

Transport of Dust Particles in Glow-Discharge Plasmas

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A theory is presented for the transport of dust particles in glow-discharge plasmas. The forces which act on the negatively charged dust particles are examined and estimated. The dominant force is shown to be dependent upon the particle size and location within the discharge.

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rf and dc glow-discharge plasmas are extensively utilized in the manufacturing of semiconductor integrated circuits. Dust particles ranging in size from tenths of microns to microns have been observed in these plasmas by several researchers [1-6]. These particles are detected hovering above electrode and wall surfaces using laser light scattering at several optical frequencies. Other authors have reported on various theoretical aspects of particle behavior in gaseous discharges [7-10]. None of these reports, however, fully explains the observed particle transport phenomena. In this Letter, a theory is outlined describing the transport of dust particles in electro-positive glow-discharge plasmas. An argon discharge with 10^{10}-cm^{-3} plasma density; electron and ion temperatures of 2.0 and 0.05 eV, respectively; and a neutral argon pressure of 100 mT is analyzed. Nucleation and growth kinetics are not discussed here.

Dust-particle charging mechanisms.—The potential difference between the dust particle and the plasma can be estimated using the orbit-motion-limited probe theory developed by Langmuir and Mott-Smith [11]. The dust particle is assumed to be spherical with uniform surface charge density and a geometrical radius r_p , which is smaller than the Debye length L_D . (The Debye length is also assumed to be less than an electron-neutral mean free path.) The dust-particle concentration is sufficiently low that particle-particle interactions which reduce the charge on the particle can be ignored. By assuming that the electrons are in thermal equilibrium, the electron density n_e is given by the Boltzmann relation

$$n_e = n_0 \exp \left[\frac{q(\phi - \phi_s)}{k_B T_e} \right], \quad (1)$$

where n_0 is the bulk plasma density, q is the electronic charge, k_B is Boltzmann's constant, T_e is the electron temperature, ϕ is the local potential, and ϕ_s is the bulk plasma potential.

The thermal electron current to the dust particle is given by

$$I_e = 4\pi r_p^2 q \frac{n_0}{4} \left[\frac{8k_B T_e}{\pi m_e} \right]^{1/2} \exp \left[\frac{q(\phi_p - \phi_s)}{k_B T_e} \right], \quad (2)$$

where m_e is the electronic mass and ϕ_p the potential at the surface of the particle. In the bulk of the glow

discharge, ion current velocities are typically less than the ion thermal velocity. The ion current entering the particle's sheath is then given by

$$I_i = 4\pi r_p^2 q \frac{n_0}{4} \left[\frac{8k_B T_i}{\pi m_i} \right]^{1/2} \left(1 - \frac{q(\phi_p - \phi_s)}{k_B T_i} \right) \quad (3)$$

for a random thermal ion current with a Maxwellian velocity distribution in the orbit motion limit [12]. T_i and m_i are the ion temperature and mass, respectively. Alternatively, the particle could be charged by the monoenergetic ion current directed toward a wall or electrode as is typical for a dust particle in the proximity of a wall sheath. In this case, the ion current to the particle is given by

$$I_i = \pi r_p^2 q n_0 v_i \left(1 - \frac{2q(\phi_p - \phi_s)}{m_i v_i^2} \right), \quad (4)$$

where v_i is the ion velocity. This represents a unidirectional current which charges the particle, rather than a spherically symmetric current as in Eqs. (2) and (3). Hence, the particle cross-sectional area rather than the surface area is used.

To calculate the potential difference between the dust particle and the plasma, i.e., $\phi_p - \phi_s$, the ion current, Eq. (3) or (4), is equated to the electron current, Eq. (2). This is analogous to the calculation of the floating potential on a collecting spherical probe in the orbit-motion-limited regime [12]. In this formulation, Eqs. (3) and (4) are equivalent when the ion velocity in these equations is replaced with the mean speed of the ions approaching the dust particle. [This includes replacing the thermal velocity and energy terms in Eq. (3) with the mean speed.] The mean speed is given by

$$v_s = \left[\frac{8k_B T_i}{\pi m_i} + v_i^2 \right]^{1/2}. \quad (5)$$

Therefore, when the directed ion velocity is low, either Eq. (3) or (4) is in the thermal limit. Alternatively, when the velocity is high, the ion current equations are in the monoenergetic beam limit.

The charge on the dust particle, Q_p , is estimated using basic electrostatic theory for a charged, conducting sphere. With the zero reference at infinity, it is given by

[13]

$$Q_p = (\phi_p - \phi_s) 4\pi\epsilon_0 r_p (1 + r_p/L_D), \quad (6)$$

where ϵ_0 is the permittivity of free space. (This approximation assumes that particle-particle interactions are negligible [14,15].) The mass M of the particle is determined by multiplying the density d by the particle volume. Consequently, the charge-to-mass ratio of a spherical particle becomes

$$\frac{Q_p}{M} = \frac{3\epsilon_0(\phi_p - \phi_s)}{r_p^2 d}, \quad (7)$$

which is inversely proportional to the radius squared.

Forces acting on the dust particle.—The first force which significantly affects the dust particle's transport is due to gravity. It has the form $k_g = Mg$ where g is the gravitational acceleration. The second force is exerted by E , the electric field; it is represented by $k_E = Q_p E$. At a particular location within the discharge, the ion velocity must be estimated as a function of the electric field for the solution of Eqs. (2) and (4). Therefore, the electrostatic force has a twofold dependency on the ion velocity. First, the electric field can be related to the ion velocity through a mobility term, and, second, the charge (and voltage) on the particle increases as the ion mean speed increases.

The third force, k_N , results from collisions with neutral gas atoms (or molecules) and is therefore proportional to the background chamber pressure. The interactions between the neutrals and the dust particle are assumed to be hard sphere, elastic collisions. (Since the dust particles considered here are much larger than neutral molecules, the interactions of the molecular dipoles with the charged particle are neglected.) Using the momentum-transfer cross section [16], the average momentum transferred per neutral is given by the average relative neutral velocity multiplied by the reduced mass and the cross-sectional area. (The reduced mass in this case is given by the neutral mass since it is much less than the dust-particle mass.) Therefore, k_N is approximated by

$$k_N = N v_R^2 m_n (\pi r_p^2), \quad (8)$$

where N is the neutral density, m_n the neutral mass, and v_R the average relative velocity between the neutrals and the dust particle. When the particle is drifting, this force is in the direction opposite to its motion. Alternatively, when there are net flows of neutral gas molecules, there is a momentum transfer to the particle in the direction of these flows, as shown experimentally by Jellum, Dougherty, and Graves [4] with thermophoresis.

The ion drag force k_i is caused by momentum transfer from the positive ion current which is driven by the electric field. This force consists of two components: the collection and the orbit forces. (In the following analysis, it is assumed that the dust-particle velocity is negligible and no ion interaction with the particle occurs outside of a

Debye length.) The collection force represents the momentum transfer from all ions that are collected by the particle. Each ion that impacts the particle transfers its original momentum, $m_i v_i$. Hence, this component is given by

$$k_i^c = n_i v_s m_i v_i \pi b_c^2, \quad (9)$$

where n_i is plasma density which is usually assumed to be equal to n_0 . The collection impact parameter is given by

$$b_c = r_p \left[1 - \frac{2q(\phi_p - \phi_s)}{m_i v_s^2} \right]^{1/2}, \quad (10)$$

based on orbit-motion-limited probe theory [11]. The orbit force [16] is given by

$$k_i^o = n_i v_s m_i v_i 4\pi b_{\pi/2}^2 \Gamma, \quad (11)$$

where

$$b_{\pi/2} = q Q_p / 4\pi\epsilon_0 m_i v_s^2 \quad (12)$$

is the impact parameter whose asymptotic orbit angle is $\pi/2$, and

$$\Gamma = \frac{1}{2} \ln \left(\frac{L_D^2 + b_{\pi/2}^2}{b_c^2 + b_{\pi/2}^2} \right) \quad (13)$$

is the Coulomb logarithm integrated over the interval from b_c to L_D . The contribution of the orbit force is zero when the collection impact parameter is greater than or equal to the Debye length.

The theory used for the ion drag force differs from the standard theory [17] for a negatively charged test particle in a uniform flux of positive ions. In the standard theory, the central test particle is considered to be a point charge. This does not properly account for the momentum transferred to the particle by ions that are collected. At low ion velocities (typically found in presheath regions), most of the ions within a Debye length are collected. Hence, in this work, the collected ions are treated in one manner and those that transfer momentum through orbits are treated in another. Moreover, typical approximations for the Coulomb logarithm are not applicable here since this term must be integrated with the proper limits to exclude those ions that are collected by the particle.

Discussion and results.—In Fig. 1, the ion drag and electrostatic and gravitational forces are depicted as a function of argon-ion velocity for 0.1-, 1-, and 10- μm particles. A standard form of the ion mobility for argon [18] has been used to determine the electric field corresponding to a given ion velocity. The variation of the charge on the particle with ionic velocity has been included in the ion drag calculation but shows little effect on the force curve. The pressure is 100 mT, the plasma density is 10^{10} cm^{-3} , the specific gravity of the particle is 2 (e.g., graphite), and the electron and ion temperatures are 2.0 and 0.05 eV, respectively. Typical neutral drag forces which oppose the particle's motion are 2.62

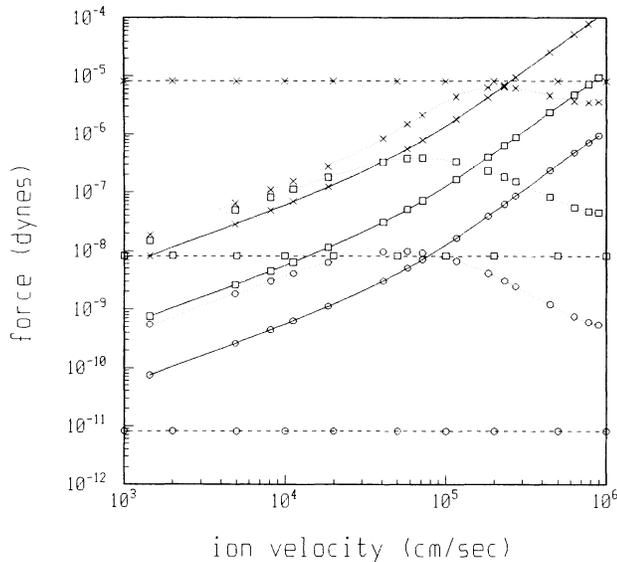


FIG. 1. Electrostatic force, ion drag, and gravitational force as a function of argon-ion velocity. These are depicted by the solid, dashed, and dotted lines, respectively. \circ , \square , and \times distinguish the 0.1-, 1.0-, and 10.0- μm particles, respectively, for each force.

$\times 10^{-12}$, 2.62×10^{-10} , and 2.62×10^{-8} dyn for the 0.1-, 1-, and 10- μm particles, respectively, for a particle velocity, v_p , of 2.2×10^2 cm/sec.

In addition to the force dependencies on ionic velocity, each of the forces has a different power-law dependency on the particle radius suggesting that the dominant force changes as the particle grows larger. The electrostatic force k_E is proportional to the particle radius since the charge is linearly dependent on the radius. The momentum transfer forces, k_N and k_i , are proportional to particle area and hence the radius squared. The ion drag force, however, only has this dependency over certain limited ranges of ion velocity. Finally, the gravitational force k_g is proportional to the particle mass which is proportional to the radius cubed.

The forces acting on the particles are sensitive to the location within the discharge. For the 0.1- μm particle, the ion drag force moves the particle towards a wall (or electrode) until it sees an electric field of approximately 14 V/cm in the wall sheath. At this point, the electrostatic and ion drag forces are in balance; the particle hovers above the wall surface. This is the point where the ion drag and electrostatic forces intersect in Fig. 1. For a 1- μm particle, this point occurs for an electric field of approximately 35 V/cm. Finally, the 10- μm particles are pulled toward the lower wall surface by gravity until an electric field of 82.6 V/cm is reached. The ion drag force is not the dominant force for particles of this size.

Particles with radii approximately 1 μm or less can be pushed by the ion drag force toward wall surfaces at the

top or bottom of a discharge vessel until the ion drag and electrostatic forces are in balance. If these particles grow in size, those at the top wall surface will eventually traverse the discharge to the bottom electrode where they will hover due to the force balance between the electrostatic and gravitational forces. Particles which hover above wall surfaces have been observed experimentally by several researchers [1,4,5,19]. Finally, if the particle continues to grow, the gravitational force pulls the particle closer to the bottom wall sheath. There it reaches a region (in the sheath) where the electron density is low. At this point, most of the particle's negative charge is neutralized and it drops out of the discharge.

Another spatial effect occurs when the particle sheath loses its symmetry about the particle. This occurs as a particle enters an electrode sheath since the plasma density decreases significantly as the electrode is approached. Consequently, the particle now has a considerable dipole moment associated with it. This dipole moment will not affect spherical particles significantly but will tend to align nonspherical (e.g., rod-shaped) particles with its direction. (The force resulting from the dipole moment interaction with the electric field is negligible in the particle transport when compared with the forces discussed in this paper.) Lastly, eddy and diffusion ion currents parallel to electrode surfaces have significant effects on particles suspended above wall surfaces due to the efficiency of the ion drag force at low ionic velocities. These parallel motions of the particulates have been observed experimentally [19].

It has recently been shown that the effective Debye length is reduced for small dust particles and low ion velocities in quiescent plasma [20]. This work is based on finite temperature, orbital motion probe theory using an isotropic monoenergetic ion distribution [21]. In this regime, the ion density increases near the particle and most likely screens the electrostatic force [14]. The ion drag force is also reduced since it is dependent on the Debye length. At higher concentrations, the dust particles coagulate at plasma-sheath boundaries. The charge on each particle is reduced and it has been postulated that they exist in a condensed phase [15]. To better approximate the effects of ion drag and include those due to screening, the trajectories of ions approaching a dust particle have to be accounted for in a self-consistent manner. Indeed, a great deal of numerical simulation is necessary to express dust-particle transport with a higher degree of accuracy than presented here. This theory does, however, elucidate the essential aspects of dust-particle transport in electropositive glow discharge plasmas.

[1] G. S. Selwyn, J. Singh, and R. S. Bennett, *J. Vac. Sci. Technol. A* 7, 2758 (1989).

[2] J. A. O'Neill, J. Singh, and G. G. Gifford, *J. Vac. Sci.*

- Technol. A **8**, 1716 (1990).
- [3] J. T. Verdeyen, J. Beberman, and L. Overset, *J. Vac. Sci. Technol. A* **8**, 185 (1990).
- [4] G. M. Jellum, J. E. Dougherty, and D. B. Graves, *J. Appl. Phys.* (to be published).
- [5] G. M. Jellum and D. B. Graves, *Appl. Phys. Lett.* (to be published).
- [6] H. M. Anderson, R. Jairath, and J. L. Mock, *J. Appl. Phys.* **67**, 3999 (1990).
- [7] A. Garscadden, in Proceedings of the Thirty-Seventh American Vacuum Society Conference, Toronto, 1990 [*J. Vac. Sci. Technol.* (to be published)].
- [8] J. Goree and T. E. Sheridan, in Proceedings of the Thirty-Seventh American Vacuum Society (Ref. [7]).
- [9] M. J. McCaughey and M. J. Kushner, *Bull. Am. Phys. Soc.* **36**, 219 (1991).
- [10] S. J. Choi, R. S. Averbach, and M. J. Kushner, *Bull. Am. Phys. Soc.* **36**, 220 (1991).
- [11] F. F. Chen, in *Plasma Diagnostic Techniques*, edited by R. H. Huddlestone and S. L. Leonard (Academic, New York, 1965), Chap. 4.
- [12] L. Schott, in *Plasma Diagnostics*, edited by W. Lochte-Holtgreven (Wiley, New York, 1968), Chap. 11.
- [13] J. O'M. Bockris and A. K. N. Reddy, *Modern Electrochemistry* (Plenum, New York, 1970).
- [14] I. Langmuir, *J. Chem. Phys.* **6**, 873 (1938).
- [15] H. Ikezi, *Phys. Fluids* **29**, 1764 (1986).
- [16] J. A. Bittencourt, *Fundamentals of Plasma Physics* (Pergamon, New York, 1986).
- [17] B. A. Trubnikov, *Reviews of Plasma Physics* (Consultants Bureau, New York, 1965), Vol. 1.
- [18] L. S. Frost, *Phys. Rev.* **105**, 354 (1957).
- [19] G. S. Selwyn, in Proceedings of the Thirty-Seventh American Vacuum Society Conference (Ref. [7]).
- [20] J. E. Dougherty, D. B. Graves, R. K. Porteous, and M. D. Kilgore, *Appl. Phys. Lett.* (to be published).
- [21] I. Bernstein and I. Rabinowitz, *Phys. Fluids* **2**, 112 (1959).