The influence of dust particles concentration on glow discharge parameters and dust particle charge

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Abstract: A self-consistent kinetic model of the influence of dust particle concentration $N_d$ on the parameters of the positive column of a low pressure DC glow discharge in noble gases based on Boltzmann equation for electron energy distribution function is presented. It is shown that the increase of $N_d$ leads to the increase of averaged electric field and ion density, and to the decrease of dust particle charge in the dusty cloud.

Keywords: Dusty plasma, Boltzmann equation, Havnes parameter, volume recombination.

1. Introduction

Dusty or complex plasma is ionized gas of electrons, ions, and negatively charged micron-sized particles [1-3]. Dusty grains can be found either in space (e.g. planet rings, interstellar molecular clouds, cometary tails) or in different technological processes (e.g. plasma chemical deposition and coating, thermonuclear reactors, etc.). In reactive plasmas used in semiconductor industry, dust particles are polydispersed fine particulates with radii in the nano- or micrometre range, which are produced from the plasma itself by the coagulation of smaller clusters or polymerization of gas dissociation products. These fine particulates form a cloud electrically levitating above the wafer and contaminate the wafer falling on it when the applied voltage on the wafer is turned off [4]. In laboratory conditions, dusty plasmas are also intensively investigated in the positive column (PC) of a DC glow discharge and in RF discharges in noble gases at low gas density. Many interesting phenomena are observed and investigated in dusty plasma, e.g. formation of dusty structures (Coulomb crystals, liquids and gases), phase transitions, vortices, wave propagation, and different kinetic processes. For the current state of the field, see recent review papers [1-3].

Laboratory dusty plasma in DC or RF discharges consists of electrons and ions with densities $n_i \approx n_e \approx 10^7-10^9$ cm$^{-3}$ and dust particle with the dust number density is usually $N_d \sim (10^2-10^4)$ cm$^{-3}$ and the charge $eZ_d = (10^{-2}-10^{-1})$ e. For a small Havnes parameter, $P_H = Z_d N_d n_i / n_e < 1$, the charge of dust particles is determined only by the plasma conditions. With the increase of $P_H$, the local parameters in the plasma region containing dust particles (the electron density and electron energy distribution function (EEDF)) change, which in turn leads to a change of the average charge of dust particles and, hence, of all properties of dusty plasma. For conditions of RF discharge used for thin films preparation in the semiconductor industry, the influence of dust particles on discharge properties was investigated with the help of particle-in-cell Monte Carlo simulations by J.P. Boeuf [5]. It was understood that each dust particle acts as an electron and ion sink, and a large concentration of dust particles will have some effect on the plasma properties and on the plasma sustainment conditions. Unfortunately, in late 1990s and 2000s, this important conclusion was almost forgotten in a great number of investigations of dusty plasmas (Coulomb structures, waves and so on) in laboratory conditions. Moreover, in almost all the papers published on dusty plasma in this period, the electron energy distribution function in dusty plasma was assumed to be Maxwellian. In the paper by Lipaev et al. (1997) [6], a simple estimation was presented where it was assumed that in dusty plasma in the PC of a glow discharge the electron and ion losses on dust particles should be compensated in ionizing collisions, and an axial electric field in a discharge tube should increase in the region containing dust particles. However, this effect has a kinetic nature and must be treated in terms of non-equilibrium EEDF, which is not Maxwellian in the PC of glow a discharge [7]. Moreover, a self-consistent consideration of mutual plasma-dust interaction should be investigated.

This paper presents the first attempt to make a self-consistent kinetic investigation of the influence of dust particle concentration $N_d$ on the parameters of the PC of a low pressure DC glow discharge in noble gases on the basis of Boltzmann equation for EEDF. It should be stressed that even without dusty particles the glow discharge in cylindrical tubes is a very complex open non-equilibrium system of neutral atoms (in different electronic states), ions and electrons. For some conditions, the low-pressure glow discharge can be self-organized (stratification of PC), with non-local processes playing an important role. The addition of dust particles increases the complexity of the description of a glow discharge. We will consider a simplified model that nevertheless can highlight the main problems of the interference of dust particle concentration and plasma conditions in a discharge.
2. Model

The charging of an individual dust particle was considered in the OML approximation for the case when the mean free path length of ions in plasma is much larger than both \( r_D \) and screening length \( \lambda \) [1]. In this paper, we did not take into account the formation of trapped ions around the negatively charged dust particle and additional collisional ion flux. In papers [8,9], it was deduced that the trapped ions lead to some shielding of the charge of a dust particle, and collisional ion flux leads to some decrease of the dust particle charge. However, this fact does not change the qualitative conclusions about the influence of dust particle concentration on the discharge parameters. It is more important for the aim of the paper to take into account the non-Maxwellian character of EEDF in non-equilibrium low temperature dusty plasma.

We consider the positive column of a glow discharge without striations in the discharge tube in a quasi-2D arrangement. Without dust particles in the tube, the axial electric field is \( E \), and the electron current density on the tube axis has some value \( J_e \). In a steady state, the creation of new electrons and ions per unit time in the discharge tube due to gas ionization by electron impact is totally compensated for by their recombination on the tube wall that is governed by the process of ambipolar diffusion. All plasma parameters depend on a reduced electric field \( E/N_Z \) (\( N_Z \) is gas density). If we immerse micron-sized dust particles with number density \( N_d \) into a discharge then electrons and ions will also take part in the recombination on the dust particle surfaces.

A dust particle charge is determined by the equality of electron and ion fluxes \( I_e = I_i \) on its surface from the surrounding plasma. In non-equilibrium plasma of a gas discharge, the electron and ion fluxes to the surface of the particle are equal to

\[
I_e = \frac{2}{m_e} \int_0^{\infty} \sigma_{\text{cap,}e}(\varepsilon) f_1(\varepsilon) d\varepsilon, \quad (1)
\]

\[
I_i = \int_0^{\infty} \sigma_{\text{cap,}i}(V) f_i(\varepsilon) d\varepsilon, \quad (2)
\]

where \( f_1(\varepsilon) \) is the energy distribution function of electrons far from the dust particle and \( f_i(V) \) is the velocity distribution function of ions. The cross sections for electrons and ions to be captured by the dust particle (according to OML theory) are equal to

\[
\sigma_{\text{cap,}e}(\varepsilon) = \varpi \left[ 1 - \frac{\varepsilon \varphi(\eta_0)}{E} \right], \quad \varepsilon > |\varphi(\eta_0)|, \quad (3)
\]

\[
\sigma_{\text{cap,}i}(V) = \varpi \left[ 1 - \frac{2 |\varphi(\eta_0)|}{MV^2} \right], \quad (4)
\]

where \( \varpi = m_e v^2/2 \) is the kinetic energy of an electron, \( \varphi(\eta_0) = -e^2Z_dF_0 \) is the particle surface potential that depends on the EEDF [7]. In this paper, we assume that the ion velocity cross section can be approximated by the shifted Maxwell distribution with the ion temperature \( T_i \) and the ion drift velocity \( V_i = \mu_i E \) (\( \mu_i \) is the ion coefficient of mobility).

The EEDF is obtained with the help of a steady state Boltzmann equation in a two term approximation for isotropic \( f_0(z,\varepsilon) \) and anisotropic \( f_1(z,\varepsilon) \) parts of EEDF. 

\[
\varepsilon = m_e v^2/2
\]

is the kinetic energy of electron:

\[
-\frac{eE_e}{3E_e} \varepsilon f_0(z,\varepsilon) = S_e(f_0) + S_a(f_0) + S_d(f_0), \quad (5)
\]

\[
-\frac{eE_i}{3E_i} \varepsilon f_i(z,\varepsilon) = -(N_e \sigma_m(\varepsilon) + N_d \sigma^r_m(\varepsilon)) f_1(z,\varepsilon), \quad (6)
\]

where \( S_e(f_0) \) is the integral of elastic, inelastic, and ionizing collisions of electrons with atoms [7]; \( S_a(f_0) \) is the term describing the recombination of electrons on the wall of a discharge tube with radius \( R \):

\[
S_a(f_0) = -f_0 / \tau_e, \quad (7)
\]

where \( \tau_e = (R/2.405)^2/D_e \) is the characteristic time of electrons losses on the wall, \( D_e \) is the coefficient of ambipolar diffusion. In particular, the balance of electron losses on the wall described by the term \( S_a(f_0) \) and electron creation in ionizing collisions with neutral atoms determines the value of the axial component of the electric field in the region of the discharge tube without dust particles. The presence of dust particles leads to the absorption and recombination of electrons with energies higher than the particle potential \( |\varphi(\eta_0)| \), \( Z_d \varepsilon^2 / n_0 \) on the particle surface. The last term in Eq. (5) determines the electron losses due to electron absorption on dust particles:

\[
S_d(f_0) = -N_d \sigma_m(\varepsilon) f_0(\varepsilon), \quad (8)
\]

In the right side of Eq. (7), the first term describes the loss of electron momentum in elastic and inelastic collisions with neutral atoms, and the last term reflects the loss of electron momentum due to the absorption on dust particles. The momentum cross section \( \sigma_m(\varepsilon) \) for scattering of electrons on the dust particle can be calculated for a given potential of the dust particle. Functions \( f_0(\varepsilon) \) and \( f_1(\varepsilon) \) determine the electron density \( n_e(z) \), and electron current density \( j_e \).

\[
\begin{align*}
\varpi & = \int_0^\infty f_0(\varepsilon) \sqrt{2m_e} d\varepsilon, \quad j_e = \frac{1}{3} \int_0^\infty f_1(\varepsilon) \sqrt{2m_e} d\varepsilon \sim n_e \mu_i E. \quad (9)
\end{align*}
\]

The total current through any discharge tube cross section is constant including the region where the cloud of dust particles is located. The axial component of the electric field \( E \) varies depending on the dust particles concentration in the cloud. The neutrality condition in dusty plasma should be satisfied:

\[
Z_d n_d + n_e = n_i. \quad (10)
\]

The presented model was calculated numerically in an iterative way. The dust particle radius \( r_0 \) and density \( N_d \), discharge tube radius \( R \), buffer gas (neon) density \( n_0 \) and
discharge current density \( j \) were the given parameters that can be changed independently (as it can be done in experiments, see, for example [10]). First of all, the solution of equations (5,6) without dust particles, \( N_D=0 \), permits us to obtain an EEDF. The axial electric field, \( E_a(N_D=0)=E_{dr} \), was determined by the balance of electron production in ionizing collisions with neutrals and their recombination on the tube wall due to ambipolar diffusion. The anisotropic part of EEDF, \( f_i(\epsilon) \), was normalized by the condition that in the centre of the discharge tube the electron current density is equal to \( j_e(N_D=0)=1mA/cm^2 \). This normalization permits us to obtain electron density \( n_e \) from Eq. (9). The charge of a single particle immersed in plasma, \( Z_d(N_D=0) \), was calculated with the help of OML theory taking into account the non-equilibrium EEDF.

Then we considered the dust particles cloud with density \( N_d \neq 0 \). The Boltzmann equations (5,6) were calculated with some value of the axial electric field \( E_a(N_d \neq 0) \), which provides the equality of electrons and ions production and the total recombination on the walls and on the particle surface. A new value of electron density \( n_e \) was calculated with the help of Eq. (9) and a new ion density \( n_i \) was obtained from neutrality condition (10). After that, a new dust particle potential \( \phi_s \) was calculated. For the given dust particle density, the OML model for dust particle potential, the Boltzmann equation for EEDF, the neutrality condition were recalculated by the iterative method until all the parameters satisfied each other. Finally, for a new dust particle density \( N_d \), the procedure was repeated until full convergence.

3. Results

In Fig. 1, self-consistent solutions for dust particle potential \( \phi_s \) and axial electric field \( E_a \) dependences on dust particle concentration \( N_d \) are presented for dust particle radii, \( r_0 = 5 \mu m \) and 1 \mu m.

Fig. 1. Dust particle surface potential (solid line – particle radius \( r_0 = 5 \mu m \), dotted line – \( r_0 = 1 \mu m \) and axial electric field (dashed line – \( r_0 = 5 \mu m \), dashed dotted line – \( r_0 = 1 \mu m \)) dependences on dust particle concentration.

It is seen that the self-consistent electric filed increases sharply with the increase of dust particle concentration in the region \( N_d r_0^2 > 10^7 \) cm\(^{-3} \) due to increased electron losses in recombination on dust particle. The dust particle potential decreases less with the increase of the dust particles concentration. It becomes only two times smaller when the product of the dust particle concentration and radius achieves the value \( N_d r_0^2 > 10^7 \) cm\(^{-2} \). The charge of dust particle \( Z_d \) is proportional to \( \phi_s r_0 \) [7]. It is interesting that for relatively small dust particle concentrations for different dust particle radii \( r_0 \) the electric field is the function of parameter \( N_d r_0^2 \), and in almost the whole \( N_d \)-region the dust particle potential is the function of parameter \( N_d r_0 \).

Fig. 2. Ion \( n_i \) (solid line), electron \( n_e \) (dashed line) and dust particle charge (dotted line) concentration dependences on dust particle concentration \( N_d = 1 \mu m \).

Fig. 3. The electron impact ionization \( S_i \) (solid line), charge recombination on the dust particle surface \( S_{Z_d} \) (dashed line), and wall recombination \( S_w \) (dashed dotted line) dependences on the dust particle concentration, \( N_d \), \( r_0 = 1 \mu m \).

In Fig. 2, the densities of ions \( n_i \), electrons \( n_e \), and the dust particles charge \( N_d Z_d \) are presented. The ion density
is obtained from the condition (10). In Fig. 3, the terms describing the production and losses of electrons and ions (in the right side of Boltzmann equation (5,6) are presented. In a steady state, the electron impact ionization is balanced by the processes of ions and electrons recombination on the discharge tube wall and on the dust particles surface. The term describing the electron recombination on dust particle surfaces, \( S_a \), is proportional to \( N_d \) and to the square of \( r_0 \) (8). The ionization term \( S_i \) increases with the increase of a reduced electric field, \( E_r/N_e \), which in turn increases with the increase of \( N_d \). It is also seen that the term \( S_a \) describing the wall recombination in the discharge tube is important only in the region of low dust particles concentration, \( N_d \).

In Fig.4, the EEDF for different dust concentration \( N_d \) and different radii \( r_0 \) are presented for the same value of a reduced electric field \( E_r/N_e \). The symbols present a self-consistent solution for different particle radii, \( r_0 = 1, 2 \) and \( 5 \) \( \mu \)m. It is seen that the EEDF has no visible peculiarities in the electron energy range \( \epsilon > -\phi_r(r_0) \) even for a high dust particle concentration, when electron recombination on dust particles becomes substantial. This result is rather unexpected and contradicts the naive conclusion that EEDF should deplete for \( \epsilon > -\phi_r(r_0) \) due to high energy electrons loss in absorption on dust particles. This fact reflects the self-consistent process of the adjustment of EEDF to a higher electric field in a dusty cloud relative to dust-free conditions in a discharge. Indeed, for the fixed electric field \( E_z \), dust particle charge \( Z_d \), and radius \( r_0 \), the non-self-consistent calculation demonstrates a substantial depletion of EEDF with the increase of dust particle concentration \( N_d \).

4. Conclusion

We have presented a self-consistent kinetic model based on the solution of the Boltzmann equation for EEDF, the OML model for a dust particle charge and the neutrality condition that describes interdependence between dust particle concentration and radii on parameters of gas discharge plasma (averaged electric field, electron and ions densities) and characteristics of individual dust particles (charges). It is shown that for dust particle concentrations in the region \( N_d r_0 \sim 10^3 \text{cm}^{-2} \) the charges of dust particles decrease but the Havnes parameter \( P_d = Z_d N_d/N_i \) increases, which means that dusty plasma can be regarded as electron depleted system, \( n_e < n_i \). In this region of dust particle concentrations (for different dust particle radii \( r_0 \)), the electric field is an increasing function of parameter \( N_d r_0^2 \), and in almost the whole \( N_d \)-region the dust particle potential is the function of parameter \( N_d r_0 \). The screening parameter \( \lambda = (T/4\pi e)^{1/2} \) for dust particles remains smaller than the mean separation between dust particles, \( (r_0 N_d)^{1/3} \). This fact justifies the approximation made in the model: electron and ion energy distribution functions are formed in the averaged electric field \( E_z(N_d) \) in the space between screened dust particles. For a higher concentration of dust particles, which can be achieved, for example, in cryogenic discharges [11], “\( \lambda \)-spheres” of dust particles become overlapped, and one should solve the non-local Boltzmann equation for EEDF in the electric field rapidly varying in space between dust particles. In this case, the complex plasma can be regarded as negatively charged dust particles immersed in the positive sea of ions that provide close coupling (attractive forces) between dusty grains.

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References