag{15}$$

This comes from the condition of charge neutrality but has not been explicitly shown in terms of characteristic parameters. Thus the lower limit of realizable ξ is determined by Γ.

Substituting explicit 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 can determine the value of f_Q. We note that, since (13) 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 $n_0 \equiv 3/4\pi r_p^3$ and $E_0 = k_BT_0 \equiv e^2/r_p$. 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.

In Fig.4, we show some examples of experimental parameters at the critical point. We assume the gas is Ar and ions are Ar^+. Along the locus of the critical point, we obtain experimental conditions by the above mentioned procedure. Due to the condition (1), the experiments of the critical point with fine particle plasmas are possible on the lines plotted in Figs.3.

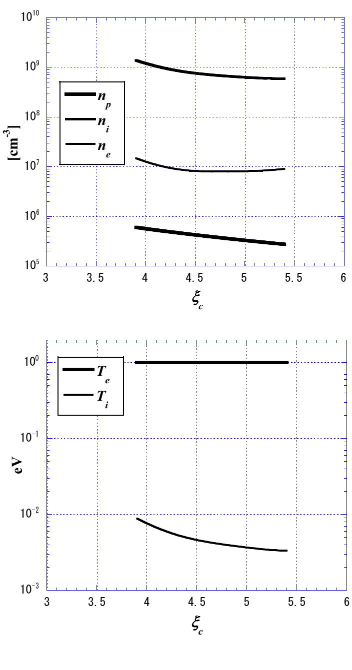

Fig. 4 Experimental conditions for $r_p = 10\mu$m. Densities (top) and temperatures (bottom).

6. Applicability to Fine Particle Plasmas

In experiments on the ground, the ion flow in the sheath gives the anisotropy of interaction between fine particles which is neglected in the above model. Many properties of fine particle plasmas, however, can be described within the isotropic interaction and we assume our system can be regarded as isotropic at least in the first approximation. Important aspects of fine particle plasmas which are not included in our model are; (a) anisotropy of interaction, (b) possible existence of attractive interactions between particles[6], (c) nonlinear screening and deviation from Yukawa interaction, and (d) thermodynamic openness.

As for (a), we assume that we have a bulk, approximately isotropic three-dimensional fine particle plasmas which may be realized under microgravity or methods to effectively cancel the gravity. The possibility (b) has the effect to make the conditions for strength of coupling in favor of the instability. As for (c), collision experiments indicate that Yukawa repulsion works at least in some range of mutual distance[7]. Though the aspects (b), (c), and (d) may influence critical conditions quantitatively, we may expect our results may be applicable to fine particle plasmas semi-quantitatively.

7. 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 given corresponding experimental conditions. In order to observe phenomena near the critical point, it is necessary to have a bulk isotropic three-dimensional system of fine particle plasmas. Though it might be 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.

This work has been supported by the Grant-in-Aid for Scientific Research (C) No.19540521 of JSPS.

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