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Charging of aerosol particles in the near free–molecule regime

A.A. Lushnikov[a] and M. Kulmala

Department of Physical Sciences, University of Helsinki, P.O. Box 64, 00014 Helsinki, Finland

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Abstract. The charging of small neutral and charged particles suspended in weakly ionized plasma is investigated under the assumption that the Coulomb + image forces give rise to the ion transport in the carrier plasma and define the rate of charging processes. Our approach is based on a BGK version of the kinetic equation [1,2] describing the ion transport in the presence of force fields created by the particle charge and the image force. A special type of the perturbation theory (with respect to the reciprocal Knudsen number) is used for calculating the rate of ion deposition onto neutral and charged particles. As the starting approximation, the free–molecule ion distribution with a floating ion flux is used for evaluating the collision term in the Boltzmann equation. The value of the ion flux as a function of the particle size is then fixed self–consistently from the solution of the Boltzmann equation with the approximated collision term. The expression for the ion flux $J(a)$ to the spherical particle of radius a is derived in the form $J = \xi(a)J_{fm}$, where J_{fm} is the free–molecule flux (no carrier plasma) and $\xi(a)$ is a correction factor taking into account the ion–molecular collisions. The latter is shown to never exceed unity and to depend weakly on the particle–ion interaction.

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1 Introduction

Charged aerosol particles have attracted the attention of scientists and technologists for already more than a century [3–8]. Not to mention such remarkable phenomena like thunderstorms, linear and ball lightnings, formation of crystal–like structures in dusty plasma [9–12] charged aerosols play considerable role in the dynamics of particle formation in the atmosphere [5,13,14]. Not less remarkable is the role of charged particles in modern aerosol technologies and aerosol measurements [7,8,15–19]. These circumstances have motivated the permanently growing interest in the studies of aerodisperse systems wherein the aerosol particles are suspended in a plasma.

The kinetics of particle charging demands an answer to the question, how effective are the elementary charging processes? This problem had been considered by numerous authors theoretically [3,5,6,20–28] and experimentally [4,7,8,14,29–31]. More or less reliable results are found only for small Kn (Kn is the Knudsen number equal to the ratio of the ion mean free path to the particle size) [3,6,20] when the ion diffuses in the continuum regime. Meanwhile, small ($Kn \gg 1$) and transition particles ($Kn \propto 1$) are of most interest, especially for the Physics of Atmospheric Aerosols. The attempts to consider the free–molecule and the near free–molecule regimes can be found e.g. in references [21,23–27]. Analytical results are known for small neutral metallic particles. For charged particles there exist rather cumbersome expressions for the charging efficiencies [21,26]. No reliable results are found for dielectric particles. In references [26,27] attempts had been made to take into account the ion collisions with the molecules of the carrier gas.

In this paper we derive rather simple expressions for the charging efficiencies of small metallic particles, which are valid in the near free–molecule regime (large Kn). The total flux of ions onto the particle surface is expressed in terms of the product of the free–molecule flux with a correction factor taking into account the collisions of ions with the carrier gas molecules.

The plasmosols are characterized by three nondimensional groups:

- the Knudsen number $Kn = l/a$, i.e., Kn is the ratio of the ion mean free path l to the particle size a. In what follows we consider only spherical particles, so a is the particle radius. This parameter characterizes the regime of ion transport toward or outward from a disperse particle;
- the group qQe^2/akT, where e is the electron charge, q, Q are the ion and particle charges respectively (in units of e). T is the temperature in K and k is the Boltzmann constant. This parameter is the ratio of the particle electrostatic energy to the mean kinetic energy of ions. When $qQe^2/akT \propto 1$ the electrostatic interaction plays an important role in the processes of particle–ion interaction;

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[a] e-mail: `alex.lushnikov@helsinki.fi`

- the third group $q^2e^2/\lambda_i kT$ characterizes the electric state of the carrier plasma, i.e., the relative role of the Coulomb interaction in the charge transport processes. Here λ_i is the mean interion distance.

In what follows we consider a weakly ionized carrier plasma containing small dispersed particles, i.e., we focus on the case $a/l \ll 1$, $qQe^2/akT \propto 1$, and $q^2e^2/\lambda_i kT \ll 1$. This case suits very well the conditions of the Earth atmosphere, wherein the Knudsen number varies over a wide range (from 1 to 100) and $e^2/akT \propto 10$ for nanometric particles. This combination of parameters allows us to consider a pair: a suspended particle + an ion moving in a neutral carrier gas. Now our task is formulated as follows: to find the ion flux toward the particle (also neutral or charged), once the concentration of ions is known far away from the particle.

The most successful semi–phenomenological theory of the particle charging process had been presented in references [20,22]. The semi–phenomenological approach of these papers combines the solution of the diffusion equation in the external field with further matching the diffusion and free–molecule fluxes at a distance of the order of the ion mean free path. Although this approach describes the existing experimental data it cannot be considered as a microscopic theory of particle charging.

Another theory [26] starts with the free–molecule approximation corrected by a straightforward account of three–body effects. This approach also agrees satisfactory with the experimental data. On the other hand, the solution of the three–body problem cannot be performed analytically and some simplifications were done whose reliability leaves room for doubt. Moreover, it is absolutely clear that the most perfect approach should rely upon a kinetic equation.

An attack in this direction had been attempted in reference [27]. The authors applied a perturbation theory with respect to the reciprocal Knudsen number. The ideology of our approach is similar to that presented in this paper, but the version of the perturbation theory proposed by us is the principal difference.

The idea of our approach is extremely simple. We split the ion velocity–coordinate distribution into two terms f_0 and f_1, the first of which does not contribute to the total flux J whereas the second one does, $f = f_0 - Jf_1$. Here J is the total flux of the ions. The Boltzmann equation for a single ion moving in the carrier plasma is linear in f and can be rewritten as $f = Kf$, with K being a linear operator. Then we find $J = (Kf_0) - J(Kf_1)$, where the parentheses stand for a linear operation producing J from f, $(f) = J$. Now $J = (Kf_0)/[1 + (Kf_1)]$. Section 2 formulates this general approach.

Thus the expression for J is a good starting point for any approximations. In this paper we use the free–molecule approximation for f_0 and f_1. The details are explained in Section 3.

The final expression for J derived in Section 4 is then used in Sections 5 and 6 for calculating the efficiencies of free condensation, the capture of polar molecules by charged particles, and the charging of small particles.

Because we focus on the dependence of J on the particle size, it is more convenient to rearrange the dimensionless groups mentioned above and to introduce

$$\tilde{a} = a/l = Kn^{-1} \quad \text{and} \quad \frac{qQe^2}{lkT} = \frac{l_c}{l}, \tag{1}$$

where $l_c = qQe^2/kT$ is the Coulomb length, a distance at which the energy of the Coulomb interaction becomes comparable to the ion thermal energy. At $T = 300$ K and $q = Q = 1$ the Coulomb length is comparable with the ion mean free path in the atmosphere at normal pressure. Indeed, $l_c = e^2/kT = 5.565 \times 10^{-6}$ cm $\approx l$.

2 Basic equations

In this section we discuss the general statement of the problem of the particle–ion interaction and the approximate approach.

The description of the ion transport toward a spherical particle in the transition regime requires the solution of the steady–state Boltzmann kinetic equation which is linear in the ion distribution $f(\boldsymbol{r}, \boldsymbol{v})$,

$$v_i \frac{\partial f}{\partial x_i} - \frac{1}{m}\frac{\partial U}{\partial x_i}\frac{\partial f}{\partial v_i} + \frac{f}{\tau_c} = R[f]. \tag{2}$$

Here $f(\boldsymbol{r}, \boldsymbol{v})$ is the ion distribution over coordinates and velocities, m is the ion mass, U is the potential of an external field, and $R[f]$ is the collision term (a linear functional of f). The collision time τ_c is defined as $1/\tau_c = R[1]$. The convention on the summation over repeating indexes is adopted.

In what follows we will consider only spherical particles. The potential U is then a function of $r = |\boldsymbol{r}|$ and the ion distribution depends only on three variables, the ion radial coordinate r, absolute ion velocity $v = |\boldsymbol{v}|$ and $\mu = \cos\theta$, with θ being the angle between the directions of $\boldsymbol{r}$ and $\boldsymbol{v}$.

In spherically symmetric systems another set of variables is more convenient. Namely, instead of r, v, μ we introduce r, E, L, with

$$E = mv^2/2 + U(r), \quad L = m|[\boldsymbol{v} \times \boldsymbol{r}]| = mvr\sqrt{1-\mu^2} \tag{3}$$

being the total ion energy and the ion angular momentum respectively. In these variables the Boltzmann equation takes the form:

$$sv_r \frac{\partial f_s}{\partial r} + \frac{f_s}{\tau_c} = R[f], \tag{4}$$

where

$$v_r = \sqrt{\frac{2}{m}\left(E - U(r) - \frac{L^2}{2mr^2}\right)} = \frac{1}{mr}\sqrt{L^2(r) - L^2} \tag{5}$$

is the radial ion velocity, $s = \pm 1$ is an auxiliary variable defining the direction of ion motion along the radial coordinate ($s = -1$ corresponds to the direction toward the particle), and

$$L(r) = \sqrt{2mr^2(E - U(r))}. \tag{6}$$

The ion flux toward the particle is expressed in terms of f as follows:

$$J = -\int \mathrm{d}^3v \int (v\,\mathrm{d}S) f(\boldsymbol{r}, \boldsymbol{v}). \tag{7}$$

The integrals on the right–hand side (rhs) of this equation are taken over all $\boldsymbol{v}$ and the surface of a sphere of radius r. The sign "$-$" in the definition of the flux makes J positive. In spherical coordinates equation (7) is rewritten as

$$J = -8\pi^2 r^2 \int_0^\infty v^3 \mathrm{d}v \int_{-1}^{1} f(r, v, \mu)\mu \mathrm{d}\mu. \tag{8}$$

The rule for replacing the variables $(r, v, \mu) \longrightarrow (r, E, L)$ readily follows from definition (3) of the variables E and L,

$$\mathrm{d}^3v \longrightarrow \frac{\pi}{m^2 r} \sum_s \frac{\mathrm{d}E\mathrm{d}L^2}{\sqrt{L^2(r) - L^2}}. \tag{9}$$

The restrictions on the intervals of integration over E and L^2 are defined by two conditions, $L^2 \leq L^2(r)$ and $L^2(r) \geq 0$. The latter one is equivalent to $E \geq U(r)$. In what follows we do not specify the limits of integrations except for the cases, where they play a decisive role.

The expression for the flux J in r, E, L variables looks as follows:

$$J = -\frac{4\pi^2}{m^3} \sum_s s \int \mathrm{d}E \int \mathrm{d}L^2 f_s(r, E, L). \tag{10}$$

Now we propose a very effective and transparent trick helping much in the analysis of many kinetic problems including the present one. The idea is simple. The Boltzmann equation (2) is just a linear equation of the form $\hat{\mathcal{L}} f = \hat{\mathcal{R}} f$, where $\hat{\mathcal{L}}$ is the differential operator on the left–hand side (lhs) of equation (2) and $\hat{\mathcal{R}}$ is a linear operator on its right–hand side (rhs). The formal solution to equation (2) is

$$f = \hat{\mathcal{L}}^{-1}\hat{\mathcal{R}} f. \tag{11}$$

Let now f_J be a solution to the Boltzmann equation with a *floating* (not yet fixed) flux J. We split f_J into two terms

$$f_J = f^{(0)} - \xi(a) f^{(1)},$$

where

$$\xi(a) = \frac{J}{J_{fm}}, \tag{12}$$

J_{fm} is the free–molecule ion flux, and the functions $f^{(i)}$ ($i = 0, 1$) are independent of J. The correction factor $\xi(a)$ defines the changes in the ion flux due to ion–molecule collisions. The ratio $\xi(a) = J/J_{fm}$ can be then found from equation (11) as

$$\xi(a) = \frac{A^{(0)}}{1 + A^{(1)}}, \tag{13}$$

where

$$A^{(i)} = \frac{4\pi^2}{m^3 J_{fm}} \sum_s \int \mathrm{d}E \int \mathrm{d}L^2 \hat{\mathcal{L}}^{-1}\hat{\mathcal{R}} f_s^{(i)}(a, E, L). \tag{14}$$

This very general expression is a good starting point for approximations: any reasonable approximate expression for f can be used.

We conclude this section by formulating the boundary condition to equation (4). We assume that no ions escape from the particle surface,

$$f_1(a, E, L) = 0. \tag{15}$$

Because the total flux J is independent of r, equation (10) can be rewritten as

$$J = \frac{4\pi^2}{m^3} \int \mathrm{d}E \int \mathrm{d}L^2 f_{-1}(a, E, L). \tag{16}$$

3 Approximations

In this section we discuss the form of the collision term and the approximate ion distribution function.

3.1 Collision term

In order to find the form of the functional $R[f]$ on the rhs of equations (2, 4) a two–body problem in the external field should be solved. The standard form of the collision term cannot be used, for the spatial scale of changing the external potential (of the order of a) is shorter than the ion mean free path. We therefore use the simplest possible form of the collision term,

$$R[f] = \frac{1}{\tau_c} n(r) M(r, E, L) Z(r), \tag{17}$$

where

$$n(r) = \frac{\pi}{m^2 r} \sum_s \int \mathrm{d}E \int \frac{\mathrm{d}L^2}{\sqrt{L^2(r) - L^2}} f_s(r, E, L) \tag{18}$$

is the ion density and

$$M(r, E, L) = \left(\frac{\beta m}{2\pi}\right)^{3/2} e^{-\beta(E - U(r))} \tag{19}$$

is the Maxwellian ($\beta = 1/kT$). The normalization factor $Z(r)$ takes into account the fact that the bound ion states are empty. We thus assume that the last collision that ion experiences right before attaching the particle surface takes place at a distance much exceeding the radius of external forces.

The normalization condition

$$\frac{Z(r)\pi}{m^2 r} \int_{\max(0, U(r))}^{\infty} \mathrm{d}E \int_0^{L^2(r)} \frac{\mathrm{d}L^2}{\sqrt{L^2(r) - L^2}} M(r, E, L) = 1 \tag{20}$$

defines the factor $Z(r)$,

$$Z(r) = \frac{\sqrt{\pi}}{2\Gamma(3/2, |\min(0, \beta U(r))|)}. \tag{21}$$

Here $\Gamma(\alpha, x) = \int_x^\infty s^{\alpha-1} e^{-s} \mathrm{d}s$ is the incomplete gamma–function.

Our next approximation concerns τ_c. We assume that $l = v\tau_c = \text{const}$ or

$$\tau_c \sqrt{\frac{2}{m}(E - U(r))} = \text{const}. \tag{22}$$

This approximation means that the ion mean free path l remains a constant rather than the collision time τ_c, as is commonly accepted [1,2]. In what follows we put $l = 1$, i.e., all distances are measured in units of l.

After these approximations the Boltzmann equation acquires the form:

$$s\sqrt{1 - \frac{L^2}{L^2(r)}} \frac{\partial f_s}{\partial r} + f_s = n(r) M(r, E, L) Z(r). \tag{23}$$

3.2 Free–molecule distribution

The steady–state free–molecule distribution satisfies the equation

$$s\sqrt{1 - \frac{L^2}{L^2(r)}} \frac{\partial f_s}{\partial r} = 0. \tag{24}$$

The most general form of the solution to equation (24) is

$$f_s(r, E, L) = B_s(E, L)\theta(L^2(r) - L^2) \tag{25}$$

with $\theta(x)$ being the Heaviside step–function. The function $B_s(E, L)$ is still arbitrary. Its dependence on E is defined by the condition that $f_s \propto e^{-\beta E}\theta(E)$ as $r \longrightarrow \infty$. The factor $\theta(E)$ excludes the bound states of ions, for they are not present far away from the particle. The dependence on L follows from the boundary condition (15),

$$B_s(E, L) = C e^{-\beta E}\theta(E)[\theta(L - L_a) + \xi(a)\delta_{s,-1}\theta(L_a - L)], \tag{26}$$

where C is the normalization constant,

$$C = n_\infty \left(\frac{\beta m}{2\pi}\right)^{3/2}, \tag{27}$$

$\delta_{i,k}$ is the Kronecker delta, and $\xi(a) = J/J_{fm}$ is the ratio of the total flux J to the free–molecule flux J_0 (see Eq. (12)),

$$J_{fm} = C\frac{4\pi^2}{m^3} \int L_a^2 e^{-\beta E} \mathrm{d}E. \tag{28}$$

This results follows from equation (16) on substitution of the distribution given by equations (25, 26) and putting $\xi(a) = 1$. The angular momentum L_a separates the ion trajectories crossing the particle surface ($L < L_a$) from those passing by the particle ($L > L_a$). It is easy to see that the distribution in this form gives the correct total ion flux (equal to J) and contains no outgoing ions at $r = a$. Indeed, the first term on the rhs of equation (26) is independent of s and thus does not contribute to the flux at all (see Eq. (10)). The integration of the second one removes J_0 from the denominator of $\xi(a)$. The factor $\theta(L - L_a)$ in the first term does not permit trajectories crossing the point $r = a$. Hence $f_1(a) = 0$.

4 Solution of kinetic equation

In order to find the flux we must first calculate the ion density profile, then solve equation (23) with the collision term found in the free–molecule approximation, and then calculate the integrals in equation (13). We will follow this plan.

4.1 Ion density profile

Equations (18, 25, 26) allow us to find the approximate ion density profile $n_a(r)$,

$$n_a(r) = n_\infty[2F(r) + \xi(a)G(r)], \tag{29}$$

where n_∞ is the ion density far away from the particle,

$$F(r) = \frac{\pi}{m^2 r}\left(\frac{\beta m}{2\pi}\right)^{3/2} \int_{E_0}^{\infty} e^{-\beta E}\mathrm{d}E \int_{L_a^2}^{L^2(r)} \frac{\mathrm{d}L^2}{\sqrt{L^2(r) - L^2}}, \tag{30}$$

and

$$G(r) = \frac{\pi}{m^2 r}\left(\frac{\beta m}{2\pi}\right)^{3/2} \int_{E_0}^{\infty} e^{-\beta E}\mathrm{d}E \int_{0}^{L_a^2} \frac{\mathrm{d}L^2}{\sqrt{L^2(r) - L^2}}. \tag{31}$$

Here the energy E_0 is defined by the condition $L_a^2 \geq 0$.

It is easy to find $P(r) = F(r) + G(r)$,

$$P(r) = \frac{e^{-\beta U(r)}}{2Z(r)}, \tag{32}$$

where $Z(r)$ is given by equation (21). The approximate ion density profile is thus

$$n_a(r) = n_\infty\left[2F(r) + \xi(a)(P(r) - F(r))\right]. \tag{33}$$

Let us find $F(r)$. The integration over L^2 in equation (30) gives

$$F(r) = \frac{1}{\sqrt{\pi}} \int_{\beta E_0}^{\infty} \mathrm{d}x\, e^{-x}\sqrt{x - \frac{a^2}{r^2}\Psi - \beta U(r)}. \tag{34}$$

In most practically important cases the function $L(r)$ reaches its minimum at $r_* > a$. The condition $\partial_r L_r^2 = 0$

rewritten as

$$r_*\frac{\partial U}{\partial r}\mid_{r=r_*}=\frac{L^2(r_*)}{mr_*^2} \tag{35}$$

defines its position as a function of a and E.

The factor Ψ depends on x and a via r_* and explicitly,

$$\Psi=\frac{r_*^2}{a^2}(x-\beta U(r_*))=\frac{\beta L_a^2}{2ma^2}. \tag{36}$$

The free–molecule flux J_{fm} is expressed in terms of Ψ as follows:

$$J_{fm}=\pi a^2 n_\infty v_T\int\limits_{x_0\theta(x_0)}^{\infty}e^{-x}\Psi\mathrm{d}x, \tag{37}$$

where $v_T=\sqrt{8kT/\pi m}$ is the thermal velocity and x_0 is the root of the equation $\Psi(x_0)=0$.

4.2 Ion distribution

According to the boundary condition to the kinetic equation (15) the ion flux J can be expressed in terms of $f_{-1}(a,E,L)$ alone, so here we solve equation (23) for the component f_{-1} with the approximate collision term containing the ion density profile from equation (33),

$$-\sqrt{1-\frac{L^2}{L^2(r)}}\frac{\partial f_{-1}}{\partial r}+f_{-1}=n_a(r)M(r)Z(r). \tag{38}$$

Instead of r we introduce the variable

$$\sigma(r)=\int\limits_a^r\frac{L(r')\mathrm{d}r'}{\sqrt{L^2(r')-L^2}}. \tag{39}$$

Then the solution to equation (38) is readily found,

$$f_{-1}(a,E,L)=\int\limits_a^\infty e^{-\sigma(r')}n_a(r')Z(r')M(r')\frac{L(r')\mathrm{d}r'}{\sqrt{L^2(r')-L^2}} \tag{40}$$

or, in a more concise form:

$$f_{-1}(a,E,L)=\int\limits_0^\infty e^{-\sigma}n_a(r(\sigma))Z(r(\sigma))M(r(\sigma))\mathrm{d}\sigma, \tag{41}$$

where $r(\sigma)$ is the solution of the equation $\sigma(r)=\sigma$.

4.3 Flux

Now we are ready to find the ion flux. Applying equation (16) to both sides of equation (40) gives $\xi(a)$ in the form of equation (13) with

$$A^{(0)}(a)=\frac{8\pi^2 n_\infty}{m^3 J_{fm}}\int\mathrm{d}E\mathrm{d}L^2 \times\int\limits_0^\infty e^{-\sigma}F(r(\sigma))M(r(\sigma))Z(r(\sigma))\mathrm{d}\sigma \tag{42}$$

and

$$A^{(1)}(a)=\frac{1}{2}A^{(0)}(a)-\frac{1}{2}. \tag{43}$$

In deriving equation (43) we used the identity

$$\frac{4\pi^2 n_\infty}{m^3 J_{fm}}\int\mathrm{d}E\mathrm{d}L^2\int\limits_0^\infty e^{-\sigma}P(r(\sigma))M(r(\sigma))\mathrm{d}\sigma=\frac{1}{2}. \tag{44}$$

In most cases of interest the integral appearing in equation (40) cannot be performed analytically. Still a very simple approximation is accessible. Its idea relies upon the fact that the term L^2 on the rhs of equation (40) can be ignored in the limit of small a. Then

$$r(\sigma)=\sigma+a. \tag{45}$$

Indeed, $\sigma\propto 1$ contributes to the integral in equation (39) whereas $L\propto L_a\ll L_r$.

Equation (45) simplifies considerably the expression for $Q(a)$. The integrations over L and E produce the multiplier J_0 and we have

$$A^{(0)}(a)=2\int\limits_0^\infty e^{-\sigma}F(\sigma+a)Z(\sigma+a)e^{\beta U\sigma+a)}\mathrm{d}\sigma. \tag{46}$$

In deriving this equation we use the fact that the product $e^{-\beta U}M\propto e^{-\beta E}$ (see Eq. (19)) is independent of r.

Now the problem of calculating the flux J reduces to the evaluation of the integral appearing in equation (42) or equation (46) for various potentials.

5 Nonsingular potentials

In this section we consider the molecular fluxes onto the particle surface assuming that the interaction potentials are nonsingular, i.e., they remain finite at $r=a$. Most widespread potentials are nonsingular, e.g., the Coulomb potential or the interaction of polar molecules with a charged particle. The common feature of these potentials is their monotonic behavior that ensures the minimum of $L(r)$ locates at $r=a$. The consideration of nonsingular potentials is much simpler than singular ones having a singularity at the particle surface. Examples of such potentials are also well–known, e.g., the image force potential which, as we will see below, plays an important role in considering the particle charging.

Two simplest examples illustrate our approach: the free condensation (no potential, $U=0$) of molecules onto the particle surface and the capture of polar molecules by charged particles.

5.1 Free condensation

Let us first consider the free condensation, $U(r)=0$. The free–molecule flux J_0 is readily found from equation (28),

$$J_{fm}=J_0=\pi a^2 v_T n_\infty. \tag{47}$$

Next, $L(r) = \sqrt{2mEr^2}$ and

$$\sigma(r) = \sqrt{2mE}\int\limits_a^r \frac{r'\mathrm{d}r'}{\sqrt{2mEr'^2 - L^2}} = \sqrt{r^2 - \frac{L^2}{2mE}} - \sqrt{a^2 - \frac{L^2}{2mE}}. \qquad (48)$$

It is easy to invert the function $\sigma(r)$,

$$r(\sigma) = \sqrt{\sigma^2 + 2\sigma a\mu + a^2}. \qquad (49)$$

Here we returned to the variable $\mu = \mu(L) = \sqrt{1 - L^2/L_a^2}$ (see Eq. (3)).

The factor $\Psi = x$ and $r_* = a$, so the function $F(r)$ has an especially simple form (see Eq. (34)):

$$F(r) = \frac{1}{2}\sqrt{1 - \frac{a^2}{r^2}}. \qquad (50)$$

Then,

$$F(r(\sigma)) = \frac{1}{2}\sqrt{\frac{\sigma^2 + 2\mu a\sigma}{\sigma^2 + 2\mu a\sigma + a^2}}. \qquad (51)$$

Equation (42) reduces to

$$A^{(0)}(a) = 2a\int\limits_0^1 \mu\mathrm{d}\mu \int\limits_0^\infty e^{-as}\sqrt{\frac{s^2 + 2\mu s}{s^2 + 2\mu s + 1}}\mathrm{d}s. \qquad (52)$$

Equations (13, 52) give the full solution to the problem. The approximation equation (43) simplifies the expression for $A^{(0)}$,

$$A^{(0)} = a\int\limits_0^\infty e^{-as}\frac{\sqrt{s^2 + 2s}}{s + 1}\mathrm{d}s. \qquad (53)$$

The correction factor $\xi(a) = J/J_0$ as the function of particle size a (measured in units of the ion mean free path l) is shown in Figure 1 (curve 1). Curves 1 and 1' reproduce the results of the present paper. In calculating curve 1' we used the approximation (45) for $r(\sigma)$. Curve 2 is the semi–empirical formula for the correction factor [22] that reproduces well the experimental data on condensation efficiencies for vapors of many substances. Even at sufficiently large particle sizes ($a = 0.5$) the deviation of the correction factor found in the free–molecule regime from the semi–empirical result is not very large.

5.2 Potential driven condensation

If the potential $U(r)$ has no singularity at $r = a$ and behaves monotonously, then the expressions for J_{fm} and $F(r)$ can be considerably simplified. In this case the angular momentum $L(r)$ reaches a minimum at the particle surface, $r_* = a$. The function Ψ reduces to $\Psi(x) = x - \beta U(a)$ and equation (37) reproduces the well–known results for the free–molecule flux,

$$J_{fm} = J_0 e^{-\beta U(a)} \qquad (54)$$

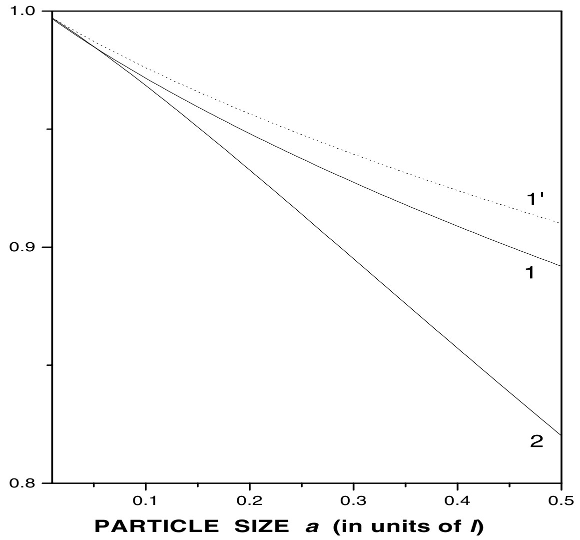

Fig. 1. Condensation of neutral molecules on a neutral particle. The nondimensional correction factor $\xi(a)$ is shown as the function of particle radius a measured in units of mean free path l. Curve 1: the correction factor in free–molecule limit (Eqs. (13, 52)). Curve 1' displays the same function found with the use of approximation (45). Curve 2 reproduces the semi–empirical size dependence of the correction factor from reference [22].

for repulsive potentials ($U(a) > 0$) and

$$J_{fm} = J_0[1 + \beta|U(a)|] \qquad (55)$$

for attractive potentials ($U(a) > 0$).

The function $F(r)$ is expressed in terms of the incomplete gamma–function $\Gamma(\alpha, x) = \int_x^\infty s^{\alpha-1}e^{-s}\mathrm{d}s$. Tedious but very straightforward algebra again gives different results for repulsive and attractive potentials.

For repulsive potentials we have,

$$F(r) = \frac{1}{\sqrt{\pi}}\sqrt{1 - \frac{a^2}{r^2}}\, e^{-\beta U(a)} \exp\left(\frac{\beta U(a) - \beta U(r)}{1 - a^2/r^2}\right) \times \Gamma\left(\frac{3}{2}, \frac{\beta U(a) - \beta U(r)}{1 - a^2/r^2}\right). \qquad (56)$$

For attractive potentials the result changes,

$$F(r) = \frac{1}{\sqrt{\pi}}\sqrt{1 - \frac{a^2}{r^2}} \exp\left(\frac{\beta|U(r)| - \beta|U(a)|(a^2/r^2)}{1 - a^2/r^2}\right) \times \Gamma\left(\frac{3}{2}, \frac{\beta|U(r)| - \beta|U(a)|(a^2/r^2)}{1 - a^2/r^2}\right). \qquad (57)$$

5.3 Condensation of polar molecules

The above theory can be readily applied to the calculations of the flux of polar molecules toward a charged particle. Although the interaction potential depends on the orientation of the polar molecule we can ignore this dependence and consider the dipoles directed toward the particle center. In this case the interaction potential is,

$$U(r) = -\frac{de^2}{r^2}, \tag{58}$$

where d is the distance between the opposite charges of the polar molecule. There are strong grounds for using equation (58). The point is that the dipole reorientation time τ_r is much shorter than the time of flight, the distance is of the order of the particle size $\tau_a \propto a/v_T$. Indeed, the dynamics of the dipole rotation is governed by the equation of motion $I\ddot{\phi} = \mathcal{F}$, where $I \propto md^2$ is the moment of inertia of the molecule, m is its mass and the rotating moment $\mathcal{F} \propto e^2d^2/a^3$. The angular acceleration $\ddot{\phi} \propto 1/\tau_r^2$. Hence, $\tau_r^2 \propto a^3m/e^2$ and $\tau_r^2/\tau_a^2 \propto amv_T^2/e^2 \propto akT/e^2 \propto a/l_c \ll 1$.

Next, it is pertinent to notice that the combination $[\beta|U(r)| - \beta|U(a)|(a^2/r^2)]/(1 - a^2/r^2)$ in equation (57) is identically zero for the potentials $U(r) \propto 1/r^2$. The function $F(r)$ (see Eq. (57)) in this case coincides with that for free condensation (Eq. (49)). The final result thus looks as follows:

$$J = \pi a^2 v_T n_\infty \left(1 + \frac{\beta e^2 d}{a^2}\right)\frac{2A^{(0)}(a)}{1 + A^{(0)}(a)}, \tag{59}$$

where the function $A^{(0)}(a)$ is given by equation (53).

6 Charging of particles

In order to use equation (42) for calculating the ion flux one first finds r_* and Ψ (see Eqs. (35, 36)). Then the function $F(r)$ found from equation (34) is substituted into equations (42) or (46). We use this scheme for calculating the rate of free condensation (no external potential) and the rate of charging a neutral or a charged particle.

6.1 General consideration

Let us consider the ion flux toward a charge particle. The incident ion interacts with the particle via the Coulomb and the image potentials,

$$U(r) = \pm\frac{qQe^2}{r} - \frac{q^2e^2}{2a}\frac{a^4}{r^2(r^2 - a^2)}, \tag{60}$$

where Q and q are the charges of the particle and the ion in units of e (Q and q are nonnegative integers, the upper sign (here "+") refers to the Coulomb repulsion). On introducing $\zeta = q/Q$ reduces equation (35) for r_* to the form:

$$\frac{q^2e^2}{2a}\left[\frac{a^4}{(r_*^2 - a^2)^2} \pm \frac{\zeta a}{r_*}\right] = E. \tag{61}$$

Although this equation cannot be resolved with respect to r_* analytically, it allows one to conclude that

$$r_* = a\rho(x/\gamma), \tag{62}$$

where we introduced $x = \beta E$, $\gamma = \beta q^2e^2/2a$. The function $\rho = \rho(u)$ satisfies the equation

$$u = \frac{1}{(\rho^2 - 1)^2} \pm \frac{\zeta}{\rho}. \tag{63}$$

It is easy to find that

$$\beta U(r_*) = \gamma\left(-\frac{1}{\rho^2(\rho^2 - 1)} \pm \frac{2\zeta}{\rho}\right) \tag{64}$$

and

$$\Psi = \gamma\rho^2\left[\frac{x}{\gamma} + \left(\frac{1}{\rho^2(\rho^2 - 1)} \mp \frac{2\zeta}{\rho}\right)\right]. \tag{65}$$

Because the function Ψ depends on the combination x/γ it is reasonable to introduce the variable $u = x/\gamma$ in the integrand of equation (37). This step reduces the expression for J_{fm} to

$$J_{fm} = J_0\gamma^2 \int\limits_{u_0\theta(u_0)}^{\infty} e^{-\gamma u}\psi(u)\mathrm{d}u, \tag{66}$$

where

$$\psi(u) = \rho^2(u)\left[u + \frac{1}{\rho^2(u)(\rho^2(u) - 1)} \mp \frac{2\zeta}{\rho(u)}\right] \tag{67}$$

and u_0 is the zero of $\psi(u)$.

The function $F(r)$ can be found from equations (34, 65) in the form $F(r) = \gamma^{3/2}\varphi(r/a, \gamma)$ with

$$\varphi(s,\gamma) = \frac{1}{\sqrt{\pi}}\int\limits_{u_0}^{\infty}\mathrm{d}u\, e^{-\gamma u}\sqrt{u + \frac{1}{s^2(s^2 - 1)} \mp \frac{2\zeta}{s} - \frac{\psi(u)}{s^2}}, \tag{68}$$

where u_0 is the positive zero of the function $\psi(u)$ or 0 if the function $\psi(u)$ has no positive zeros.

We conclude this paragraph by deriving a very useful identity. Let us differentiate both sides of equation (67) over u and apply equation (63). We get

$$\frac{\mathrm{d}\psi}{\mathrm{d}u} = \rho^2(u). \tag{69}$$

This remarkable identity helps much in simplifying the final expressions for J_{fm} and $F(r)$.

In what follows we apply the last five equations for finding J_{fm}, $F(r)$, and $A^{(0)}$ from equations (66, 34, 46). Our final goal is to restore the correction factor $\xi(a) = J/J_{fm}$,

$$\xi(a) = \frac{J}{J_{fm}} = \frac{2A^{(0)}}{1 + A^{(0)}}. \tag{70}$$

6.2 Charging of neutral particles

A neutral metallic particle interacts with ions via image forces whose potential is always attractive ($Q = 0$ and

$\zeta = 0$ in equations (61–65). Equation (63) is then solved analytically to give

$$\rho^2(u) = 1 + \frac{1}{\sqrt{u}} \tag{71}$$

and

$$\psi = u + 2\sqrt{u}. \tag{72}$$

Equations (66, 72) allow us to reproduce the well–known result for the free–molecule flux [21],

$$\begin{aligned} J_{fm} &= \pi a^2 v_T n_\infty \gamma^2 \int\limits_0^\infty e^{-\gamma u}\,(u + 2\sqrt{u})\mathrm{d}u \\ &= \pi a^2 v_T n_\infty \left(1 + \sqrt{\frac{\pi\beta q^2 e^2}{2a}}\right). \end{aligned} \tag{73}$$

The function $F(r)$ is readily found from equations (34, 72) in the form $F(r) = \gamma^{3/2}\varphi(r/a, \gamma)$ with

$$\varphi(s,\gamma) = \frac{1}{\sqrt{\pi}} \int \mathrm{d}u\, e^{-\gamma u} \sqrt{u + \frac{1}{s^2(s^2-1)} - \frac{u + 2\sqrt{u}}{s^2}}. \tag{74}$$

Equation (46) gives

$$A^{(0)}(a) = a\sqrt{\pi}\gamma^{3/2} \int\limits_0^\infty \frac{e^{-as}\varphi(s+1,\gamma)}{\Gamma(3/2, \nu(s))} e^{-\nu(s)}\mathrm{d}s, \tag{75}$$

where

$$\nu(s) = \frac{\gamma}{s(s+2)(s+1)^2}.$$

Now combining equations (13, 43) with the above two equations gives the ion flux. The result of calculations are presented in Figure 2.

6.3 Opposite polarities

In this case equation (63) has the form

$$u = \frac{1}{(\rho^2 - 1)^2} - \frac{\zeta}{\rho} \tag{76}$$

and cannot be solved analytically. It is important, however, that the function $u(\rho)$ has a zero at $\rho = \rho_\zeta$. As we will see this fact plays a very important role.

The function ψ has the form in this case

$$\psi(u) = \rho^2(u)\left[u + \frac{1}{\rho^2(u)(\rho^2(u) - 1)} + \frac{2\zeta}{\rho(u)}\right]. \tag{77}$$

Now, let us integrate by parts the rhs of equation (66) and introduce the variable ρ instead of u. Equation (69) allows then to simplify the expression for J_{fm},

$$\frac{J_{fm}}{J_0} = \gamma\psi(0) + \gamma\int_1^{\rho_0} e^{-\gamma u(\rho)}\left(\frac{4\rho^3}{(\rho^2-1)^3} - \zeta\right)\mathrm{d}\rho \tag{78}$$

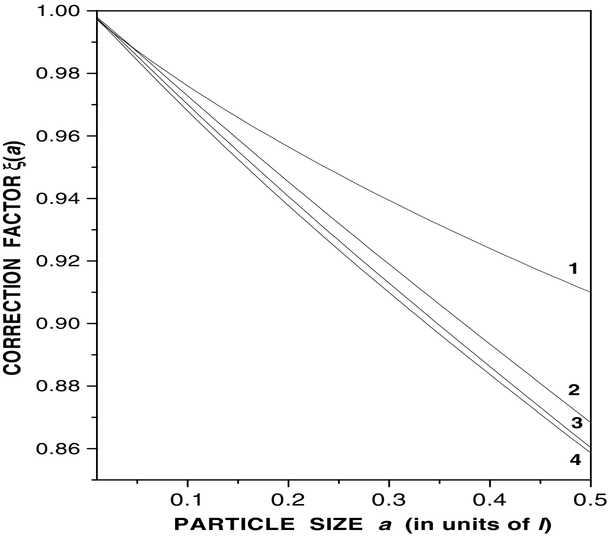

Fig. 2. Charging of a neutral particle. Shown is the nondimensional correction factor $\xi(a)$ as a function of particle size (in units of mean free path). Curve 1, $q = 0$ (neutral particle + neutral molecule), Curve 2–4 correspond to $q = 1$, 2, 3 respectively. It is seen that the intensity of the interaction does not strongly affect the curves.

Table 1. The coefficients of expansion of $J(a)$ (Eq. (79)) and the value of ρ_0 (Eq. (78)).

q	Q	a_1	a_0	a_{-1}	ρ_0
1	1	3.7996	2.2203	−2.1065	1.4901
1	2	6.6158	1.8214	−0.6241	1.3496
2	1	2.2288	2.8318	−7.3286	1.6828

where the function $u(\rho)$ is given by equation (76), ρ_0 is the root of the equation $u(\rho) = 0$. The values of ρ_0 and $\psi(0)$ depend on the ratio $\zeta = q/Q$.

At large γ the integral on the rhs of equation (66) can be easily estimated, for small $u \propto 1/\gamma$ contributes to the integral. We have

$$J_{fm} = \pi a^2 v_T n_\infty (a_1\gamma + a_0 + a_{-1}\gamma^{-1}\ldots). \tag{79}$$

The coefficients a_i and the values of the root of $u(\rho)$ (Eq. (76)) are collected in Table 1 for three combinations of charges $q = 1$ $Q = 1$, $q = 1$ $Q = 2$, $q = 2$ $Q = 1$. The results for $q = 2$ $Q = 2$ are identical to those for $q = 1$ $Q = 1$. The dependence of J_{fm} on q and Q enters also via γ (see the definition after Eq. (62)).

In calculating $Q(a)$ we use the approximation (45). Again, the function $F(r)$ can be found from equations (34, 64). Now the flux J is restored from equations (13, 46). The results of the numerical calculations are presented in Figure 3.

6.4 Similar polarities

The potential

$$U(r) = \frac{qQe^2}{r} - \frac{q^2e^2}{2a}\frac{a^4}{r^2(r^2-a^2)} \tag{80}$$

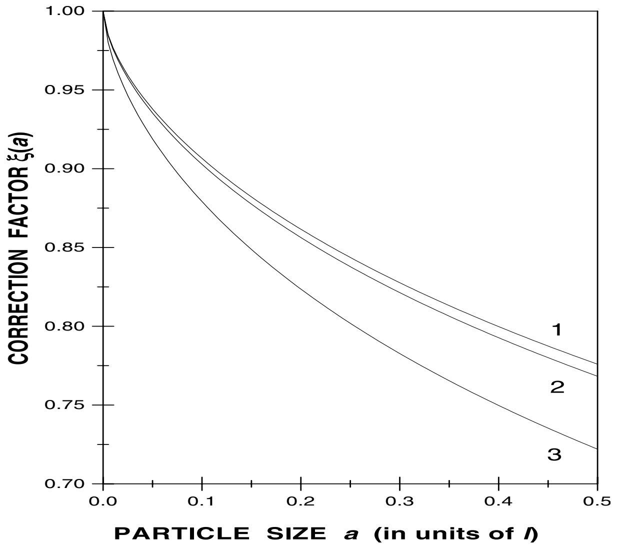

Fig. 3. Charging of a charged particle of charge Q by ions of opposite polarity (charge q). Shown is the nondimensional correction factor $\xi(a)$ as a function of particle size (in units of mean free path). Curves 1–3 refer to the charge combinations $q = 1$ $Q = 1$, $q = 1$ $Q = 2$, and $q = 2$ $Q = 1$. It is seen that the intensity of the interaction does not strongly affect the curves.

describes the interaction of like–charged ion and particle. It contains the Coulomb repulsion and the attraction due to the image forces.

For the potential equation (80), equations (63, 66, 67) look as follows:

$$u = \frac{1}{(\rho^2-1)^2} + \frac{\zeta}{\rho}. \tag{81}$$

The function $u(\rho)$ has no zeros

$$\psi(u) = \rho^2(u)\left[u + \frac{1}{\rho^2(u)(\rho^2(u)-1)} - \frac{2\zeta}{\rho(u)}\right] \tag{82}$$

and

$$J_{fm} = \pi a^2 n_\infty v_T \gamma^2 \int\limits_{u_0}^{\infty} e^{-\gamma u}\psi(u)\mathrm{d}u. \tag{83}$$

The interval of integration $u > u_0$ is determined by the condition $\psi(u) \geq 0$.

Differentiating both sides of equation (82) over u and using equation (81) again yields $\psi' = \rho^2$. On integrating by parts the rhs of equation (83) and introducing the variable ρ instead of u yields a more convenient expression for J_{fm},

$$\frac{J_{fm}}{J_0} = \gamma\int_1^{\rho_1} e^{-\gamma u(\rho)}\left(\zeta + \frac{4\rho^3}{(\rho^2-1)^3}\right)\mathrm{d}\rho, \tag{84}$$

where the function $u(\rho)$ is given by equation (81), ρ_1 is the root of the equation $\psi(u(\rho)) = 0$.

At large γ the integral on the rhs of equation (84) can be easily estimated, for small $u - u_0 \propto 1/\gamma \ll 1$ contributes to the integral.

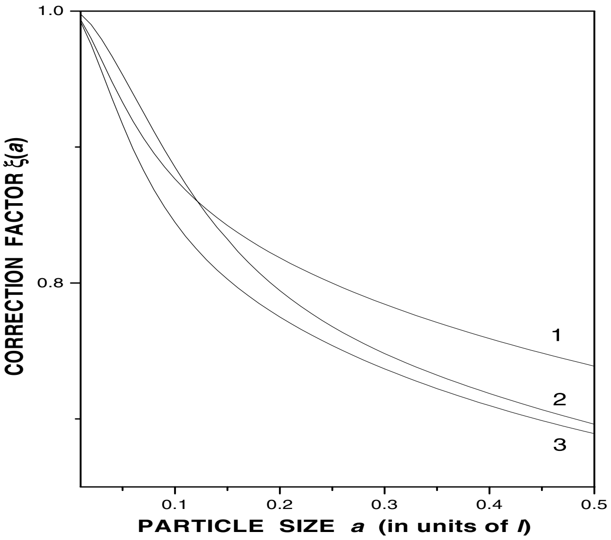

Fig. 4. Charging of a charged particle (charge Q) by ions of similar polarity (charge q). Flux of ions of charge q to the similarly charged particle of charge Q. Shown is the nondimensional correction factor $\xi(a)$ as a function of particle size (in units of mean free path). Curves 1–3 refer to the charge combinations $q = 1$ $Q = 1$, $q = 1$ $Q = 2$, and $q = 2$ $Q = 1$. It is seen that the intensity of the interaction does not strongly affect the curves.

Table 2. The coefficients of expansion of $J(a)$, the exponent u_0, (Eq. (85)), and the value of ρ_1 (Eq. (84)).

q	Q	b_0	b_{-1}	r_1	u_0
1	1	2.6165	−1.409	1.6179	1.001
1	2	2.0378	−0.4687	1.4275	2.3296
2	1	3.5426	−36813	1.8822	0.4203

As $u \longrightarrow u_0$ the function $\rho(u)$ approaches ρ_1 and

$$J_{fm} = \pi a^2 v_T n_\infty e^{-u_0\gamma}(b_0 - b_{-1}\gamma^{-1}\ldots). \tag{85}$$

The coefficients b_i and the values of ρ_1 and u_0 are presented in Table 2 for three combinations of charges $q = 1$ $Q = 1$, $q = 1$ $Q = 2$, $q = 2$ $Q = 1$. The results for $q = 2$ $Q = 2$ are identical to those for $q = 1$ $Q = 1$. The dependence of J_{fm} on q and Q enters also via γ (see the definition after Eq. (62)).

Figure 4 presents the results of the numerical calculations of the correction factor for a similarly charged particle and an ion.

6.5 Comparison with experiment

We compared the results of our calculations for the efficiency of particle charging with the experimental data in reference [29]. Figure 5 (solid line) clearly demonstrates that the first–order calculation does not give a satisfactory agreement with the experimental data. There could

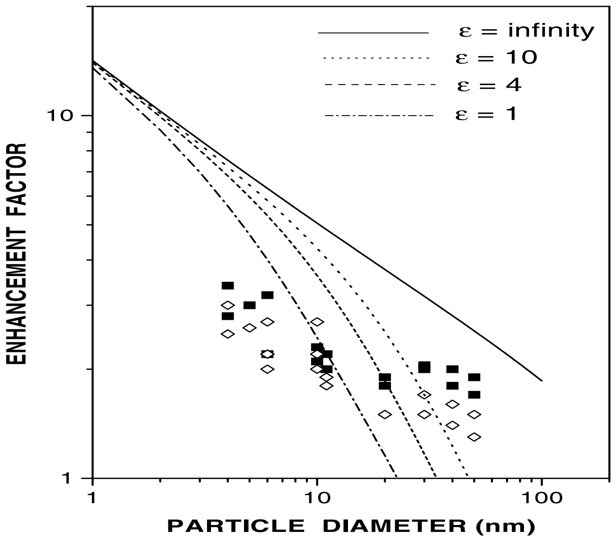

Fig. 5. Charging of a neutral particle. Shown is the enhancement factor (J/J_0) for charging neutral particles. The experimental data in reference [29] are compared with the result of our theoretical calculation presented in Section 6.3 (solid line). The other three lines demonstrate the effect of diminishing the ion concentration at distances $r = a + \varepsilon l$ of the order of the mean free path from the particle surface. Calculations are done for $\varepsilon = 10, 4, 1$.

be two reasons for this:

(i) the approximation already stops working at $Kn \approx 10$,
(ii) the dielectric permeability $\epsilon_d = 6$ is not high enough to reproduce the results for metallic particles (our theory works only at $\epsilon_d = \infty$).

Here we demonstrate explicitly that the first reason is responsible for the theory giving the excessive charging efficiencies and outline a route for improving the situation.

The ion flux onto the particle can always be written as

$$J = \alpha(a) n_\infty, \tag{86}$$

i.e., the flux is proportional to the ion density far away from the particle. If we calculate the efficiency $\alpha(a)$ approximately, let us say, $\alpha \approx \alpha_{fm}$, then n_∞ should be replaced by $n_\epsilon = n(a + \epsilon l)$, where $\epsilon \propto 1$ is a constant. However, the ion density at the distances from the particle surface of the order of mean free path can differ considerably from n_∞ due to the diffusion resistance of the carrier gas [20].

Let us rewrite equation (86) in the form

$$J = \alpha_{fm}(a) n_\epsilon. \tag{87}$$

Our next step assumes applying the diffusion equation for finding the ion density $n_\epsilon = n(a + \epsilon l)$. As $n(r)$ we use the solution to the diffusion equation with *fixed* flux J,

$$n^{(J)}(r) = e^{-\beta U(r)} \left(n_\infty - \frac{J}{4\pi D} \int\limits_r^\infty e^{\beta U(r')} \frac{\mathrm{d}r'}{r'^2} \right). \tag{88}$$

Then we obtain the equation for J,

$$J = \alpha_{fm} n^{(J)}(a + \epsilon l). \tag{89}$$

On solving this equation gives,

$$J = \alpha n_\infty \tag{90}$$

with

$$\frac{\alpha}{\alpha_{fm}} = \frac{e^{-\beta U(a+\epsilon l)}}{1 + \dfrac{\alpha_{fm} e^{-\beta U(a+\epsilon l)}}{4\pi D} \displaystyle\int\limits_{a+\epsilon l}^\infty e^{\beta U(r')} \frac{\mathrm{d}r'}{r'^2}}. \tag{91}$$

We can go even further and rewrite equation (89) in the form:

$$J = \alpha_{fm1} n^{(J)}(a + \varepsilon l), \tag{92}$$

where α_{fm1} is the charging efficiency found for the near free–molecule regime. In this case the value of $\varepsilon > \epsilon$ is expected to grow (compared to the free–molecule regime). This very version of equation (91) was used for calculating the curves plotted in Figure 5.

7 Discussion and conclusion

In this paper we have analyzed the kinetics of charging tiny particles suspended in a weakly ionized carrier plasma. Our approach relies upon a perturbation solution of the Boltzmann equation. Instead of attempting to construct an expansion of the total ion flux with respect to the smallness parameter (the reciprocal Knudsen number in our case) we use a self–consistency procedure for fixing the value of the flux. This step has allowed us to develop the perturbation theory separately for the numerator and denominator of the final exact expression for the flux (see Eq. (13)).

Another important step that has effectively simplified the solution of the Boltzmann equation and the derivation of the expressions for the free–molecule ion fluxes is the use of the set of variables (r, E, L^2) instead of the traditional one (r, v, μ). This very step has allowed us to construct the expression for the free–molecule distribution function (Eqs. (25, 26)) satisfying the boundary condition (15) and find a very simple and effective method for calculating the free–molecule fluxes and corrections due to ion–molecule collisions.

The final results (the ion fluxes) are presented in the form $J = J_{fm}\xi(a)$. We have expressed both multipliers on the rhs of this equation in terms of rather simple integrals (Eqs. (78, 84)). The analysis of the asymptotic behavior of these integrals has clearly shown the importance of the mirror forces. For example, in the case of similarly charged particles $q = Q = 1$ the exponent on the rhs of equation (85) is half of the value found from the consideration of the Coulomb repulsion alone.

The results of our calculation clearly demonstrate that the corrections due to ion–molecule collisions are almost independent of the interaction intensity. This effect is expressed especially clearly in the case of the interaction of polar molecules with charged particles, where the correction factor $\xi(a)$ is identical to that for free condensation.

The comparison of the above approach with the existing experimental data on particle charging in the near free–molecule regime shows that the first–order calculations are not enough to take into account the effect related to diminishing the ion concentration at distances of the order of the mean free path from the particle compared to the concentration of the ions far from the particle. Our simple model in Section 6.5 introduces this effect explicitly. The calculated charging efficiencies satisfactorily reproduce the existing experimental data.

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