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Charging of particles in a plasma

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Abstract. Several models that predict the charge of particles in a plasma are reviewed. The simplest is based on orbit-limited probe theory. This basic model can be improved by adding several effects: charge reduction at high dust densities, electron emission, ion trapping and fluctuations. The charge is reduced at high dust densities, when a significant fraction of the charge in the plasma resides on the particles, depleting the plasma. Electron emission due to electron impact or ultraviolet exposure can cause a particle to have a positive charge, which has useful implications for plasma processing, since particles are confined in a discharge only if they have a negative charge. Ion trapping occurs due to ion–neutral collisions within the attractive Debye sphere of a negatively charged particle. Trapped ions reduce the net electric force on a particle. A particle's charge fluctuates because the currents collected from the plasma consist of discrete charges arriving at the particle at random intervals. The root mean square fractional fluctuation level varies as $0.5\langle N\rangle^{-1/2}$ where $\langle N\rangle = \langle Q\rangle/e$ is the mean number of electron charges on the particle.

1. Introduction

A dust particle in a plasma gains an electric charge and responds to electric forces. The charge can range from zero to hundreds of thousands of electron charges, depending on the particle size and the plasma conditions. It arises from collecting electrons and ions from the plasma and sometimes from emitting electrons. In a plasma in which emission processes are unimportant, the equilibrium charge is negative because the flux of electrons to an uncharged surface is high relative to that of ions. On the other hand, when electron emission is significant, the equilibrium charge is positive.

A calculation of the charge on a particle is the starting point of every theory of dusty plasmas. Here I review the common 'orbit-limited' theory of charge collection, and then I present some effects that are often neglected in this model, but may have a significant impact on the particle's transport. These effects are: a reduction in charge due to high dust density, positive charging by electron emission, a reduction in electric forces due to ion trapping and charge fluctuations. These effects have been presented already in the literature; the purpose of this paper is to review them and to give practical formulae with illustrative examples.

2. The basic model: orbit-limited theory

Most dusty plasma charging theories are based on theories of electrostatic probes in plasmas. These theories predict the electron and ion currents to the probe. The currents are termed 'orbit-limited' when the condition $a \ll \lambda \ll \lambda_{\mathrm{mfp}}$ applies, where a is the particle radius, λ is the Debye length and λ_{mfp} is a collisional mean free path between neutral gas atoms and either electrons or ions [1, 2]. In that case, the currents are calculated by assuming that the electrons and ions are collected if their collisionless orbits intersect the probe's surface. It is assumed that the currents are infinitely divisible; that is, the discrete nature of the electronic charge is ignored. The latter assumption must be reversed to account for the fluctuations of the particle, as shown later.

For the collection of Maxwellian electrons and ions, characterized by temperatures T_e and T_i, the orbit-limited currents for an isolated spherical particle are [3]

$$\begin{aligned} I_e &= I_{0e}\exp(e\phi_s/kT_e) && \phi_s < 0 \\ I_e &= I_{0e}(1 + e\phi_s/kT_e) && \phi_s > 0 \\ I_i &= I_{0i}\exp(-z_i e\phi_s/kT_e) && \phi_s > 0 \\ I_i &= I_{0i}(1 - z_i e\phi_s/kT_e) && \phi_s < 0. \end{aligned} \tag{1}$$

Here ϕ_s is the surface potential of the particle relative to the plasma and z_i is the electronic charge of the ions. The coefficients I_{0e} and I_{0i} represent the current that is collected for $\phi_s = 0$, and are given by $I_{0\alpha} = n_\alpha q_\alpha (kT_\alpha/m_\alpha)^{1/2}\pi a^2 f_\alpha(w, v_{\mathrm{th}})$, where n_α is the number density of plasma species α. Here $f_\alpha(w, v_{\mathrm{th}})$ is a complicated function of the thermal velocity $v_{\mathrm{th}} = (2kT_\alpha/m_\alpha)^{1/2}$ and the drift velocity w between the plasma and the particles [3]. Simple expressions are available in the limiting cases of

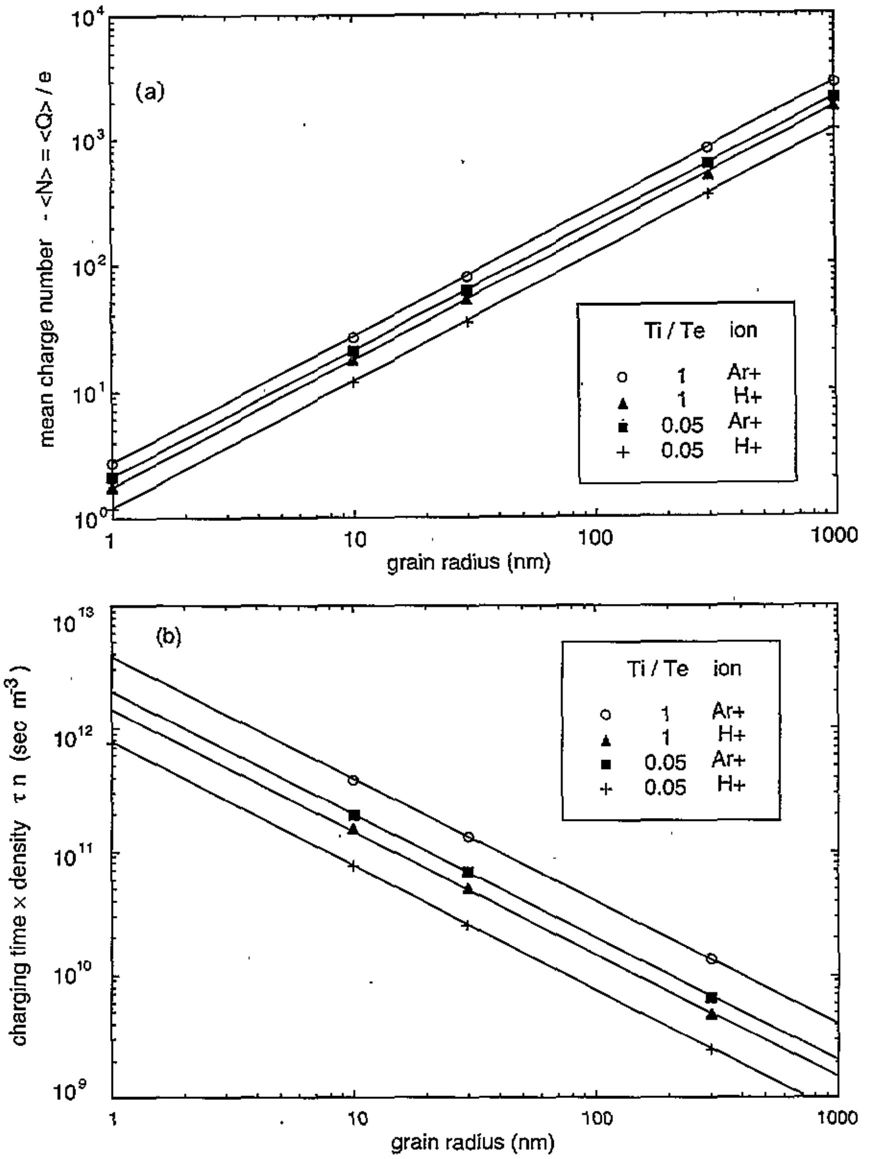

Figure 1. (*a*) Mean charge number $\langle N \rangle = \langle Q \rangle / e$ and (*b*) charging time τ as a function of particle radius for four combinations of temperature ratios and ion masses. These values are for the basic orbit-limited charging model, assuming non-drifting Maxwellians and no electron emission.

small and large drift velocities:

$$I_{0\alpha} = 4\pi a^2 n_\alpha q_\alpha (kT_\alpha / 2\pi m_\alpha)^{1/2} \qquad w/v_{th} \ll 1 \qquad (2a)$$

$$I_\alpha = \pi a^2 n_\alpha q_\alpha w[1 - 2q_\alpha \phi_s / (m_\alpha w^2)] \qquad w/v_{th} \gg 1. \qquad (2b)$$

In some laboratory discharges, the ions or electrons may drift at a significant speed. For example, the ions enter the electrode sheath at the ion acoustic speed, which is much faster than the ion thermal speed.

The charge Q is related to the particle's surface potential ϕ_s, with respect to a plasma potential of zero, by

$$Q = C\phi_s \qquad (3)$$

where C is the capacitance of the particle in the plasma. For a spherical particle satisfying $a \ll \lambda$, the capacitance is [3]

$$C = 4\pi\varepsilon_0 a. \qquad (4)$$

The standard 'continuous charging model' of particle charging in a plasma neglects the discrete nature of the electron's charge. The particle's charge is assumed to vary smoothly, rather than in integer increments. A particle with zero charge that is immersed in a plasma will gradually charge up, by collecting electron and ion currents, according to

$$\mathrm{d}Q/\mathrm{d}t = \sum I_\alpha. \qquad (5)$$

To find the equilibrium, one can set $\mathrm{d}Q/\mathrm{d}t = 0$ in equation (5). This yields the steady-state potential ϕ_f and steady-state charge $\langle Q \rangle$,

Table 1. Coefficients for ϕ_f, Q and τ appearing in equations (6) and (7). These values were found by a numerical solution of the continuous charging model, assuming non-drifting Maxwellians and no electron emission. From [5].

m_i (amu)	T_i/T_e	K_ϕ ($\mathrm{V\,eV^{-1}}$)	K_Q ($\mu\mathrm{m^{-1}\,eV^{-1}}$)	K_τ ($\mathrm{s}\,\mu\mathrm{m\,cm^{-3}\,eV^{-1/2}}$)
1	0.05	−1.698	−1179	7.66×10^2
1	1	−2.501	−1737	1.51×10^3
40	0.05	−2.989	−2073	2.05×10^3
40	1	−3.952	−2631	3.29×10^3

$$\phi_f = \langle \phi_s \rangle = K_\phi T_e$$
$$\langle Q \rangle / e = K_Q a k T_e \tag{6}$$

where the coefficients K_ϕ and K_Q are functions of T_i/T_e and m_i/m_e, and they must be determined numerically. Useful values for these coefficients are listed in table 1, and illustrative values of the charge are shown in figure 1(*a*). When electron emission is neglected, the floating potential and K_ϕ are both negative, since the electrons have higher thermal velocity than ions.

Note that ϕ_f is independent of the particle's size, but it depends on the plasma temperatures. On the other hand, the charge $\langle Q \rangle$ is proportional to the particle's radius, $\langle Q \rangle \propto a$. For example, a sphere in a hydrogen plasma with $T_e = T_i$ has the Spitzer [4] potential $\phi_f = -2.50\,kT_e/e$.

The charging time τ is inversely proportional to the plasma density. It depends on the particle size, temperature and ion mass according to [5]

$$\tau = K_\tau \frac{(kT_e)^{1/2}}{an} \tag{7}$$

where K_τ is a function of T_i/T_e and m_i/m_e. The fact that τ is inversely proportional to both a and n means that the fastest charging occurs for large particles and high plasma densities. One can define τ as the time required for a particle's charge to reach a fraction $(1 - e^{-1})$ of its equilibrium value, when it is initially uncharged. [5] Using this definition, the constant K_τ has the values summarized in table 1. Illustrative values of the charging time are shown in figure 1(*b*).

No dust particle is perfectly spherical, and so one should ask how much the sphericity assumption limits the theory's validity. This assumption appears twice in the model: the capacitance in equation (4) and the currents in equation (1). For the capacitance, the shape does not matter greatly as long as one chooses for a the typical size of the particle. The electron and ion currents are dominated by the shape of the electrostatic equipotential surfaces around the particle. The electric perturbation caused by the particle extends into the plasma a distance characterized by the shielding length, λ. Since the case treated here is $a \ll \lambda$, the equipotentials are distorted from a spherical shape only in a small central part of a spherical region of radius λ. Consequently, the sphericity assumption will introduce only a small error, as long as $a \ll \lambda$, as it is in most dusty plasmas.

3. Reduction of the charge due to high particle density

So far, I have considered the case of a single isolated particle, but this assumption is often unsuitable for modelling dusty laboratory plasmas, since they can have high particle concentrations. Several theorists have demonstrated that, as the dust number density is increased, the particle's floating potential and charge are reduced, due to electron depletion on the particles. [9] This electron depletion also modifies the plasma potential. The crucial parameter is Havnes's value P, which is basically the ratio of the charge density of the particles to that of the electrons. When $P > 1$, the charge and floating potential are significantly diminished, while for $P \ll 1$ the charge and floating potentials approach the values for an isolated particle (see section 2). In practical units, P is given by [6]

$$P = 695\, T_{eV} a_{\mu m} N_{cm^{-3}} / n_{cm^{-3}} \tag{8}$$

where N and n are the dust and electron number densities, respectively. This expression is written in a form for a mono-dispersive size distribution; a more general expression accounting for size dispersion is offered by Havnes *et al* [6].

Havnes *et al* [6] solved the charge balance equations, and reported useful analytic expressions for the particle's floating potential ϕ_f (referenced to the plasma potential) and the plasma potential ϕ_p (referenced to a dust-free plasma). These are functions of the parameter, P,

$$e\phi_f/kT = (K_\phi + a_1 P)/(1 + b_1 P + b_2 P^2)$$
$$e\phi_p/kT = (c_1 P + c_2 P^2)/(1 + d_1 P + d_2 P^2) \tag{9}$$

where the coefficients K_ϕ, b, c and d are listed in table 2.

Table 2. Coefficients for charge and plasma potential, assuming non-drifting Maxwellians with $T_e = T_i$, singly charged ions, and no electron emission. From [6].

Coefficient	Ion mass (amu) 1	32
K_ϕ	−2.5	−3.9
a_1	−0.764	−1.14
b_1	1.09	1.1
b_2	0.12	0.0754
c_1	−1.26	−1.98
c_2	−0.21	−0.252
d_1	1.04	1.17
d_2	0.112	0.0917

A representative plot of equation (9) is shown in figure 2.

In a RF discharge, the dust density is often high enough to attain $P \gg 1$. Consider for example the dust density measurements of Boufendi *et al* [7]. In a silane RF discharge, particles grew to a radius $a = 115$ nm, as determined by electron microscopy. Mie scattering indicated a particle density of $1 \times 10^8\,\mathrm{cm}^{-3}$, while the ion density was $5 \times 10^9\,\mathrm{cm}^{-3}$, based on ion saturation current measurements using a Langmuir probe. I assume $T_e = 2$ eV, which is probably accurate to within a factor of three. This yields $P = 3.2$ (accurate to within the same factor of three), corresponding to a 60% reduction in the particle's charge (according to equation (9) and figure 2).

4. Electron emission

Electrons can be emitted by the particle due to electron mpact, UV exposure, thermionic emission and field emission. The first two are probably the most important for laboratory dusty plasmas. Electron emission constitutes a positive current with respect to the particle, and, if it is large enough, it can cause the particle to be positively charged. Even if the particle is not always positive, it might sometimes fluctuate to a positive level, as described in sections 6 and 7.

4.1. Secondary electron emission

The secondary emission yield δ depends on both the impact energy E and particle material. The yield is generally much larger for electron impact than for ion impact. For bulk materials, the energy-dependence of the electron-impact yield is [8, 9]

$$\delta(E) = 7.4\delta_m (E/E_m) \exp[-2(E/E_m)^{1/2}]. \qquad (10)$$

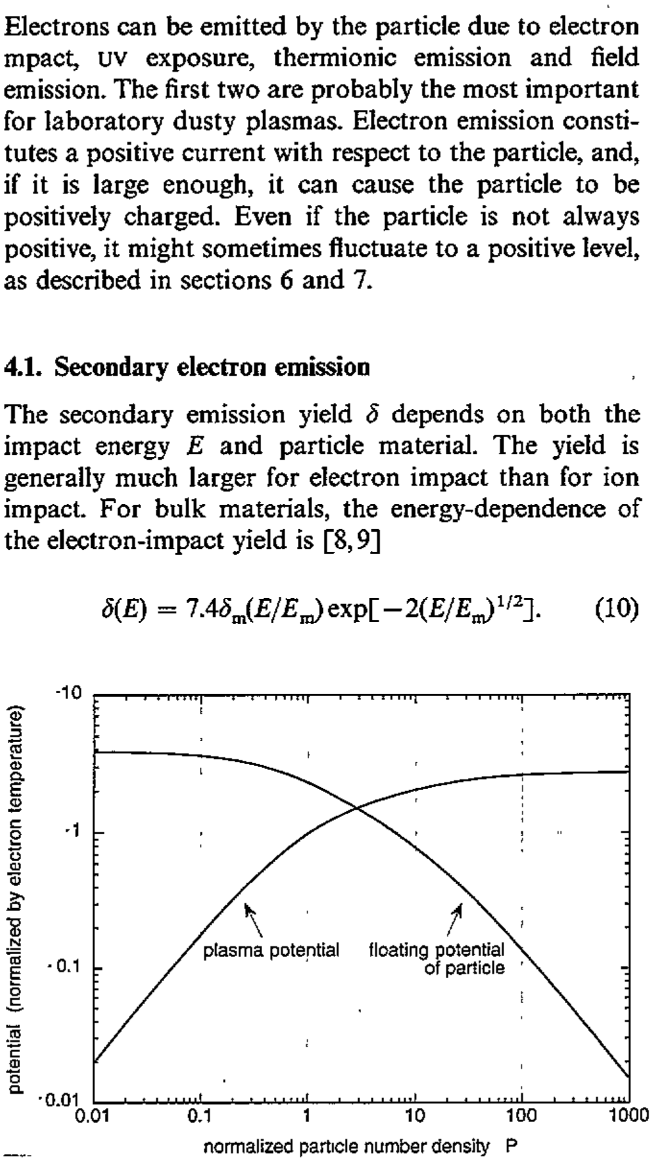

Figure 2. Dependence of floating potential $e\phi_f/kT$ and plasma potential $e\phi_p/kT$ on particle number density. These data are from equation (9) and table 2, for singly charged ions of mass 32 amu, with non-drifting Maxwellians and $T_e = T_i$. The normalized particle density is $P = 695\, T_{eV} a_{\mu m} N_{cm^{-3}}/n_{cm^{-3}}$.

The peak yield δ_m is at energy E_m, and both of these are material constants. Graphite, for example, has $\delta_m = 1$ and $E_m = 250$ eV, while for quartz $\delta_m = 2.1$–4 and $E_m = 400$ eV [8].

Secondary emission from small particles is significantly enhanced above the value for bulk materials. This was shown by Chow *et al* [10], whose theory included geometric effects. Scattered electrons escape more easily from a small particle than from a semi-infinite slab of material, and so δ is enhanced.

Expression (10) is for mono-energetic electrons of energy E. It must be remembered that electrons in a plasma have a distribution function. Assuming a Maxwellian primary electron distribution with temperature T_e, Meyer-Vernet [8] found the secondary currents I_{sec} due to an impinging electron current I_e,

$$\begin{aligned} I_{sec}/I_e &= 3.7\,\delta_m F_5(E_m/4kT_e) \qquad \phi_s < 0 \\ I_{sec}/I_e &= 3.7\,\delta_m \exp[(-e\phi_s/k)(T_s^{-1} - T_e^{-1})] \\ &\quad \times F_{5B}(E_m/4kT_e) \qquad \phi_s < 0 \end{aligned} \qquad (11)$$

where

$$F_5(x) = x^2 \int_0^\infty t^5 \exp[-(xt^2 + t)]\,dt$$

$$F_{5B}(x) = x^2 \int_B^\infty t^5 \exp[-xt^2 + t)]\,dt$$

$$B = [(e\phi_s/kT_e)(4kT_e/E_m)]^{1/2}$$

and T_s is the temperature of the emitted electrons, typically $1 < T_s < 5$ eV.

By including these currents into the charging balance the particle potential can become positive [8, 9]. For Maxwellian electrons, a switch in polarity occurs at an electron temperature of 1–10 eV, depending on δ_m. The reason this happens at temperatures well below the energy for peak emission E_m is the contribution of electrons in the tail of the distribution.

4.2. Photoelectric emission

Absorption of UV radiation releases photoelectrons and hence causes a positive charging current. Just like secondary electron emission, it can make the particle positively charged [9].

Electron emission depends on the material properties of the particle (its photoemission efficiency). It also depends on the particle's surface potential, because a positively charged particle can recapture a fraction of its photoelectrons. Taking this into account, the photoemission current is [9]

$$\begin{aligned} I_\nu &= 4\pi a^2 \Gamma_\nu \mu \qquad & \phi_s \leqslant 0 \\ I_\nu &= 4\pi a^2 \Gamma_\nu \mu \exp(-e\phi_s/kT_p) \qquad & \phi_s > 0. \end{aligned} \qquad (12)$$

Here Γ_ν is the UV flux and μ is the photomission efficiency ($\mu \approx 1$ for metals and $\mu \approx 0.1$ for dielectrics). Equation (12) assumes an isotropic source of UV and that the photoelectrons have a Maxwellian energy spectrum with a temperature T_p.

A laboratory plasma is a source of UV, due to electron-impact excitation of neutrals. However, there has been no analysis known to the author of whether this UV radiation can be strong enough to alter the charge significantly. In space plasmas, it is well known that dust and other objects often charge to positive polarity due to UV exposure.

5. Ion trapping

A particle's negative charge creates a Debye sheath, which is an attractive potential well for positive ions. A passing ion can become trapped in this well when it suffers a collision within the particle's Debye sphere, simultaneously losing energy and changing its orbital angular momentum. It remains trapped there, in an orbit bound to the particle, until it is detrapped by another collision [11].

Trapped ions are important because they shield the charged particle from external electric fields. Since these fields provide the particle's levitation and confinement in the discharge, shielding must be modelled in order to understand confinement. This shielding works the same way as in an atom, where orbital electrons screen the charge of the nucleus. The effectiveness will vary with the number of trapped ions.

Untrapped ions do nothing to screen the particle's charge from an electric field. The author believes there has been some confusion in the literature over how Debye shielding works. Untrapped ions do contribute to reducing the force applied *by* the particle on other distant charges, but they do not reduce the force applied *to* the particle by an electric field. Only trapped ions can do that.

Ion trapping has been ignored often in dusty plasma theories, probably because it is not easy to deal with analytically. At least two numerical methods [11, 12] have been reported recently. The methods are useful for estimating the number of trapped ions, N_{trap}. However, this value has been reported for only a limited number of conditions, to date.

Both methods involve simulating ion motion in the field of the charged particle by integrating the equation of motion. They also both include collisions. In a code with a fixed time step that is short compared with the mean time between collisions, this is done typically by using a Monte Carlo method. The collision probability $1 - \exp(-\Delta t|v|/\lambda_{mfp})$ is evaluated at each time step and compared with a random number between 0 and 1 to determine whether a collision took place during that time step.

Choi and Kushner [12] developed a three-dimensional particle-in-cell (PIC) code, where all the ion orbits were tracked in a simulation box. Electrons and ions are absorbed by the particle, allowing dynamic simulation of the particle's charge and the surrounding electrostatic potential. Ions are subject to collisions, and those that are trapped are counted. This number of trapped ions fluctuates in time, as individual ions became trapped and then lost.

Goree [11] handled the incoming flux of incoming ions, one at a time, as individual test particles. The number of trapped ions at steady state is computed by assuming a balance between collisional trapping and detrapping. Assuming that ion–neutral collisions are dominant, this theory predicts that N_{trap} is independent of the mean free path, and increases with the plasma ion density. The model was implemented as a Monte Carlo code for many test ions selected from the incoming ion flux. If an ion becomes trapped, its orbit is followed until it is eventually scattered into an untrapped trajectory by further collisions. This code showed that $N_{trap} \gg 1$ when the ion density is $\gg 10^6\,\mathrm{cm}^{-3}$, which it always is in plasma processing discharges, indicating that ion trapping will cause significant electrical screening. These results were for $a = 10\,\mu\mathrm{m}$ and a plasma with $\lambda_D = 100\,\mu\mathrm{m}$ and an ion-neutral mean free path much shorter than λ_D. A limitation of this theory is that is uses a prescribed electrostatic potential. This makes the simulation valid only when the number of trapped ions is small, $N_{trap} \ll Q/e$. In principle it could be extended to compute the potential self-consistently for the actual electron and ion densities, as is done in the simulation by Choi and Kushner [12].

6. Charge fluctuations

The standard continuous charging model described in section 2 neglects the fact that the electron and ion currents collected by the particle actually consist of individual electrons and ions. The charge on the particle is an integer multiple of the electron charge, $Q = Ne$, where N changes by -1 when an electron is collected and by z_i when an ion is absorbed. Electrons and ions arrive at the particle's surface at random times, like shot noise. The charge on a particle will fluctuate in discrete steps (and at random times) about the steady-state value $\langle Q \rangle$.

Several models have been reported recently to predict the fluctuation level. Choi and Kushner's PIC simulation [12] yielded a time series for the charge of an isolated particle, for a particular set of parameters. The charge clearly fluctuated about a mean value. Tsytovitch [13] developed an analytic theory that is unique because it was not for an isolated particle, but rather for a cloud of particles in the plasma. Taking into account how the fluctuation of the charge on one particle affects the charge on a neighbour, he found that the fluctuation level increases with particle number density.

Cui and Goree [5] used a numerical method, where the problem for an isolated particle was cast in terms of a probability per unit time of collecting an electron or ion from the plasma. To do this, they first converted the current I_α of the continuous charging model into a probability per unit time $(\mathrm{d}P/\mathrm{d}t)_\alpha$ of collecting an ion or electron, by $(\mathrm{d}P/\mathrm{d}t)_\alpha = I_\alpha/q_\alpha$. This probability depends

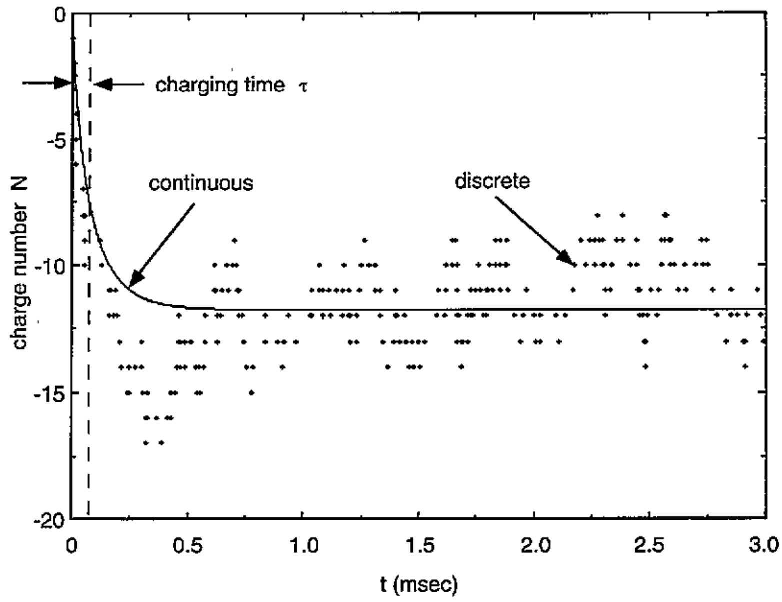

Figure 3. Temporal evolution of charge number $N = Q/e$, for a small particle ($a = 10\,\mathrm{nm}$) in an H^+ plasma with $T_i/T_e = 0.05$ and $n = 10^{15}\,\mathrm{m}^{-3}$. When discrete electronic charges are taken into account, fluctuations of the particle's charge are apparent, due to electrons and ions arriving at random times. From [5].

in a realistic way on the particle's potential; when the particle becomes more negative, it becomes less likely to collect an electron, for example. This probability is used with a random number generator to determine the times when an individual electron or ion is collected, and te charge is advanced by -1 or $+z_i$, accordingly.

Cui and Goree's simulation [5] begins with a particle that is initially uncharged, and it is allowed to continue for a long time after reaching a teady state. Figure 3 shows the early part of the time series for the particle's charge. The charge builds up from zero toward an equilibrium charge $\langle Q/e \rangle = \langle N \rangle$. The continuous model gives a smooth curve for $Q(t)$, while the discrete model reveals the discrete nature of Q, with random time steps and fluctuations about the smooth curve from the continuous model.

The fractional fluctuation is strongest for smallest particles. It obeys $\Delta Q/\langle Q \rangle = 0.5\langle Q/e \rangle^{-1/2}$, for a wide range of plasma and particle parameters. The square-root scaling is the same as in counting statistics, where the fractional uncertainty of a count N is $N^{-1/2}$. The power spectrum of the fluctuations is dominated by very low frequencies, with half the spectral power lying at frequencies below $0.024\tau^{-1}$. Here τ is the charging time, as defined in equation (7). At higher frequencies, the spectral power diminishes as the second power of frequency, f^{-2}.

7. Contamination control

Here I suggest a speculative idea for contamination control during plasma processing, based on an understanding of the charging processes described in this paper.

Contamination might be controlled by inducing a positive charge on the particles. A positive charge is important because it is believed that only negatively charged particles are confined in a laboratory discharge. A discharge (in the absence of significant egative io density) has a natural electric potential that tends to confine negatively charged particles. By promoting electron emission, the particles will charge positively and be expelled from the plasma. They will either strike the electrode or escape radially from the discharge.

In the case of secondary emission, electron emission can be promoted by heating the electrons, perhaps by operating with a low gas pressure or using an electron-heating source such as microwave power. For photoemission, one could deliberately illuminate the plasma with a UV source. It may be useful to know that the particle's charge can fluctuate to a positive value even if it is not possible to charge it positively all of the time.

To be effective, a contamination control method must either prevent growth of particles to a harmful size or transport them away from the substrate. The technique proposed here could serve the first purpose, and perhaps the second. Some particles might be forced to land on the substrate by promoting a positive charge. This would be acceptable if it happens while they are still nano-particles, which are too small to cause a defect. Provided that the source of UV or electron heating is applied constantly, or pulsed rapidly, any particle that begins growing will be expelled from the plasma before it has time to grow to a harmful size.

Acknowledgments

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