The role of molecular ions in cryogenic dusty plasmas of dc discharge in neon

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Abstract. The diffusion-drift model of positive column of glow discharge in neon have been used to study the effect of molecular ions in the interaction of plasma with dust particles of micron size. Simulations have been carried out at a pressure of 20 – 100 Pa and gas temperature of 77 K. The electric field of the discharge, the concentrations of discharge components, including metastable atoms and molecular ions, and the charge of dust particles have been simulated.

1. Introduction

Plasma of electric discharges of noble gases is widely used in engineering at low temperatures [1, 2]. Plasma with dust particles (dusty plasmas) is used in various technical devices for plasma coating, surface modification, in plasma-chemical reactors with rf or dc discharges for production of fine powders and novel materials [3-5] and in other processes of plasma-cryogenic synthesis [6]. The dusty plasma simulates the properties of non-ideal systems observed in the micro-world in a wide range of gas temperature including cryogenic [7-10].

The composition of the plasma (the partial concentrations of atomic ions, molecular ions and metastable atoms) is determined by the parameters of the discharge and the gas density. Usually, a large number of metastable atoms (A*) is contained in the noble gas plasma. When the gas is cooled at constant pressure, the gas density increases and the concentration of molecular ions (A₂⁺) also increases. The presence of excited atoms and molecular ions in the plasma can influence the interaction of the plasma with dust particles, which determines the conditions for the existence and charging of dust formations in the plasma.

The glow discharge in neon, taking into account the influence of metastable atoms on its properties at pressures up to 187 Pa at cryogenic and room temperature, was studied in [2]. The gas pressure in the discharge tube was maintained constant in the course of cooling. The gas density in a discharge corresponded to the measured temperature at given pressure, namely, was proportional to the relation of 295 K to its measured value. It was noted that an increase in the concentration of molecular ions that may be observed with an increase of the gas density in the result of discharge cooling, can affect the longitudinal electric field of the discharge. With an increase of concentration of molecular ions the role of associative ionization increases. In positive-column plasma of rare-gases, the molecular ions appear for the most part in the reaction of termolecular association (conversion) of atomic ions A⁺ + 2A → A₂⁺ + A, as well as in reaction of associative ionization, while their destruction and loss proceed in the process of dissociative recombination in collisions with electrons A₂⁺ + e⁻ → A + A [11-13] and diffusion to the wall of the discharge tube. It was noted in [2] that associative
The simulation of plasma-dust systems has been carried out in a discharge with a radius of 3. Results to gas temperature. In neon, dissociative ionization occurs in reactions of the interaction of excited atoms (Ne*) with the formation of molecular ions (Ne$_2^+$) and electrons (e') as Ne$^*$ + Ne$^*$ → Ne$_2^+$ + e' [14] and leads to a decrease in the electric field. The role of metastable atoms in the interaction of plasma with dust particles was studied in [15-19] at room temperature for various dust particle distributions in dust structures, but no molecular ions were considered. This paper is devoted to the investigation of neon plasma with dust particles at cryogenic temperature. In this paper, we numerically study the influence of molecular ions on discharge parameters and the charge of dust particles in a dc glow discharge in neon at $T = \text{77 K}$ and a pressure up to 100 Pa (measured at 295 K).

2. Numerical model

Numerical study of the effect of molecular ions on the parameters of glow discharge plasma in neon and the charge of dust particles was carried out on the basis of the diffusion-drift model of positive column of a glow discharge in neon with microparticles [15, 16]. The model takes into account the formation, drift, diffusion and losses of electrons, atomic and molecular ions and metastable neon atoms in the plasma volume, on the walls of the discharge tube and on dust particles. In addition to the previously considered processes, the formation of molecular ions in the reaction of the termolecular association Ne$^+$ + 2Ne → Ne$_2^+$ + Ne, with the rate constant taken from [11], as well as their losses in the process of dissociative recombination of Ne$_2^+$ + e' → Ne + Ne in collisions with electrons with the rate constant taken from [13]. As in [13], it was assumed that the rate of formation of molecular ions in the process of associative ionization can be neglected in comparison with the rate of termolecular association (ion conversion).

The average electron energy and their transport coefficients were calculated using the BOLSIG+ package [20]. The mobility of molecular ions was taken from [21].

The charge of dust particles was calculated taking into account the ion-atom collisions in the CEC approximation (collision enhanced collection) [22], and the fluxes of metastable atoms to dust particles were simulated in the gas kinetic approximation. The possible heating of ions [23] was not taken into account, and the temperature of the heavy components of plasma was assumed to be equal to gas temperature.

3. Results

The simulation of plasma-dust systems has been carried out in a discharge with a radius of $R = 8\ \text{mm}$, at neon pressure $P = 20 - 100\ \text{Pa}$ and a discharge current $I = 0.6\ \text{mA}$. To simulate the dust structures formed by spherical dust particles with a diameter of 4.14 µm, the dust cloud radius $r_d$ with a density distribution of dust particles $n_d(r)$, described by a step blurred function, have been chosen:

\begin{align}
    n_d(r) &= n_{d,0}, \quad r \leq r_d \\
    n_d(r) &= n_{d,0} \exp[10(r_d - r)/R], \quad r > r_d
\end{align}

where $n_{d,0}$ is the concentration of dust particles on the discharge axis. The two different sets of the parameters $r_d$ and $n_{d,0}$ of dust particle distribution were studied: $r_d = 2\ \text{mm}$, $n_{d,0} = 10^{11}\ \text{m}^{-3}$ and $r_d = 0.5\ \text{mm}$, $n_{d,0} = 10^{12}\ \text{m}^{-3}$, that were observed in experiments [9, 10]. To analyze the effect of the plasma composition on the charge of dust particles, simulations also have been made of the charge of a solitary test dust particle introduced in the discharge. Examples of calculation are presented in figures 1 and 2 for a pressure of 84 Pa and discharge current of 0.6 mA.

Figure 1 shows the radial profiles of the concentrations of plasma particles in a discharge without dust particles (a) and in a discharge with a dust structures (b, c). It was discovered that the concentrations of electrons (1) and atomic ions (2) in discharge without dust particles (figure 1(a)) differ little from each other, since the concentration of molecular ions (4) is small in comparison with
Figure 1. Radial profiles of electron concentration $n_e$ (1), atomic ions $n_i$ (2), metastable atoms $n_m$ (3) and molecular ions $n_{i2}$ (4) in the positive column of discharge at $P = 84$ Pa, $I = 0.6$ mA in a discharge without dust particles (a) and in presence of dust structures with various distributions of dust particles (b) and (c).

the concentration of atomic ions (2). In presence of dust structures, the ion and electron profiles principally differ. One can see in figure 1(b, c) that in the region of localization of the dust structure the total concentration of ions is much higher than the concentration of electrons due to the effect of a plasma-dust trapping [24, 25].

Simulations show that in the discharge with dust particles the rate of termolecular conversion increases inside the dust structure. This leads to the fact that within the dust structure, the fraction of molecular ions ($n_{i2}/n_i$) is significantly higher than in a discharge without dust particles increasing with the increase of $n_d$.

The corresponding values of the longitudinal electric field were calculated, which amounted to 11.89 V/cm in the discharge without dust particles, 15.64 V/cm for $r_d = 2$ mm and $n_{d,0} = 10^{11}$ m$^{-3}$, and 12.00 V/cm for $r_d = 0.5$ mm, $n_{d,0} = 10^{12}$ m$^{-3}$. Without taking into account the molecular ions, the corresponding values were 11.94, 15.4, and 11.96 V/cm, respectively. Apparently, the main factor responsible for the influence of molecular ions on the plasma parameters is the higher mobility of molecular ions compared to atomic ions. The concentration of molecular ions in a discharge without dust particles increased with increasing gas pressure and, when the pressure converted to the value at room temperature, was close to the data given in [13] obtained at a temperature of 300 K.

One can analyze the change of the of dust particle charge $Z_d$ in presence the molecular ions. Figure 2 represents the charge of a dust particle in a discharge with the same values of neon pressure and discharge current, but various distributions of dust particles. At $n_{d,0} = 10^{11}$ m$^{-3}$, the change in the charge of dust particles when the molecular ions were taken into account, was negligible. However, at a higher concentration of dust particles $n_{d,0} = 10^{12}$ m$^{-3}$ observed in dust structures formed at cryogenic
temperature [10], the charge of dust particles decreased by several percent with account of molecular ions.

The simulations have shown that the flux of molecular ions practically had no direct effect on the charge of a solitary dust particle in the investigated range of parameters, their effect was mainly due to a change in an electron temperature. Though, within a dense dust structures with a deficit of free electrons in a plasma, the charge of a dust particles may decrease due to the higher mobility of molecular ions than atomic ones. The account of molecular ions can improve the accuracy of the results in the simulation of dense dust structures observed at cryogenic temperatures [10].

4. Conclusion
The effect of molecular ions on the parameters of dusty plasmas at cryogenic temperature in dc discharge in neon have been studied. It have been found that the role of molecular ions in dusty plasmas is higher than in a discharge without dust particles at the same neon pressure and discharge current. In the presence of dust particles in the discharge, the fraction of molecular ions increases in the region of localization of dust structure. At dust particle concentrations up to $10^{11} \, \text{m}^{-3}$, molecular ions do not significantly affect the electron temperature, and there is no significant effect of their flux to the dust particle surface, on the dust particle charge. When the concentration of dust particles increases by an order of magnitude, the fraction of molecular ions increases, and their effect on the charge of dust particles becomes noticeable. Thus, taking into account molecular ions one can improve the accuracy of simulation with dense dust structures with dust particle concentration exceeding $10^{12} \, \text{m}^{-3}$, which were observed at cryogenic temperatures.

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