CORRELATIONS AND FORCES IN STRONGLY COUPLED DUSTY PLASMAS

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A systematic procedure based on the Born—Green—Yvon (BGY) hierarchy is developed to find the higher order correlation functions and the interaction potential of strongly coupled, charged dust particles in the liquid phase, in terms of their measurable structure factor. The possibility to describe the gas-liquid transition of dusty plasmas in the framework of percolation theory is discussed.

Introduction

Dusty plasmas can be very different physical systems in different parameter regimes. One basic parameter is the coupling constant Γ, the ratio of mean potential energy to kinetic energy. Although the potential of dust-dust interaction in dusty plasmas is still not known, it is believed that Γ can be larger than 1 in some cases, which corresponds to a strongly coupled system of dust particles: dust liquid and dust crystal. In the opposite case, $\Gamma \leq 1$, the system can be treated with the methods of kinetic theory [1—5], but these methods are not valid for strongly coupled systems.

Dusty plasmas in the strongly correlated but disordered phase ("liquid") can provide a test for theories of simple liquids, due to the unique possibility to follow the kinetic processes and obtain accurate measurements of their structure.

All current theories of simple liquids start from the assumption of a pair potential of interaction and develop approximations for the two- and three-body correlation functions, which can be used then to calculate the thermodynamic quantities [6]. The approximate models, like Percus—Yevick, Hypernetted Chain, Mean Spherical Model, have no sound theoretical basis, as is the case for the kinetic theory of gases, where a small parameter Γ allows the truncation of the hierarchy of kinetic equations, and their justification relies only on the comparison with experimental results.

In dusty plasmas, on the other hand, due to the absorption of electrons and ions on dust, the dust-dust interaction potential is not known and the possibility of a long-range attraction between dust particles of like charge has also been discussed. But dusty plasmas also offer the unique possibility to measure the pair correlation function (or the structure factor) of dust particles with great accuracy, contrary to ordinary or charged fluids where neutron or x-ray scattering techniques are required. These also have problems with measurements at long wavelengths, strictly related to the long-range behaviour of the interaction potential.

Therefore, in this work, the usual method of fluid theory will be turned upside down: it will be assumed that the dust-dust interaction potential is unknown, while the dust pair correlation function is known from measurements, and a theory is presented to find the interaction potential and higher order correlation functions from structure data. A test of the theory and of the truncation of the kinetic hierarchy will also be given, again in terms of structure data.

Finally the possibility to describe the gas-liquid transition of dust particles in the framework of percolation theory will be discussed.

1. Structure and Forces

Here, the static equilibrium properties of strongly coupled, charged dust particles are considered in the framework of the BGY hierarchy [7], assuming that plasma particles (electrons and ions) contribute to the screening of dust particles and to the overall neutrality in the form of a superposition of screening clouds around grains at the positions $\{\mathbf{R}\} = (\mathbf{R}_1, \ldots.., \mathbf{R}_N)$ in the form

$$\rho_p(\mathbf{r}, \{\mathbf{R}\}) = \sum_i \sigma(\mathbf{r} - \mathbf{R}_i), \tag{1}$$

where ρ_p is the plasma charge density. In this case, the total electrostatic potential can be written as

$$\Phi(\mathbf{r}, \{\mathbf{R}\}) = \sum_i u(\mathbf{r} - \mathbf{R}_i), \tag{2}$$

where $u(r)$ is the screened potential around one dust particle. This assumption could be valid when the intergrain separation is larger than the screening length,

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and it allows us to treat the dusty plasma as a one-component system of dressed dust particles. The n-particle correlation function $g^{(n)}$ can be defined in terms of the potential of mean force $W^{(n)}$ as

$$g^{(n)}(\mathbf{R}_1, ..., \mathbf{R}_n) \equiv e^{-\beta W^{(n)}(\mathbf{R}_1,.....,\mathbf{R}_n)}, \tag{3}$$

where $\beta^{-1} = k_B T$, the thermal energy of the system, and the potential of mean force is defined as

$$\frac{\partial}{\partial \mathbf{R}_1}\beta W^{(n)}(\mathbf{R}_1, ..., \mathbf{R}_n) =$$

$$= -\frac{\partial}{\partial \mathbf{R}_1}\ln \int ... \int e^{-\beta E(\{\mathbf{R}\}}d\mathbf{R}_{n+1}....d\mathbf{R}_N, \tag{4}$$

where $E(\{\mathbf{R}\})$ is the total interaction energy of the system of N particles at the positions $\mathbf{R}_1, ..., \mathbf{R}_N$.

It can be shown that an exact relation between the pair correlation function $g(r)$ ($r \equiv R_{12} = |\mathbf{R}_1 - \mathbf{R}_2|$) and the three-body correlation function $g^{(3)}$ can be established using the potential of mean force, in the form of the so-called force equation

$$\frac{\partial}{\partial \mathbf{R}_1}W(r) = \frac{\partial}{\partial \mathbf{R}_1}u(r)+$$

$$+n_d \int \frac{g^{(3)}(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3)}{g(r)}\frac{\partial}{\partial \mathbf{R}_1}u(R_{13})d\mathbf{R}_3, \tag{5}$$

where n_d is the dust density. Assumptions (1), (2) and definition (3) lead to the following general form for the pair correlation function:

$$g(r) = e^{-\beta u(r)}e^{-A(r)}. \tag{6}$$

Here, the function $A(r)$ can be calculated, in principle, if the local screening density $\sigma(r)$ and potential $u(r)$ are known. To the lowest order $A = 0$, it is

$$g_0(r) = e^{-\beta u(r)}, \tag{7}$$

$$W_0(r) = u(r), \tag{8}$$

$$W_0^{(3)}(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3) = u(R_{12}) + u(R_{13}) + u(R_{23}), \tag{9}$$

and the Kirkwood superposition form follows for the three-body correlation function as

$$g_0^{(3)}(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3) = g(R_{12})g(R_{13})g(R_{23}). \tag{10}$$

Below, we use the force equation (5) to find the next order corrections and the unknown interaction potential in terms of quantities related to the dust structure factor

$$S(k) = 1 + n_d \int (g(r) - 1)\, e^{i\mathbf{k}\cdot\mathbf{r}}d\mathbf{r}. \tag{11}$$

Since $S(k)$ is easily measurable in dusty plasmas, the present results allow us to find the interaction potential and higher order correlations beyond the zero-order approximation and therefore of greater validity in the strongly coupled regime. The detailed calculation can be found in [9], where this method was first proposed for liquid metals. First, we notice that, using the zero-order results (7)—(10) in the force equation considered now as an equation for the unknown potential $u(r)$ gives the first-order result

$$u_1(r) = -\frac{1}{\beta}\ln g(r) + \frac{1}{8\pi^3 n_d}\int (S(k) - 1)^2 e^{i\mathbf{k}\cdot\mathbf{r}}d\mathbf{k} \tag{12}$$

which expresses the potential entirely in terms of structure data. This first-order result coincides with the results from both the Percus—Yevick (PY) and hypernetted-chain (HNC) equations. To the next order, first the correction to the Kirkwood superposition approximation (10) is found as

$$g^{(3)}(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3) = g(R_{12})g(R_{13})g(R_{23})e^{f(R_{13},R_{23})}. \tag{13}$$

Introducing the notations

$$T(k) = S(k) - 1, \tag{14}$$

$$T(\mathbf{k}, \mathbf{k}') = (S(\mathbf{k}') - 1)\,(S(|\mathbf{k} + \mathbf{k}'|) - 1)\,, \tag{15}$$

the function f is given by

$$f(R, R') =$$

$$= \frac{1}{(8\pi^3 n_d)^2}\int T(k)T(k')T(\mathbf{k}, \mathbf{k}')e^{-i\mathbf{k}\cdot\mathbf{R}}e^{-i\mathbf{k}'\cdot\mathbf{R}'}d\mathbf{k}d\mathbf{k}', \tag{16}$$

being again entirely expressed in terms of the dust structure factor. Using this result in the force equation leads then to the second-order potential, beyond the PY and HNC approximations,

$$\beta u_2(r) = \beta u_1(r) -$$

$$-\frac{1}{(2\pi n_d)^3}\int T(k)T(k')F(\mathbf{k}, \mathbf{k}', \mathbf{r})d\mathbf{k}d\mathbf{k}', \tag{17}$$

where

$$F(\mathbf{k}, \mathbf{k}', \mathbf{r}) = T^2(|\mathbf{k} + \mathbf{k}'|)e^{i\mathbf{k}'\cdot\mathbf{r}} -$$

$$-T(|\mathbf{k} + \mathbf{k}'|)\int T(k'')T(|\mathbf{k} + \mathbf{k}' + \mathbf{k}''|)e^{i\mathbf{k}''\cdot\mathbf{r}}d\mathbf{k}''. \tag{18}$$

The iterative procedure can be carried out, in principle, and the question, as usual with strongly coupled systems, concerns the convergence: there is no small parameter that allows the truncation of the BGY hierarchy. But dusty plasmas can perhaps provide, for the first time, a check of second-order results. In fact, it is possible to express [8] the difference between $g^{(3)}$ and the superposition value (10) in terms of the isothermal pressure derivative of $S(k)$ (the isothermal compressibility in the long- wavelength limit): if this quantity can be measured in strongly coupled dusty plasmas, it will provide an essential test for the present first-order result (13) for the three-body correlations and, consequently, for the second-order result for the interaction potential. But it can also be shown that the quantity

$$H(k) = n_d^2 \int e^{-i\mathbf{k}\cdot\mathbf{r}} d\mathbf{r} \int H(r,s) d\mathbf{s}, \tag{19}$$

where

$$H(r,s) = g^{(3)}(r,s) - g_0^{(3)}(r,s), \tag{20}$$

can again be entirely expressed in terms of structure data [9], if the assumption $\exp(f) \simeq 1 + f$ is made in (13), as

$$H(k) = -\frac{T(k)}{8\pi^3 n_d} \int T^2(q) \left(T(|\mathbf{k}+\mathbf{q}|) + T(|\mathbf{k}-\mathbf{q}|)\right) d\mathbf{q} - \\ -\frac{1}{(8\pi^3 n_d)^2} \int T(q)T(q')T(|\mathbf{q}+\mathbf{q}'|) G(\mathbf{k},\mathbf{q},\mathbf{q}') d\mathbf{q} d\mathbf{q}', \tag{21}$$

where

$$G(\mathbf{k},\mathbf{q},\mathbf{q}') = \\ = \frac{1}{8\pi^3 n_d} \int T(q'')T(|\mathbf{q}+\mathbf{q}''|)T(|\mathbf{q}+\mathbf{q}'+\mathbf{q}''-\mathbf{k}|) d\mathbf{q}'' + \\ + T(q)\left(T(|\mathbf{q}-\mathbf{k}|) + T(|\mathbf{q}+\mathbf{q}'-\mathbf{k}|)\right) + T(|\mathbf{k}+\mathbf{q}'|) + \\ + T(|\mathbf{k}-\mathbf{q}-\mathbf{q}'|). \tag{22}$$

Thus, structure data can allow us to check difference (20) and provide a test for the present result (13) for the three-body correlations and therefore for the interaction potential. Most important, measurements at long wavelengths can be done in dusty plasmas and these can provide information on the long-range behavior of the dust-dust interaction. It has been postulated that a long-range attraction between negatively charged dust particles is possible due to the over-screening by ion fluxes to the dust surfaces [10]. The comparison of structure data with a solution of the Mean Spherical Model with an attractive tail also seems to suggest the presence of long-range attraction [11], but the result is still controversial due to the approximations in the models. The use of the present theory to extract the potential from structure data could help resolve this problem.

2. Percolation Theory

For a long time, attempts have been made to interpret the gas-liquid transition of simple liquids in terms of the formation of an “infinite” physical cluster of particles (percolation) at the transition point [12]. If some attraction between dust particles exists, it would then be possible to apply percolation theory to the dust liquid transition. Since, in dusty plasmas, it could be possible to see, measure, and follow the development of physical clusters, these systems could provide a new unique test for the theories of percolation.

Percolation theory was originally developed for lattice systems, such as the Bethe lattice and the ferromagnetic Ising model, and extended to fluids by Hill [13] who introduced the idea of physical clusters of interacting particles: n particles belong to the same cluster if they are pairwise bound in the phase space(the negative total energy). Series expansions for the average number and the size of physical clusters of particles, in the framework of Mayer's diagrammatic expansions, were given in [14]. The theory of the pair connectedness $P(r_i, r_j)$ was developed in [15]. This is defined such that

$$n^2 P(r_i, r_j) d\mathbf{r}_i d\mathbf{r}_j$$

is the probability that particles i and j are in volume elements $d\mathbf{r}_i$ and $d\mathbf{r}_j$ and are physically bound, i.e. belong to the same physical cluster, in a system of N particles in volume V with density $n = N/V$ and potential energy given by Eq.(2). The activity and density expansions, a relation of the Ornstein—Zernicke type, and the Percus—Yevick approximation have been given for the pair connectedness [15] in analogy with the results for the pair correlation function.

In analogy with the theory of simple liquids, where the long-wavelength limit of the structure factor gives the system's isothermal compressibility, it was shown [15] that the long-wavelength limit of the Fourier transform of the pair connectedness is related to the critical percolation density $n_p(T)$: the density (a function of temperature T), where an infinite physical cluster of particles appears in the system, is possibly related to the point of the gas-liquid transition $n_g(T)$.

The formalism of the pair connectedness allows a description of both percolation and condensation in the pressure-density plane to find the relationship between $n_p(T)$ and $n_g(T)$: on a given isotherm, is $n_p(T)$ lower or

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higher than $n_g(T)$? In other words, does the formation of an infinite physical cluster of particles (percolation) occur before, after, or simultaneously with condensation?

In the framework of the theory developed in [15], it is possible to express the mean cluster number and size as functions of the thermodynamic variables of the system (assuming the interaction potential is known) and to study the behavior and the distribution of physical clusters and the occurrence of percolation. The results depend, of course, on the level of approximation and, as stated before, there is no check of their validity. In ordinary fluids, clusters cannot be "seen" to test the theory. Dusty plasmas offer the possibility to actually see the clusters formed in the system and to follow their dynamics and development as the system approaches the "liquid" phase. As for the theory in the previous section, it would then be possible to turn the method around: consider the size and distribution of clusters as known quantities and test the theories and approximations with results from measurements.

Conclusions

Dusty plasmas in the strongly coupled disordered phase ("liquid") can provide a test for the established theories of simple liquids and charged fluids and for the theory of percolation.

The unique possibility to follow the kinetic processes in these systems and to accurately measure their structure as well as the size and distribution of physical clusters of dust particles, may allow one to find the dust-dust interaction potential and the three-particle distribution function from structure data and to test the truncation of the kinetic hierarchy and the validity of the Kirkwood superposition approximation, which is the basis for most of the current models of liquids.

If the dust-dust interaction has an attractive tail, the results of percolation theory could be applied to dusty plasmas as they approach the condensation point, and these results could be tested against the measurable distribution of physical clusters of dust particles, providing tests for the theoretical models.

It should be pointed out that the theories discussed in the present work are referred to infinite free systems. Dusty plasmas are often electrically confined in laboratory experiments and the present results should be used with data from microgravity experiments or be extended to include the effects of external fields.

1. *Tsytovich V.N., de Angelis U.* // Phys. Plasmas. — 1999. — **6**. — P. 1093—1106.
2. *Tsytovich V.N., de Angelis U.* // Ibid. — 2000. — **7**. — P. 554—563.
3. *Tsytovich V.N., de Angelis U.* // Ibid. — 2001. — **8**. — P. 1141—1153.
4. *Tsytovich V.N., de Angelis U.* // Ibid. — 2002. — **9**. — P. 2497—2506.
5. *Tsytovich V.N., de Angelis U.* // Ibid. — 2004. — **11**. — P. 496—506.
6. *Cole G.H.A.* An Introduction to the Statistical Theory of Classical, Simple Dense Fluids. — Oxford: Pergamon Press, 1967; *Hansen J.P., McDonald I.R.* Theory of Simple Liquids. — London: Academic Press, 1976.
7. *Hill T.L.* Statistical Mechanics. — New York: McGraw-Hill, 1956.
8. *Egelstaff P.A., Page D.I., Heard C.* // J. Phys. C. — 1971. — **4**. — P. 1453—1461.
9. *de Angelis U., March N.H.* // Phys. Chem. Liquids. — 1977. — **6**. — P. 225—242.
10. *Tsytovich V.N., Morfill G.E., Thomas H.* // Fiz. Plasmy. — 2002. — **28**. — P. 675—707 [Engl. trans.: Phys. Repts. — 2002. — **28**. — P. 623—651].
11. *de Angelis U., Forlani A., Masiello G.* // Phys. Plasmas. — 2000. — **7**. — P. 3198—3209.
12. *Sator N.* // Phys. Repts. — 2003. — **376**. — P. 1—39.
13. *Hill T.L.* // J. Chem. Phys. — 1955. — **23**.— P. 617—622.
14. *Coniglio A., de Angelis U., Forlani A., Lauro G.* // J. Phys.A. — 1977. — **10**. — P. 219—228.
15. *Coniglio A., de Angelis U., Forlani A.* // J. Phys.A. — 1977. — **10**. — P. 1123—1139.

КОРЕЛЯЦІЇ І СИЛИ У СИЛЬНОВЗАЄМОДІЮЧІЙ ЗАПОРОШЕНІЙ ПЛАЗМІ

У. ді Ангеліс

Р е з ю м е

Систематичну процедуру, основану на BGY-ієрархії, застосовано для знаходження кореляційних функцій вищого порядку і потенціалу взаємодії сильнозв'язаних заряджених порошинок у рідкій фазі у термінах вимірюваного структурного фактора. Обговорюється можливість описати перехід газ—рідина запорошеної плазми в рамках теорії перколяції.