Three-Particle Correlations in Nonideal Dusty Plasma

O. S. Vaulina, O. F. Petrov, V. E. Fortov, A. V. Chernyshev, A. V. Gavrikov, and O. A. Shakhova
Institute of High Energy Density, Russian Academy of Sciences, 13/19 Izhorskaya, Moscow, 125412 Russia
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Results are given of experimental investigation of three-particle correlations for liquid plasma-dust structures formed in the electrode layer of a high-frequency capacitive discharge. The obtained three-particle correlation functions for experimental and numerical data are analyzed and compared with the superposition approximation. The forming of clusters of macroparticles in plasma-dust systems being analyzed is revealed.

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The dusty plasma of gas discharges is a partly ionized gas which, in the majority of cases, contains negatively charged dust particles of micron size. Gas-discharge chambers are extensively used for the investigation of the properties of dusty plasma owing to the presence of electric fields capable of containing such macroparticles both in the earth gravity field and in the radial (perpendicular to the force of gravity) direction. Micron-sized dust particles in a gas-discharge plasma assume a significant negative charge $[(\sim 10^3\text{–}10^5 e)]$ and may form quasistationary plasma-dust structures similar to a liquid or a solid [1–5].

The equilibrium properties of a liquid are fully described by a set of probability density functions $g_s(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_s)$ of location of particles at points $\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_s$. In the case of isotropic pair interaction, the physical properties of a liquid (such as pressure, energy density, etc.) are fully defined by the binary correlation function $g(r) = g_2(|\mathbf{r}_1 - \mathbf{r}_2|)$ [6–9], which in turn depends on the type of potential of interaction between particles and on its temperature. However, even in the approximation of pair interaction, higher-order correlation functions ($s > 2$) are of considerable interest. For example, information about the three-particle correlation function $g_3(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$ is of importance in calculating the physical characteristics of the medium (such as entropy, coefficients of thermal expansion, etc.) that depend on the derivatives of $g(\mathrm{r})$ with respect to temperature $\partial g(r)/\partial T$ or density $\partial g(r)/\partial \rho$. The function $g_3(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$ defines the probability of simultaneous detection of three particles in the vicinity of points $\mathrm{r}_1, \mathrm{r}_2, \mathrm{r}_3$. Unlike the binary function $g(\mathrm{r})$, the $g_3(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$ depends on three space coordinates and, accordingly, enables one to obtain additional information about the structure of particles. The superposition approach

$$g_3(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) \approx g_3^{\mathrm{sp}}(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) = g(\mathbf{r}_1 - \mathbf{r}_2) g(\mathbf{r}_2 - \mathbf{r}_3) g(\mathbf{r}_3 - \mathbf{r}_1) \quad (1)$$

is most frequently employed to approximate the three-particle correlation function. This approach is based on the disregard of terms of the form $U(\mathbf{r}_1 - \mathbf{r}_2, \mathbf{r}_3 - \mathbf{r}_1)$ in the system Hamiltonian, which are not reduced to pair interactions. Relation (1) is often used in calculating integral equations in the kinetics of interacting particles, as well as in recovering pair interparticle potentials, using the hyperchain approach [8–12]. Nevertheless, the available results of numerical studies, performed for the hard-sphere interaction or for the Lennard-Jones potentials, demonstrate the inadequacy of the approximation (1) even for low particle density [10–12]. The experimental verification of this approximation for real liquids is made difficult by the fact that, unlike the binary correlation function whose determination may be based on the inversion of the structure factor $S(k)$ measured by standard spectroscopic methods [13], no direct determination of the three-particle correlation function is possible without information on the particle's coordinates. Indirect diagnostic methods are used to analyze three-particle correlation in real liquids, such as the measurements of $S(k)$ at several pressures with constant temperature to be followed by the determination of $\partial g(r)/\partial \rho$, which in turn contains information about $g_3(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$ [9,10]. Extracting such information requires additional data on the isothermal compressibility of the medium under investigation.

In contrast to real liquids, the laboratory dusty plasma is a good experimental model for studying the properties of nonideal systems because, owing to their size, dust particles may be video filmed, which significantly simplifies the use of direct diagnostic methods. Experimental studies of dusty plasma may play an important part in verifying existing, and developing new, phenomenological models for strongly coupled liquids. Such models are of great significance because, owing to strong interparticle interaction, the theory of liquids lacks a small parameter that could be used for an analytical description of its state and thermodynamic characteristics, as is possible in the case of gases.

It is customary to assume that dust particles in a plasma interact with one another through the intermediary of screened Coulomb potential [14,15], $\phi = eZ_p \exp(-r/\lambda)/r$, where r is the distance, λ is the screening radius, and Z_p is the dust charge. The dynamics of particles interacting with a screened potential (Yukawa systems) were investigated in studies [16–18]. The results of numerical simulation demonstrate that the viscosity of buffer gas has almost no effect on the correlation of particles and that the effective coupling parameter, $\Gamma^* = \Gamma(1 + k + k^2/2)\exp(-k)$, may be used to analyze their phase state [here $\Gamma = (Z_p e)^2/(T_p r_p)$, $r_p = n_p^{-1/3}$ is the mean interparticle spacing, T_p and n_p denote the kinetic temperature and concentration of dust, respectively, and$k = r_p/\lambda$ is the screening parameter]. In the case of $k < 6$, the value of Γ^* fully defines the form of the binary correlation function $g(r)$ for liquid systems. These results are in good agreement with the experimental studies of the transport characteristics of dust structures in the plasma of gas discharges of different types [19–21].

This Letter contains the results of an experimental investigation of three-particle correlation for liquid plasma-dust structures formed in the electrode layer of a radio-frequency (rf) capacitive discharge. A schematic of the experimental facility is shown in Fig. 1. The experiments were performed in argon at pressure P of 2 to 10 Pa with discharge power W of 2 to 10 W. The dust component was provided by particles of formaldehyde melamine of radius $a_p \approx 1.7$ μm and density $\rho_p \approx 1.5$ g cm^{-3}. In the experimental conditions the macroparticles formed four to eight dust layers. In all cases being analyzed, the observed structures were systems of the liquid-type with the mean interparticle spacing r_p varied from 260 to 350 μm. The diagnostics involved the illumination of the single layer of dust cloud by a laser sheet (thick, $\sim$250 μm), after which this monolayer was video filmed using a CCD camera (frame frequency, 25 s^{-1}). Fragments of video image of macroparticles registered are shown in Fig. 2.

The video record was treated using special computer codes. An additional brightness analysis of particle's video images was performed for their registration in a single dust layer. To correct the fluctuations of separate dust particles out of the layer [22], pair correlation functions $g(r)$ and triplet correlation functions $g_3(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3)$ averaged over a period of 2 to 2.5 s under constant experimental conditions were obtained. The pair functions $g(r/r_p)$ are given in Fig. 3 for different discharge parameters. The cross sections of the triplet functions $g_3(r_{12}, r_{23}, r_{31})$ ($r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$) for a fixed value of r_{12} equal to the most probable interparticle spacing $r_p^{\max}(r_{12} = r_p^{\max})$ determined by the position of $g(r)$ maximum are given in Fig. 4, together with the results of calculation of $g_3^{\rm sp}(r_{12}, r_{23}, r_{31})$ within the approach (1). In order to represent these functions [$g_3(r_{12}, r_{23}, r_{31})$ and $g_3^{\rm sp}(r_{12}, r_{23}, r_{31})$] in a "two-dimensional" form convenient for comparison, they were normalized to the maximum of $g_3(r_{12}, r_{23}, r_{31})$: black color corresponds to unity, and white color corresponds to $g_3 = g_3^{\rm sp} = 0$. The deviation of the measured function $g_3(r_{12}, r_{23}, r_{31})$ from $g_3^{\rm sp}(r_{12}, r_{23}, r_{31})$ is given in the caption of Fig. 4 and was calculated proceeding from the relative mean-square error,

$$\delta = \frac{1}{N^{1/2}}\left[\sum_{i=1}^{N}\{g_3(r_{12}, r_{2i}, r_{i1}) - g_3^{\rm sp}(r_{12}, r_{2i}, r_{i1})/g_3(r_{12}, r_{2i}, r_{i1})\}^2\right]^{1/2}, \quad (2)$$

where N is the total number of elements of space $d\mathbf{r}_i$ in the vicinity of the point with coordinate $\mathbf{r}_i$. The magnitude of δ under the experimental conditions varied from $\sim$30% to $\sim$60%. It is to be reminded that the correlation functions were calculated only for the particles of a single dust layer which fitted into the plane of the laser sheet.

Visual comparison of the results reveals that the recorded structures exhibit the forming of a close-range orientational order of dust particles, which is reflected in the emergence of maxima of $g_3(r_{12}, r_{23}, r_{31})$ in nodes of

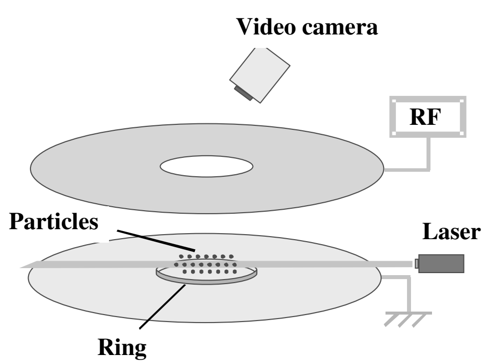

FIG. 1. Schematic of the facility for experiments in a capacitive rf discharge.

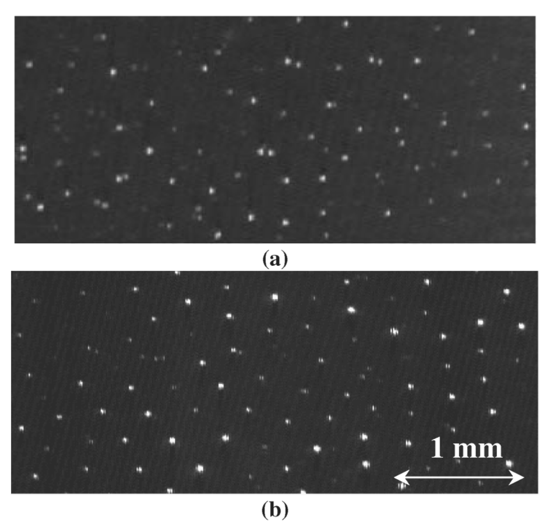

FIG. 2. Video image of dust cloud particles in the electrode layer of the discharge for different experiments: (a) $P = 5$ Pa, $W = 9$ W; (b) $P = 7$ Pa, $W = 10$ W.

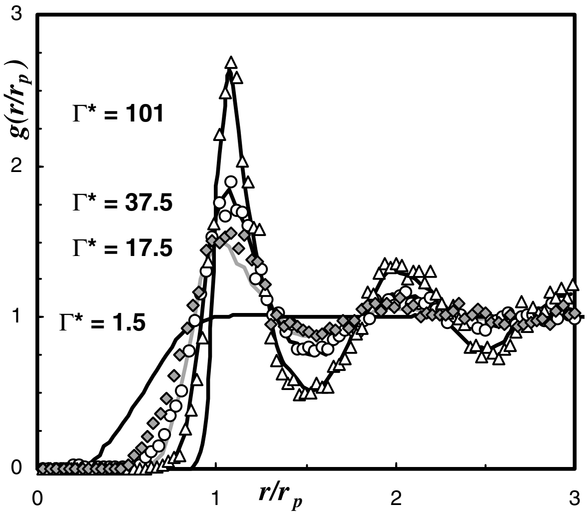

FIG. 3. Pair correlation functions $g(r/r_p)$ measured in the experiments: (◆) $P = 5$ Pa, $W = 9$ W; (○) $P = 3$ Pa, $W = 2$ W; (△) $P = 7$ Pa, $W = 10$ W; and those obtained by numerical simulation for different values of Γ^* (solid lines) indicated in the figure.

hexagonal cluster, s shown by dashed lines in Figs. 4(a) and 4(b). As the maximum of the pair correlation function increases (see Fig. 3), the magnitude of these maxima located at distances r close to $r_p^{\max}$ grows, and new maxima arise at distances $r \approx 2r_p^{\max}$. This effect does not show up when the approximation $g_3^{\rm sp}(r_{12}, r_{23}, r_{31})$ is analyzed. Some experimental conditions [see Fig. 4(c)] were characterized by the simultaneous presence in dust systems of both hexagonal clusters and cluster structures similar to the arrangement of particles on the faces of cubic lattices of different types. These elements may also be observed in experimental video recording [see Fig. 2(a)].

Numerical simulation was performed in order to compare the experimental results with the correlation of particles in Yukawa systems. The calculations were performed for a three-dimensional system (at $k < 6$) using Langevin's method of molecular dynamics with periodic boundary conditions for 512 independent macroparticles and the cutoff of pair potential equal to $7r_p$. In order to simulate the experimental conditions of observation of dust particles in the laser sheet plane, the computational cells were divided into layers of thickness $\sim r_p/2$, and the correlation of macroparticles was investigated in each layer separately. Nevertheless, under experimental conditions the intergrain potential may have non-Yukawa form, or it can be essentially asymmetric (in the direction orthogonal to the dust layer plane) due to ion wake-field effects [23]. This can lead (as one will show below) to a difference between numerical and experimental results.

Three-particle correlation functions and their superposition approaches are illustrated in Fig. 5 for three different Γ^* at $r_{12} = r_p^{\max}$. The error δ for the presented numerical data was higher than the value of δ recorded in laboratory experiments. The obtained pair correlation functions are given in Fig. 3 for different coupling parameters Γ^*. One can readily see that the experimental results well fit systems with $\Gamma^* \sim 100$, 37.5, and 17.5. In the latter case, the differences between calculated and measured $g(r)$ are most pronounced because the experimental curve exhibits a broader first maximum associated with the inhomogeneity of the analyzed structure due to a formation of dust clusters of different types.

Analysis of the calculation results reveals that the pronounced maxima of $g_3(r_{12}, r_{23}, r_{31})$ in the system being simulated arise with the emergence of such maxima for a pair correlation function, i.e., at $\Gamma^* > 5$ [16,17,24]. In so doing, the analyzed cross section of three-particle function ($r_{12} = r_p^{\max}$) has a form similar to that given in

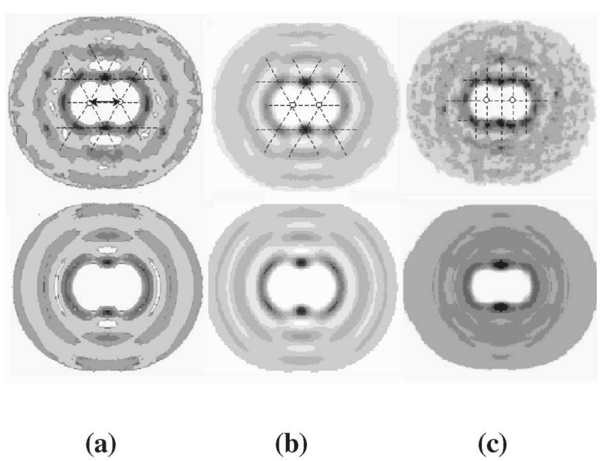

FIG. 4. Three-particle correlation functions g_3 (top row) and superposition approximation $g_3^{\rm sp}$ (bottom row) measured for different experiments: (a) $P = 7$ Pa, $W = 10$ W, $\delta = 0.61$; (b) $P = 3$ Pa, $W = 2$ W, $\delta = 0.28$; (c) $P = 5$ Pa, $W = 9$ W, $\delta = 0.3$.

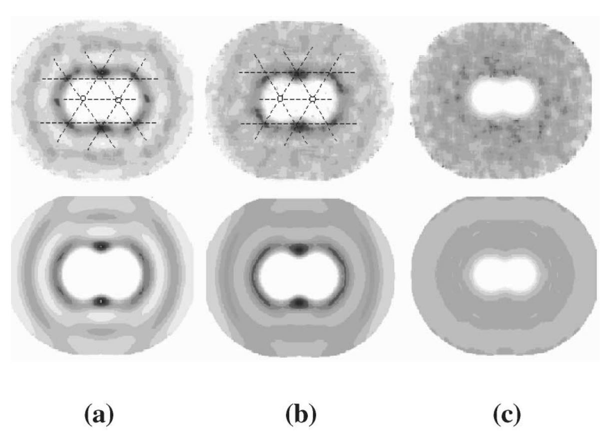

FIG. 5. Three-particle correlation functions g_3 (top row) and superposition approximation $g_3^{\rm sp}$ (bottom row) obtained by numerical simulation for different values of the parameter Γ^*: (a) $\Gamma^* = 37.5$, $\delta = 0.62$; (b) $\Gamma^* = 17.5$, $\delta = 0.61$; (c) $\Gamma^* = 1.5$, $\delta = 0.14$.

Fig. 5(b). As Γ^* increases from 5.5 to 22, the maxima of $g_3(r_{12}, r_{23}, r_{31})$ increase, and, at $\Gamma^* > 25$, the emergence of regular clusters of macroparticles is registered in the system [see Fig. 5(a)]. The shape of such clusters is close to the hexagonal shape observed in laboratory experiments [see Figs. 4(a) and 4(b)]. Nevertheless, in some experimental situation the shape of observed dust clusters may be considerably different from hexagonal shape [see Fig. 4(c)]. This may be associated with differences of the experimental conditions from the simulated problem, for example, with non-Yukawa-type of experimental potential. In spite of this, for homogeneous systems, good agreement is observed between three-particle correlation functions determined for numerical and experimental data, at least in the cases where similar agreement is observed between the shapes of their binary correlation functions [see Figs. 3, 4(b), and 5(a)].

The emergence of regular dust clusters in both simulated and experimental systems may be indicative of the fact that, as the coupling parameter increases, a short-range orientational order starts forming in the arrangement of macroparticles at distances exceeding mean interparticle spacing. Revealing the nature of the observed effects requires additional investigations to determine the characteristic times of orientational processes (mean times of existence of a certain orientation in the arrangement of particles) and their dependence on the coupling parameter.

As Γ^* increases to exceed 40–50, the form of the three-particle correlation function for numerical experiment comes to depend on the orientation of the plane of the "laser sheet," i.e., on the orientation of plane parallel layers into which the computational volume is divided. This latter case may be attributed to the fact that, as the parameter Γ^* increases, the dynamics of the simulated system become similar to those of a solid and may be treated within the "jump's" theory developed for molecular liquids [6]. The main point of this theory is that the molecule is in an equilibrium ("settled") state for a period of time required for imparting an activation energy sufficient for breaking potential bonds with neighboring molecules and for the transition into the environment of other molecules (to a new settled state). The agreement of the dynamics of Yukawa systems with this theory is supported by the numerical investigations of [17,18].

In conclusion, we will emphasize once again that it is for the first time ever that results of an experimental investigation of three-particle correlation for dust particles in a plasma are published. Analysis of the experimental results has revealed that the difference of superposition approximation from the recorded three-particle correlation function ranges from 30% to 60% for the analyzed cross sections of $g_3(r_{12}, r_{23}, r_{31})$ at $r_{12} = r_p^{\max}$. The forming of regular clusters of macroparticles was observed both in experimental plasma-dust systems and in simulated structures. The numerical calculations have demonstrated that the forming of such clusters in the Yukawa systems is observed for $\Gamma^* > 25$, which well agrees with the results of [18]. Note further that experimental investigations of three-particle correlation enable one to obtain additional information about the physical properties of plasma-dust systems and may be used for structure analysis of complex plasma.

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