Influence of dust-particle concentration on gas-discharge plasma

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A self-consistent kinetic model of a low-pressure dc glow discharge with dust particles based on Boltzmann equation for the electron energy distribution function is presented. The ions and electrons production in ionizing processes as well as their recombination on the dust-particle surface and on the discharge tube wall were taken into account. The influence of dust-particle concentration $N_d$ on gas discharge and dust particles parameters was investigated. It is shown that the increase of $N_d$ leads to the increase of an averaged electric field and ion density, and to the decrease of a dust-particle charge and electron density in the dusty cloud. The results were obtained in a wide region of different discharge and dusty plasma parameters: dust particles density $10^5 - 10^8$ cm$^{-3}$, discharge current density $10^2 - 10^4$ mA/cm$^2$, and dust particles radius 1, 2, and 5 $\mu$m. The scaling laws for dust-particle surface potential and electric filed dependencies on dust-particle density, particle radius and discharge currents were revealed. It is shown that the absorption of electrons and ions on the dust particles surface does not lead to the electron energy distribution function depletion due to a self-consistent adjustment of dust particles and discharge parameters.

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I. INTRODUCTION

Dusty or complex plasma is an ionized gas of electrons, ions, and negatively charged micronized particles [1–3]. Dust grains can be found either in space (e.g., planet rings, interstellar molecular clouds, and 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 by 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 radio frequency (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, vortexes, 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 $n_i = n_e \sim 10^7 - 10^9$ cm$^{-3}$, dust particles with the dust number density $N_d \sim (10^2 - 10^3)$ cm$^{-3}$, and the charge number $eZ_d = (10^3 - 10^6)e$. For a small Havnes parameter, $P_H = Z_d N_d / n_e < 1$, the charge of dust particles is determined only by 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 the conditions of RF discharge used for thin films preparation in the semiconductor industry, the influence of dust particles on discharge properties was investigated in 1992 with the help of particle-in-cell Monte Carlo simulations by Boeuf [5]. Recently, the dusty plasma of RF discharge was investigated by Denysenko et al. [6–8] with the help of Boltzmann equation for EEDF. Different models for reactive dusty plasma of RF discharge were also presented by Goedheer et al. [9] and Schweigert et al. [10]. It was understood that each dust particle acted as an electron and ion sink, and a large concentration of dust particles would have some effect on the plasma properties and on the plasma sustainment conditions. The electron and ion losses on dust particles should be compensated in ionizing collisions, and an averaged electric field in a discharge should increase in the region containing dust particles.

The main aim of this paper is to develop a self-consistent kinetic model of a low-pressure dc glow discharge with dust particles on the basis of Boltzmann equation for EEDF and to describe the influence of dust particles on the parameters of the positive column of a discharge in different noble gases in the wide range of dust-particle concentration $N_d$ (from $10^6$ cm$^{-3}$ to $10^9$ cm$^{-3}$), radii $r_d$, and discharge current density.

II. MODEL

It should be stressed that even without dusty particles the glow discharge in cylindrical tubes is a very complex open nonequilibrium 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 a positive column) with nonlocal processes playing an important role. The addition of dust particles increases the complexity of the description of a glow discharge. Here we consider a simplified model that nevertheless can highlight...
the main problems of the interference of dust-particle concentration and plasma conditions in a discharge.

The charging of an individual dust particle is considered in the Orbital Motion Limited (OML) approximation for the case when the mean-free-path length of ions \( l_i \) in plasma is much larger than both dust-particle radius \( r_0 \) and screening length \( \lambda_i \) [11,12]. In this paper, we do not take into account the formation of trapped ions around the negatively charged dust particle and additional collisional ion flux. In recent papers [13–19] it was deduced that the trapped ions led to some shielding of the charge of a dust particle, and collisional ion flux led 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 discharge parameters. It is more important for the aim of the paper to take into account the non-Maxwellian character of EEDF in nonequilibrium low-temperature dusty plasma of dc discharge.

We consider the positive column of a glow discharge without striations in the discharge tube in a quasi-two-dimensional (quasi-2D) arrangement. Without dust particles in the tube, the axial electric field is \( E_z \) 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. It is known that EEDF depends only on a reduced electric field \( E_z/N_e \) (\( N_e \) is the gas density). If we immerse micronized 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. Electrons recombination on the dust particle can be regarded in the Boltzmann equation for EEDF as a volume recombination.

A dust-particle charge is determined by a zero total current of electrons and ions \( (I_e+I_i=0) \) on its surface from the surrounding plasma. In nonequilibrium plasma of a gas discharge, the electron and ion currents to the surface of the particle are equal to

\[
I_e = -e \sqrt{2 \frac{m_e}{\pi \epsilon_0}} \int_0^\infty \sigma_{cap,e}(\epsilon) f_0(\epsilon) e \, d\epsilon,
\]

\[
I_i = \epsilon \int_0^\infty \sigma_{cap,i}(V) F_1(V) V_d^2 \, d\epsilon,
\]

where \( f_0(\epsilon) \) is the isotropic part of the electron energy distribution function far from the dust particle and \( F_1(V) \) 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

\[
\sigma_{cap,e}(\epsilon) = \frac{\pi r_0^2}{1 - \frac{|e \varphi(r_0)|}{\epsilon}}, \quad \epsilon > |e \varphi(r_0)|;
\]

\[
\sigma_{cap,e}(\epsilon) = 0, \quad \epsilon < |e \varphi(r_0)|
\]

where \( e=m_e \omega^2/2 \) is the kinetic energy of an electron, \( \varphi \approx \varphi(r_0) = -eZ_d/r_0 \) the particle surface potential that depends on the EEDF.

In this paper, we assume that the ion velocity distribution can be approximated by the shifted Maxwell distribution with the ion temperature \( T_i \), and the ion drift velocity \( \vec{V}_d \) is \( \mu_i \vec{E} \) (\( \mu_i \) is the ion coefficient of mobility). The ion flux can be easily obtained [1,20],

\[
I_i(r,z) = \sqrt{\frac{2T_i}{\pi m_i}} \pi a^2 \eta_i(z) \left[ \exp \left( -\frac{m_i V_i^2}{2T_i} \right) \right. + \left. (1 + m_i V_i^2/T_i + 2W_i/T_i) \sqrt{\pi} \epsilon_0 \left( \frac{m_i V_i^2}{2T_i} \right) \right].
\]

In this paper, in order to obtain the electron distribution function, \( F(\vec{v}) \), the kinetic Boltzmann equation was used,

\[
\frac{\partial F}{\partial t} + \vec{v} \cdot \frac{\partial F}{\partial \vec{v}} - \frac{e}{m_e} F \frac{\partial F}{\partial \vec{v}} = S_i(F)
\]

where \( S_i \) is the total collisional integral, \( m_e \) is the mass of the electron. In a steady state in a homogeneous DC glow discharge with the given value of axial electric field \( E_z \), the kinetic Boltzmann equation is written as

\[
-\frac{e}{m_e} F \frac{\partial F}{\partial v_z} = S_i(F)
\]

For a weak electric field directed along axis \( z \) the assumption of low anisotropy relative to direction \( z \) is quite appropriate. The first two terms of the expansion in Legendre polynomials were taken into account,

\[
f(\epsilon, \frac{v_x}{v}, \frac{v_y}{v}) = \frac{1}{2\pi(2m)^{3/2}} \int_{-1}^{1} f_0(\epsilon) + f_1(\epsilon, \frac{v_x}{v}, \frac{v_y}{v}) \, d\epsilon.
\]

Here \( f_0 \) is the isotropic part and \( f_1 \) is the anisotropic part of EEDF. Using expansion (7) in Eq. (6) and integrating over angles one can obtain the Boltzmann equation for the isotropic and anisotropic parts of EEDF,

\[
-\frac{eE_z}{3e} \frac{\partial f_0}{\partial \epsilon} = S_e^{\text{el}}(f_0) + S_i^{\text{el}}(f_0) + S_m(f_0) + S_x(f_0)
\]

\[
-\epsilon E_z \frac{\partial f_0}{\partial \epsilon} = -(N_e \sigma_m(e) + N_d \sigma_m^{\text{el}}(e)) f_1(e),
\]

where \( S^{\text{el}}(f_0), S^{\text{in}}(f_0), \) and \( S_m(f_0) \) are the integrals of elastic, inelastic, and ionizing collisions of electrons with atoms,

\[
S^{\text{el}}(f_0) = m_e \frac{1}{M_i \omega^2} \frac{\partial}{\partial v} \left[ v^3 \nu_m(v) \left( f_0 + \frac{k_g T_i}{m_i} \frac{\partial f_0}{\partial v} \right) \right].
\]
The ionization term is
\[ S_{\text{ion}}(f_0) = N_d \frac{1}{v} (2v^2 - v_i^2) \frac{\sigma_{i}(v^2 + v_i^2)f_0(v^2 + v_i^2)}{-v^2 \sigma_{\text{ion}}(v^2)f_0(v^2)}. \]
(11)

The presented model was calculated numerically in an iterative way. The dust-particle radius \( r_d \) and density \( N_d \) discharge tube radius \( R \), the gas (neon) density \( N_g \) and discharge current density \( j_e \) were the assigned parameters that can be changed independently (as it can be done in experiments, see, for example, [21,22]). First of all, the solution of Eqs. (8) and (9) without dust particles \((N_d=0)\) permits us to obtain the EEDF. In this case, the axial electric field, \( E_z(N_d=0) = E_{0z} \), is determined by the balance of electron production in ionizing collisions with neutrals and their recombination on the tube wall as a result of ambipolar diffusion. The anisotropic part of EEDF, \( f_1(e) \), was normalized by the condition that in the center of the discharge tube the electron current density is equal to \( j_e(N_d=0) \). This normalization permits us to obtain electron density \( n_e \) from Eq. (15). The charge of a single particle immersed in plasma, \( Z_d(N_d=0) = Z_d(0) \), was calculated with the help of OML theory taking into account nonequilibrium EEDF.

Then we consider the dust particles cloud with density \( N_d \neq 0 \). Boltzmann Eqs. (8) and (9) were calculated with some value of the axial electric field \( E_z(N_d \neq 0) \), which provides an equal production of electrons and ions and their 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. (15) and a new ion density \( n_i \) was obtained from neutrality condition (16). After that, a new dust-particle potential \( \varphi_{d} \) was calculated. For the given dust-particle density, the OML model for the dust-particle potential, the Boltzmann equation for EEDF, and the neutrality condition were recalculated using the iterative method until all the parameters correlated with each other. Finally, for a new dust-particle density \( N_d \), the procedure was repeated until full convergence.

III. RESULTS

In Fig. 1, self-consistent solutions for dust-particle potential, \( \varphi_{d} \), and axial electric field, \( E_z \), dependencies on dust-particle concentration, \( N_d \), are presented for dust-particle radii, \( r_0 = 1.2 \) and \( 5 \) \( \mu m \). It is seen that the self-consistent electric filed increases sharply with the increase of dust-particle concentration in the region \( N_d/r_0^2 > 10^{-2} \) \( {cm}^{-1} \) due to increased electron losses in the process of recombination on dust particles. There is a smaller decrease of particle potential with the increase of the dust-particle concentration. It becomes only two times smaller when the product of the dust-particle concentration and radius achieves the value \( N_d r_0 > 10^{-2} \) \( {cm}^{-2} \). 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 \( N_d/r_0 \).

In Fig. 2, the densities of ions \( n_i \), electrons \( n_e \), and the dust particles charge \( N_d/Z_d \) are presented for different discharge current densities: \( j_z = 1 \) and 10 mA/cm\(^2\). Due to weak dust-particle charge dependence on dust-particle concentration, \( Z_d(N_d) \), the product \( N_d/Z_d \) is almost proportional to \( N_d \). Since the electron current is constant for each dependence, the electron density decreases almost inversely proportional to the electric field. The ion density is obtained from the condition of quasi neutrality [Eq. (16)]. In the region of high dust-particle concentration \( (P_H \gg 1) \) the dust particles and plasma parameters are determined mainly by dust particles and ions. For the regime \( j_z = 10 \) mA/cm\(^2\) the initial electron and ion densities are 10 times higher than for \( j_z = 1 \) mA/cm\(^2\). For higher discharge currents the region of high Havnes parameter \( P_H \gg 1 \) appears at higher dust-particle concentration.

In Fig. 3, the frequencies of the production and losses of electrons and ions [which correspond to the terms in the right side of Boltzmann Eq. (8)] are presented for two values of initial reduced electric field, \( E_0/p \). The higher value of initial electric field corresponds to the higher frequencies of electron-impact ionization (12) and recombination on wall (13), and, consequently, to the smaller value of discharge tube radius, \( R \). The ionization term \( S_{ion} \) increases with the increase of reduced electric field, \( E_0/N_d \), which in turn increases with the increase of \( N_d \). It is seen that the term \( S_{r} \), describing the recombination on the discharge tube wall is important only in the region of low dust-particle concentration, \( N_d \). From Fig. 3, it is also seen that for a smaller tube radius (higher initial electric field) more dust is needed before the recombination on the dust starts to play a role. It should be stressed that for large values of dust-particle concentration, \( N_d \), the solutions for all plasma parameters (EEDFs, electron and ion densities, frequencies of electron-impact ionization and recombination on the particles, average electric fields and dust-particle surface potential) do not depend on the values of initial electric field in discharge tube.

Self-consistent solutions were also made for different values of discharge current density: \( j_z = 0.1 \) mA/cm\(^2\), \( j_z = 1 \) mA/cm\(^2\), and \( j_z = 10 \) mA/cm\(^2\). For given particle radius and different discharge currents, the electric-field dependencies are almost the same, while the particle surface-potential dependencies on \( N_d \) approximately obey the scaling law \( \varphi_s = \Phi(N_d/r_0/j_z) \). In Fig. 4, the dependencies of the particle surface potential on the scaling parameter \( N_d/r_0/j_z \) are presented for different particle radii and different discharge current densities \( (r_0 = 1, 2, 5 \ \mu m \ and \ j_z = 0.1, 1, \ and \ 10 \ mA/cm^2) \). All the data are seen to lie onto the same curve. This fact may seem to be unexpected because Boltzmann Eqs. (8) and (9) are nonlinear respective to EEDF due to the implicit de-
dependence of self-consistent dust-particle potential \( |\varphi_d| \) on EEDF. However, the condition of quasineutrality (16) plays an important role. The condition (16) can be written in the form

\[
e(n_i - n_e)/j_z = (N_d r_0 |\varphi_d|/j_z).
\]

For a small concentration of dust particles, electron and ion densities are proportional to the discharge current density, and \( n_i = n_e \). In this case, dust-particle potential \( |\varphi_d| = eZ_d/r_0 \) depends only on the value of EEDF irrespective to the value of \( j_z \) (the left side of curve in Fig. 4). In the case of very large dust-particle concentration \( N_d \), the ions density \( n_i \) is much larger than the electrons density, \( n_i \gg n_e \), and the ratio \( n_i/j_z \) is equal to some constant, which depends on the complex \( N_d r_0 |\varphi_d|/j_z \). It means that \( |\varphi_d| \approx (N_d r_0/j_z)^{-1} \). In Fig. 4, the right branch of the curve can be approximated as \( |\varphi_d| \approx (N_d r_0/j_z)^{-1} \). The smaller radii \( r_0 \) of dust particles (or the higher discharge current) the higher concentration of dust particle \( N_d \) is needed to influence the dusty plasma parameters.

The calculations were also made for different inert gases, i.e., argon, neon, and helium (see Fig. 5). The absolute values of dust particles potential differ for different gases due to different ionic masses and different EEDFs (electron temperatures) for the given reduced electric field.

In Fig. 6, the electron energy distribution functions for different dust concentration \( N_d \) and different radii \( r_0 \) are presented for the same value of a reduced electric field \( E_z/N_d \). The symbols present a self-consistent solution for different particle radii, \( r_0 = 1, 2 \) and 5 \( \mu m \). It is seen that under these conditions the EEDF has no visible peculiarities in the electron energy range \( e > -e\varphi_d(r_0) = e^2Z_d/r_0 \) 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 \( e > -e\varphi_d(r_0) \) due to high energy electrons loss in the course of 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, the nonself-consistent calculation demonstrates a substantial depletion of EEDF with the increase in dust-particle concentration \( N_d \).

The presented results show that for some high dust-particle concentration \( (N_d r_0^2 > 10^{-1} \text{ cm}^{-1}) \) the electric field dramatically increases. In this region, the ionic component of current density becomes important, especially due to the increase of ions concentration and decrease of electrons concentration. 2D self-consistent consideration of electron kinetics, drift and diffusion of ions and dust particles coupled with the Poisson equation should be applied in this region.

### IV. 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 the interdependence between characteristics of individual dust particles (concentration and radii) and parameters of gas-discharge plasma (averaged electric field, electron and ions densities, and discharge current densities). It is shown that for dust-particle concentrations in the region \( N_d \sigma_0 \sim 10^0 - 10^2 \) cm\(^{-2}\) the charges of dust particles decrease but the Havnes parameter \( P_H = Z_d N_d / n_e \) increases, which means that dusty plasma can be regarded as electron depleted system, \((n_e < n_i)\). The following scaling laws were obtained: for different dust-particle radii \( r_d \), the electric field is an increasing function of parameter \( N_d r_0 / j_z \), for all presented data the dust-particle potential is the function of parameter \( N_d r_0 / j_z \). The screening parameter \( \lambda_j = (T_j / 4 \pi m_j)^{1/2} \) for dust particles remains smaller than the mean separation between dust particles, \( l_p \sim N_j^{1/3} \). This fact justifies the approximation made in the model: electron and ion energy distribution functions are formed in the averaged electric field \( E_x(N_d) \) in the space between screened dust particles. For a higher concentration of dust particles, which can be achieved, for example, in cryogenic discharges \cite{23}, “\( \lambda \) spheres” of dust particles become overlapped, and one should solve the nonlocal 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. Free electrons will be contained in such plasma in very little amounts necessary only to compensate for pair recombination of ions and electrons on the surfaces of dust particles. This situation is similar to the one in metals, where heavy positive ions are immersed in the negative sea of electrons.

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