Phase transitions and transformation of dust structures in neon dc discharge at cryogenic temperature

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Abstract. The transformation of dust structures under changes of discharge current in neon dc discharge at a pressure of 20 Pa and the discharge tube temperature of 77 K has been studied. A nonmonotonic dependence of radial and axial sizes of the dust formations on the discharge current have been detected and analyzed. It has been found that dust clouds consisted of a mixture of dust particles and clusters formed from dust particles. The first and second-order phase transitions in dust structures and the mesomorphic state of dusty plasma have been found. The transformation of the dust structure to spherical form has been detected.

1. Introduction

Dusty plasma is an open nonequilibrium dissipative nonideal system with strong Coulomb interaction. Therefore, dust structures can be used to simulate nonideal systems in microworld, including those forming at low and cryogenic gas temperatures. The dust clouds in plasma can exist in forms analogous to the thermodynamic states of matter [1], and phase transitions between these states can be observed. The structure of dust particles can ‘melt’ and ‘evaporate’ like a matter. In this case, the long-range order between the dust particles is violated, while the short-range order is maintained. This allows one to describe the dusty plasma in the approximation of the thermodynamic system through the parameter of non-ideality (the kinetic temperature of particles) and to represent its properties in the hydrodynamic approximation through the transport characteristics of a continuous medium [2].

The interest in the studies of characteristics of gas discharge plasma with dust particles cooled to cryogenic temperature is stimulated by the possibility of using such plasmas in novel plasma-chemical technologies. These studies are relevant for the development of the theory of nucleation, growth and agglomeration of nanostructures and coagulation of micro- and nanoparticles in plasma [3-5], and may promote the related sciences such as low-temperature and quantum chemistry [6, 7].

At low and cryogenic gas temperatures in some gases, the structures consisting of dust clusters can be formed [5, 8-11], and the Bose condensation can occur [12]. The process of cluster formation at cryogenic cooling was accompanied by a decrease in the distances between the dust particles [8-11]. As well as individual dust particles, these clusters can form the lattice sites similar to atoms in a solid, and also exist in a ‘liquid’ and ‘gaseous’ states. The discharge current is known to exert the greatest influence on the topological parameters of the dust structure in dc discharge. For instance, the influence of the current on the radial size of dust structure was observed in the discharge plasma of a number of gases with various dust particles [13-15]. In this work the influence of the discharge current on the shape, size, composition of complex dust structures consisting of dust clusters, and their phase state has been studied at cryogenic gas temperature.
2. Experimental

The parameters of dust structures formed by spherical plastic particles with a diameter of 4.14 μm, which were injected into dc discharge in neon at a pressure of 20 Pa, have been experimentally studied in a glass discharge tube (1) with an inner diameter of 16.5 mm and a length of 20 cm (figure 1). The discharge current $I$ was varied from 0.6 to 3.2 mA. To measure the voltage drop $\Delta U_c$ in the positive column of the discharge and to provide confinement of dust particles (2) at the measurement region, two narrow ring electrodes (3) spaced apart, were located in the discharge tube near the cathode (4). The discharge tube was placed in an optical cryostat (5), where it was cooled to the boiling point of liquid nitrogen $T=77$ K. Also at different values of the discharge current, the total voltage of the discharge $U$, and the sizes of the dust structure - the radial $D$ (diameter) and the axial $L$ (length) were measured. Using the electrical measurement data, the values of the linear power input $Q=EI$ and the longitudinal electric field $E=\Delta U_c/l$ in the discharge were calculated. Length $l$ is the distance between ring electrodes. The gas temperature in the discharge was assumed to be equal to the temperature of the discharge tube near the investigated sector containing dust particles. Images of dust structures (6) were recorded with a microscope (7) in the axial section in the reflected light of a plane laser beam (8). According to the obtained images, the composition, shape, degree of order and dynamic stability of the components of dust structures were analyzed. The experimental setup is described in detail elsewhere [9, 10, 16].

3. Results and discussion

At room temperature of the discharge (about 295 K), an increase in the discharge current leads, as a rule, to an increase in the radial $D$ and a decrease in the axial $L$ size of the dust structures [11, 14]. In neon dc discharge at room temperature, the dust structures were formed by individual dust particles (figure 2 (a)). With decreasing gas temperature, the individual dust particles formed clusters (figure 2 (b, c)). At a lower gas temperature, the clusters become ordered in axial direction (figure 2 (c)). When the clusters were formed, typical features of a first-order phase transition, at which the particle density increased sharply and continued to increase with decreasing gas temperature were observed. The process of formation of clusters can be considered as ‘condensation’ and ‘deposition’ of dust particles [11].

Figure 1. Experimental scheme:
1- glass discharge tube,
2- confinement of dust particles,
3- measuring ring electrodes,
4- cathode, 5- optical cryostat,
6- dust structure image,
7- microscope, 8- laser beam,
9- laser, 10- monitor and video-recorder, 11- video-camera,
12- anode, 13- voltmeter,
14- current meter, 15- high voltage power supply.
Figure 2. (a-c) Fragments of dust structure at decreasing gas temperature.

At $T=77$ K the dust structures can be multicomponent [10, 11]. Multicomponent system is represented by a mixture of dust particles and clusters formed by dust particles. The composition of the dust system varies with gas pressure and discharge current. One-component dust systems consist of simple clusters (figure 2 (c)). Simple clusters are threadlike clusters consisting of several dust particles located along the discharge axis. Threadlike clusters form homogeneous structures in the ‘liquid’ state, which were observed at higher neon pressures $P \geq 120$ Pa [10, 11]. In our case, complex clusters consisting of threadlike clusters, formed with decreasing gas pressure. In contrast to the dust structures at room temperature, upon varying the discharge current, the nonmonotonous dependencies of the radial $D$ and axial $L$ sizes of the dust structure on the discharge current $I$ (figure 3, circles) have been observed in cryogenic multicomponent systems.

Figure 3. Dependencies of the radial $D$ (black circles) and axial $L$ (red circles) sizes of dust cloud, the longitudinal electric field over the dust cloud diameter $E/D$ (blue triangles), and the total discharge voltage $U$ (green squares) on the discharge current $I$.

In figure 3, one can distinguish three characteristic regions (I - III), divided with dashed lines. Regions II and III are analogous in the dependences on the current of the shape of the dust structures formed by individual dust particles observed at room gas temperature. The change in the slope of the $D(I)$ curve in region III is caused by the change in the discharge mode. Here, a decrease in the discharge voltage $U$ (squares, figure 3) has been observed; it led to a decrease in the electric field $E$. Regions II and III are characterized by constant ratio of the longitudinal electric field to the radial size of the dust structure, $E/D=\text{const}$ (triangles, figure 3). This result indicates the applicability of one-dimensional model of dusty plasma [17-19] at cryogenic temperatures.
Figure 4. (a) Dependencies of the dust structure volume $V$ (black circles), the longitudinal electric field $E$ (pink squares) and the linear power input $Q$ (blue triangles) on the discharge current $I$. (b-e) Images of dust structures at phase inversion points.

Figure 4 represents the dependencies of the dust structure volume $V$, linear power input $Q$ and longitudinal electric field $E$ on the discharge current $I$. The images of dust structures (figure 4 (b-e)) correspond to the regions I and II in figure 3. These images correspond to characteristic points of phase inversion. From the analysis of the dust structure images and from $V(I)$ dependence, one can conclude that there are the evidences of the first- and second-order phase transitions in dust structures, which follow the changes of the discharge current. At $I = 0.631$ mA (figure 4 (b) (region Ia)), the dust structure is in the ‘crystalline’ state. The dust ‘crystal’ is formed by cluster chains, which consist of multi-dimensional clusters. An increase of the current to 0.633 mA (figure 4 (c) (region Ib)) is accompanied by a decrease in the symmetry (up to its disappearance) of the dust structure. Thus, the evidence of a second-order phase transition has been observed. At a higher current (figure 4 (d) (region Ib)), a multicomponent mixture consisting of cluster chains and simple clusters was observed.
inside the dust structure; this indicated the ‘mesomorphic’ state of dust system. This is a state where reconstructive transition of allotropic structures from a crystal to an alloy of a crystal in an isotropic liquid and vice versa can occur. At $I = 0.691$ mA (figure 4 (e) (region II)), there was an abrupt jump in the dust structure volume, i.e. the density of the dust cloud decreased sharply, which indicated that it was a first-order phase transition. Here, the ‘melting’ of complex dust clusters and partial ‘melting’ of simple dust clusters were observed. In region II (figure 3 and figure 4), the dust system exists as a mixture consisting of individual dust particles and simple clusters. The unstable dynamic state of the dust particles resembled a ‘boiling liquid’. The development of oscillations and rotations of individual dust particles was observed. The intensity of dust particle motion increased with decreasing pressure. As the discharge current increased (region II), the concentration of individual dust particles and the intensity of their motion decreased. The dust system in region II exists in the ‘liquid’ state. In this region, the change in the dust structure size with the change of the discharge current occurs analogously to that observed in dust structures at room temperature.

**Figure 5.** (a) Dependencies of the relative size of the dust structure $L/D$ (green circles), the longitudinal electric field $E$ (pink squares) and the linear power input $Q$ (blue triangles) on the discharge current $I$. (b-e) Images of dust structures at different values of discharge current.

This behavior is illustrated by the dependence of $L/D$ on $I$ (green circles) in figure 5 (a). At a current of about 1.15 mA $L/D=1$; in this case, the dust structure takes the spherical shape (figure 5 (b)). The spherical dust clouds were observed in discharges of various types and their formation was associated with various conditions and gas discharge parameters (not only gas temperature). The spherical dust clouds were first observed in neon dc discharge at room temperature of the gas [20]. In dc discharge in air, the spherical dust structures formed under the influence of an external magnetic field [21] (figure 6 (a)). The spherical dust structures also formed in a radio-frequency discharge, when an active and grounded electrodes were shifted in the vertical discharge tube (figure 6 (b)). When a spherical cloud formed, the distances between dust particles decreased from 170 μm to 80-90 μm. At a decrease of gas temperature, an increase in number density of charged dust particles occurs as a result of a decrease in the Debye radius [22].

At cryogenic temperatures, the spherical dust structures were observed for the first time in a glow discharge of direct current in helium at $T = 4.2$ K [4]. In [23], the transition to the spherical shape of a dust cloud was obtained as a result of a decrease in the temperature of the helium plasma from 175 to 135 K. A sphere-like structure at $T=77$ K was obtained as a result of decrease of gas pressure and
discharge current in helium [24]. The spherical dust structures can simulate the state of a liquid from the point of view of compliance with the Eötvös rule for the magnitude of surface tension force, which increases with increasing the number density and decreasing the temperature of the liquid. The presence of ‘surface tension’ in the dust structure is indicated by the dynamics of the drop-like shape of the dust structure [25] and the dynamics of its separation into two spherical parts under the influence of an external temperature field [13, 26]. For the dust structure in ‘liquid’ state, the surface tension can be represented through the binding energy of dust particles [13], which is comparable to the value of the binding energy of a dust molecule [27].

The spatially ordered planar structures of spherical shape formed from charged water droplets above its surface [28]. The round form of such structures, in the opinion of the authors of [29], indicated the presence of a surface tension. Similar structures can exist in atmospheric clouds and fogs [29].

![Images of spherical dust structures (axial section) at room temperature: (a) in a dc discharge with a radial magnetic field [20] and (b) in a radio-frequency discharge.](image)

**Figure 6.** Images of spherical dust structures (axial section) at room temperature: (a) in a dc discharge with a radial magnetic field [20] and (b) in a radio-frequency discharge.

4. Conclusions
The impact of the discharge current on the size, shape and composition of complex Coulomb dust structures consisting of dust clusters at cryogenic temperature has been studied. The first and second-order phase transitions have been found. It has been found that with increasing current, the complex clusters melted, giving a mixture of components consisting of simple clusters, complex clusters, and individual dust particles. In the ‘liquid’ state of dusty plasma, the formation of a spherical dust structure has been observed.

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