Surface States and the Charge of a Dust Particle in a Plasma

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We investigate electron and ion surface states of a negatively charged dust particle in a gas discharge and identify the charge of the particle with the electron surface density bound in the polarization-induced short-range part of the particle potential. On that scale, ions do not affect the charge. They are trapped in the shallow states of the Coulomb tail of the potential and act only as screening charges. Using orbital-motion limited electron charging fluxes and the particle temperature as an adjustable parameter, we obtain excellent agreement with experimental data.

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Motivation.—The calculation of the charge of a macroscopic object in an ionized gas is one of the most fundamental problems of plasma physics. It occurs in space-bound plasmas, where the charge of satellites is of interest [1], in astrophysical plasmas, where one wants to know the charge of interstellar grains [2], and in laboratory gas discharges, where charged dust particles are either contaminants, which need to be controlled, or constituents, whose collective properties are the subject of study [3,4].

For laboratory plasmas, the particle charge has been measured in a number of experiments [5–9]. Throughout it is thereby assumed that the particle's surface potential, and hence its charge, is the one which balances at the grain surface the total electron with the total ion charging flux: $j_e = j_i$. In almost all cases, however, the charges obtained from this condition, which is equivalent to forcing the net charge of the particle to be quasistationary, are too high. Usually, the approximations for the fluxes, mostly orbital-motion limited (OML) [10–12], are blamed for the disagreement and various modifications of the OML theory have been proposed. Although this leads sometimes to reasonable quantitative results [5,6], we suspect on fundamental grounds that irrespective of the fluxes $j_e = j_i$ is not the condition which fixes the charge (or potential) of the particle.

The condition $j_e = j_i$ is part of the Boltzmann-Poisson description of the plasma-particle interaction. Its natural length scale is thus the length on which the Coulomb potential varies. There are however microscopic processes near the surface of the particle, most notable sticking and desorption of electrons, which affect the charge but take place on a much shorter length scale. Once these processes are incorporated it is clear that the charge of the particle is not determined by the quasistationarity of the net charge but by the individual quasistationarity of the electron and ion densities bound to the particle. This condition implies the former but not vice versa. It is thus more restrictive and leads to lower charges.

In this Letter we describe a surface model, which accounts for plasma- and surface-induced processes, and calculate the charge of the particle, and its partial screening due to trapped ions, without relying on the condition $j_e = j_i$. Instead, we force the electron and ion densities bound to the particle to be quasistationary by balancing, individually on effective surfaces, electron and ion charging fluxes with electron and ion desorption fluxes.

Surface states.—We start with an investigation of the bound states in the *static* interaction potential of an electron (ion) and a dust particle with radius R, dielectric constant ϵ, and charge $-eZ_p$. The potential contains a polarization-induced part, arising from the electric boundary condition at the grain surface, and a Coulomb tail due to the particle's charge [13]. Defining $\xi = (\epsilon - 1)/2(\epsilon + 1)Z_p$, measuring distances from the grain surface in units of R and energies in units of $\bar{U} = Z_p e^2/R$, the interaction energy at $x = r/R - 1 > x_b$, where x_b is a cutoff below which the grain surface is not perfect anymore, reads

$$V_{e,i}(x) = \pm\frac{1}{1+x} - \frac{\xi}{x(1+x)^2(2+x)} \approx \begin{cases} 1 - \xi/2x & \text{electron} \\ -1/(1+x) & \text{ion.} \end{cases} \quad (1)$$

The second line approximates the relevant parts of the interaction energy very well and permits an analytical calculation of surface states. A gas discharge usually contains enough electrons which can overcome the particle's Coulomb barrier $\bar{U} \sim O(\text{eV})$. These are the electrons which may get bound in the polarization-induced short-range part of the potential, well described by the above approximate expression. Ions, on the other hand, having a finite radius $r_i^{\text{size}}/R = x_i^{\text{size}} \approx 10^{-4}$, cannot explore the potential at these distances. The long-range Coulomb tail is most relevant to them, which is again well described by the approximate expression.

In order to determine bound states from the Schrödinger equations corresponding to $V_{e,i}(x)$ we have to specify boundary conditions. Clearly, the wave functions $u_{e,i}(x)$ have to vanish for $x \to \infty$, irrespective of the potentials. The boundary condition at x_b, in contrast, depends on the

potential for $x \leq x_b$, that is, on the surface barrier. For our purpose, it is sufficient to take the simplest barrier model: $V_{e,i}(x \leq x_b) = \infty$ with $x_b = 0$ for electrons and $x_b = x_i^{\text{size}}$ for ions. The wave functions vanish then also at x_b and the surface states are basically rescaled hydrogen-type wave functions.

The left panel of Fig. 1 shows s-type electron and ion probability densities $|u_{e,i}(x)|^2$ (our reasoning does not depend on the angular momentum) for a melamine-formaldehyde (MF) particle ($R = 4.7$ μm, $\epsilon = 8$, and $Z_p = 6800$) in a helium discharge with plasma density $n_e \approx n_i = 0.62 \times 10^9$ cm^{-3}, ion temperature $k_B T_i = 0.04$ eV, and electron temperature $k_B T_e = 2.2$ eV [14]. The Rydberg series of electron surface states is only a few Angstroms away from the grain boundary. At these distances, the spatial variation of $V_e(x)$ is comparable to the de Broglie wavelength of electrons approaching the particle: $\lambda_e^{dB}/R \sim |V_e/V_e'| \sim 10^{-4}$. Hence, the trapping of electrons at the surface of the particle is a quantum-mechanical effect not included in the classical description of the plasma-particle interaction. For ions, on the other hand, the lowest surface states, which carry quantum-mechanical features, are unimportant. Being cold and heavy, ions will be bound in a continuum of states below the ion ionization threshold which consists essentially of classical trapped orbits, as can be seen from the $|u_i(x)|^2$ for the state labeled by $k(20\,000)$. That ions behave classically is not unexpected. Their de Broglie wavelength is much smaller then the scale on which the potential varies: $\lambda_i^{dB}/R \sim 10^{-5} \ll |V_i/V_i'| \sim 1$ for $x \gtrsim 10^{-3}$.

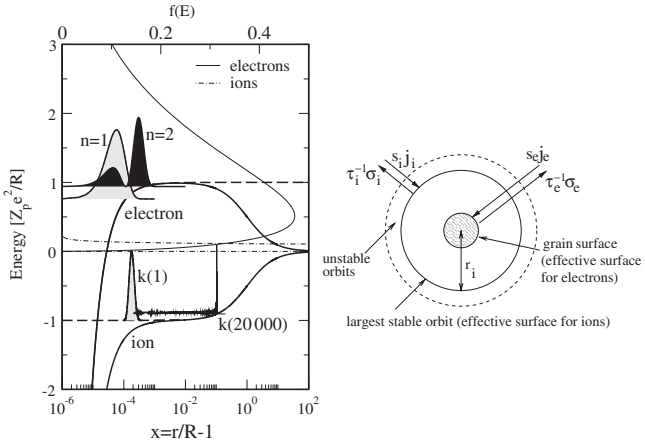

FIG. 1. Left panel: Potential energy for an electron (ion) in the field of a MF particle ($R = 4.7$ μm, $Z = 6800$) [14] and s-type probability distributions shifted to the binding energy and maxima normalized to one. Dashed lines are the potentials used in the Schrödinger equations and thin lines are the bulk energy distribution functions for the hosting discharge. Right panel: Illustration of the surface model to be discussed in the main text. At quasistationarity, surface charges $\sigma_{e,i}$ bound at $r_e \approx R$ and $r_i \approx (2\pi\sigma_{\text{cx}} n_g)^{-1}$, respectively, balance charging fluxes $s_{e,i} j_{e,i}$ with desorption fluxes $\tau_{e,i}^{-1}\sigma_{e,i}$.

Model.—We now use the properties of the surface states to construct a model for the charge of the particle. Within the sheath of the particle, the density of free electrons (ions) is much smaller than the density of bound electrons (ions). In that region, the quasistationary charge (in units of $-e$) is thus approximately

$$Z(x) = 4\pi R^3 \int_{x_b}^{x} dx'(1 + x')^2 [n_e^b(x') - n_i^b(x')] \quad (2)$$

with $x < \lambda_i^D = \sqrt{kT_i/en_i}$, the ion Debye length, which we take as an upper cutoff, and $n_{e,i}^b$ the density of bound electrons and ions. The results presented above suggest to express the density of bound electrons by an electron surface density: $n_e^b(x) \approx \sigma_e \delta(x - x_e)/R$ with $x_e \sim x_b \sim 0$ and σ_e the quasistationary solution of

$$d\sigma_e/dt = s_e j_e - \tau_e^{-1}\sigma_e, \quad (3)$$

where j_e is the electron charging flux from the plasma and s_e and τ_e are, respectively, the electron sticking coefficient and electron desorption time due to inelastic collisions between electrons and the particle (see right panel in Fig. 1). We will argue below that once the particle has collected some negative charge, not necessarily the quasistationary one, there is a critical ion orbit at $x_i \sim 1$–$10 \gg x_e$ which prevents ions from hitting the particle surface. Thus, the particle charge is simply

$$Z_p \equiv Z(x_e < x < x_i) = 4\pi R^2 (s\tau)_e j_e. \quad (4)$$

For an electron to get stuck at (to desorb from) a surface it has to loose (gain) energy at (from) the surface. Since electrons with rather low and rather high energies are, respectively, reflected by the Coulomb and surface barrier of the particle, sticking (desorption) primarily affects electrons at energies slightly above $\bar{U}$. Assuming this group of electrons after overcoming the Coulomb barrier to be in quasiequilibrium with the surface electrons, absolute reaction rate [15] allows us to estimate

$$(s\tau)_e \approx \frac{h}{k_B T_p} \exp\left[\frac{E_e^d}{k_B T_p}\right], \quad (5)$$

where h is Planck's constant, T_p is the particle temperature, and E_e^d is the negative of the binding energy of the surface state from which desorption most likely occurs. This phenomenological equation relates a combination of kinetic coefficients, which individually depend on the dynamic interaction, to an energy which can be deduced from the static interaction alone. To go beyond Eq. (5) necessitates a quantum-kinetic treatment of the inelastic electron-particle interaction.

Equation (4) is a self-consistency equation for Z_p. More explicitly, combined with Eq. (5), and using the OML electron charging flux, which is a reasonable approximation because, on the plasma scale, electrons are repelled from the particle, it reads

$$Z_p = 4\pi R^2 \frac{h}{k_B T_p} e^{E_e^d/k_B T_p} j_e^{\mathrm{OML}}(Z_p) \quad (6)$$

with $j_e^{\mathrm{OML}} = n_e\sqrt{k_B T_e/2\pi m_e}\exp[-Z_p e^2/Rk_B T_e]$. Thus, Z_p depends on the radius R, the plasma parameters n_e and T_e, and the surface parameters T_p and E_e^d.

Results.—To estimate E_e^d we imagine an electron with energy just above $\bar{U}$ approaching the grain. By necessity, it comes very close to the surface (see left panel in Fig. 1). For any realistic surface barrier, the wave function will therefore leak into the grain and the electron will strongly couple to the excitations of the grain which provide the thermal reservoir encoded in T_p. Hence, the electron will quickly relax to the lowest surface state. The $n = 1$ state for the infinitely high barrier is an approximation to that state. Thus, $E_e^d \approx R_0(\epsilon - 1)^2/16(\epsilon + 1)^2$, where R_0 is the Rydberg energy. T_p cannot be determined as simply. It depends on the heating and cooling fluxes to and from the grain and thus on additional surface parameters [16]. We use T_p therefore as an adjustable parameter. To reproduce, for instance, with Eq. (6) the charge of the particle in Fig. 1, $T_p = 395$ K implying $E_e^d \approx 0.51$ eV and $(s\tau)_e \approx 0.4 \times 10^{-6}$ s.

In Fig. 2 we analyze within our approach the pressure dependence of the charge of a MF particle with $R = 1$ μm in the bulk of the neon discharge of Ref. [5]. Since the plasma parameters entering Eq. (6) are known [5], T_p is again the only free parameter. Fixing T_p at a particular value gives the isothermal particle charges $Z_p(T_p)$. From $Z_p(T_p) = Z_{\mathrm{exp}}$ follows then the T_p required to reproduce the data. The predicted increase of T_p with pressure is realistic. Indeed, assuming $T_p \simeq T_g$, with T_g the gas temperature, T_p is in accordance with what one would expect from the pressure dependence of T_g in noble gases [16]. For comparison we also plot the particle charges deduced from $j_e^{\mathrm{OML}} = j_i^{\mathrm{OML}} + j_i^{\mathrm{CX}}$ with $j_i^{\mathrm{OML}} = n_i\sqrt{k_B T_i/2\pi m_i}[1 + Z_p e^2/Rk_B T_i]$ the OML ion charging flux and $j_i^{\mathrm{CX}} = n_i(0.1\lambda_i^D/l_{\mathrm{cx}})\sqrt{k_B T_i/2\pi m_i}(Z_p e^2/Rk_B T_i)^2$ the ion flux due to charge-exchange (CX), where $l_{\mathrm{cx}} = (\sigma_{\mathrm{cx}} n_g)^{-1}$ is the scattering length, σ_{cx} is the cross section, and $n_g = p/k_B T_g$ is the gas density [5]. Although for $\sigma_{\mathrm{cx}} = 2 \times 10^{-14}$ cm^2 (which may be unrealistically large [17]) the agreement with the data is good, the radius dependence of Z_p at fixed pressure shown in the inset indicates that something must be wrong with the flux balance criterion. The data clearly appear to be closer to the nonlinear R dependence obtained from the surface model than to the linear one resulting from $j_e^{\mathrm{OML}} = j_i^{\mathrm{OML}} + j_i^{\mathrm{CX}}$.

Figure 3, showing the R dependence of Z_p for MF particles confined in the sheath of an argon discharge [7], provides additional support for our model. To approximately account for the fact that particles with different radius experience different plasma environments, we included the depletion of n_e in the sheath by replacing n_e in j_e^{OML} by $n_e \exp[e\Phi(z_{\mathrm{eq}}(R))/k_B T_e]$ with $\Phi(z)$ the sheath potential and $z_{\mathrm{eq}}(R)$ the equilibrium position of the particle with radius R [7]. When the grains are not too deep in the sheath ($R < 5$ μm), we find excellent agreement with the data for $T_p = 420$ K. Our approach fails, however, for $R > 5$ μm (see inset). We attribute this to the ad-hoc description of j_e which may not capture the total electron charging flux close to the electrode.

Equation (6) depends on the assumption that once the particle is negatively charged ions are trapped far away from the grain surface. Indeed, a recent study based on the

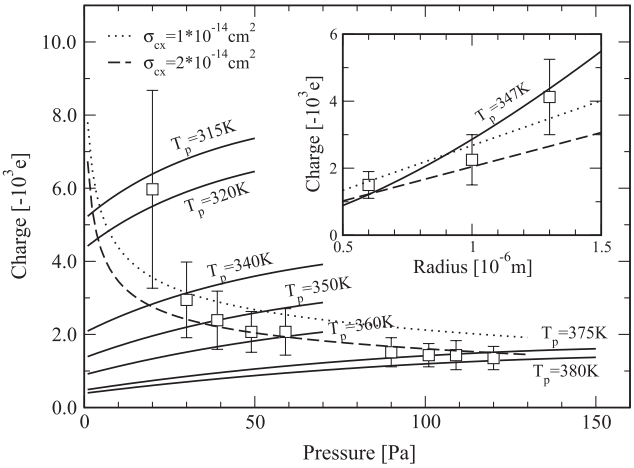

FIG. 2. Pressure dependence of the charge of a MF particle with $R = 1$ μm in the neon discharge of Ref. [5] (squares). Solid lines denote the (isothermal) charges deduced from the surface model whereas dotted and dashed lines are the charges obtained from $j_e^{\mathrm{OML}} = j_i^{\mathrm{OML}} + j_i^{\mathrm{CX}}$. The inset shows the radius dependence of the charge for $p = 50$ Pa.

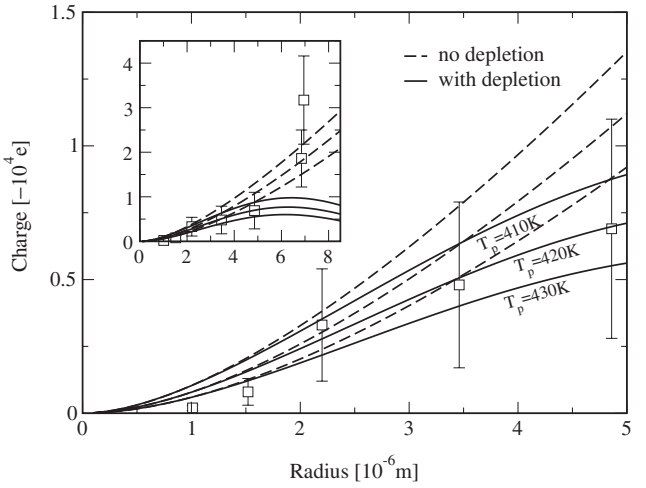

FIG. 3. Radius dependence of the charge of a MF particle in the sheath of an argon discharge at $p = 6.67$ Pa [7] (squares). Solid (dashed) lines give the charges deduced from the surface model when the depletion of n_e in the confining sheath is (is not) included in the OML electron charging flux.

Boltzmann-Poisson equations has shown that charge-exchange collisions lead to a local pileup of ions in the sheath of the particle [18,19]. We come to the same conclusion from the surface physics point of view. Similar to an electron, an ion gets bound to the grain only when it looses energy. Because of the long-range attractive ion-grain interaction, the ion will be initially bound far away from the grain surface (see left panel in Fig. 1). The coupling to the excitations of the grain is thus negligible and only inelastic processes due to the plasma are able to induce transitions to lower bound states. Since the interaction is classical, inelastic collisions, for instance, charge-exchange between ions and atoms, act like a random force. Energy relaxation can be thus envisaged as a destabilization of orbits whose spatial extension is comparable to or larger then the scattering length. Smaller orbits are unaffected because the collision probability during one revolution is vanishingly small. For a circular orbit, a rough estimate for the critical radius is $r_i = R(1 + x_i) = (2\pi\sigma_{\mathrm{cx}} n_g)^{-1}$, which leads to $x_i \sim 5 \gg x_e \sim 0$ when we use the parameters of the helium discharge of Fig. 1 and $\sigma_{\mathrm{cx}} = 0.32 \times 10^{-14}$ cm^2 [20]. Thus, there is a relaxation bottleneck at x_i and ions are trapped deep in the sheath of the particle.

To determine the partial screening due to trapped ions we model the ion density n_i^b accumulating in the vicinity of the critical orbit by a surface density σ_i which balances at x_i the ion charging flux with the ion desorption flux (see right panel in Fig. 1). Mathematically, this gives rise to a rate equation similar to (3) but now for the ions. At quasistationarity, the ion surface density is thus $\sigma_i = (s\tau)_i j_i$. Although Eq. (5) assumes excitations of the grain to be responsible for sticking and desorption we expect a similar expression (with E_e^d, T_p replaced by E_i^d, T_g) to control the density of trapped ions. From Eq. (2) we then obtain $Z(x_i < x < \lambda_i^D) = Z_p - Z_i$ with

$$Z_i = 4\pi R^2 (1 + x_i)^2 \frac{h}{k_B T_g} e^{E_i^d(Z_p)/k_B T_g} j_i^B \qquad (7)$$

the number of trapped ions. Since the critical orbit is near the particle-sheath-plasma boundary, it is fed by the Bohm ion flux $j_i^B = 0.6 n_i \sqrt{k_B T_e / m_i}$. The ion desorption energy is the negative of the binding energy of the critical orbit, $E_i^d(Z_p) = -V_i(x_i)\bar{U}(Z_p) = 4\pi\sigma_{\mathrm{cx}} a_B n_g Z_p R_0$, and depends strongly on Z_p and x_i. For the situation shown in Fig. 1 we obtain $E_i^d \approx 0.37$ eV and $(s\tau)_i \approx 0.6 \times 10^{-8}$ s when we use $T_g = T_p = 395$ K, the particle temperature which reproduces $Z_p \approx 6800$. The ion screening charge is then $Z_i \approx 148 \ll Z_p$ which is the order of magnitude expected from molecular dynamics simulations [21]. Thus, even when the particle charge is defined by $Z(x_i < x < \lambda_i^D)$ it is basically given by Z_p.

Summary.—We constructed a surface model to calculate the charge (partial screening) of a particle in a plasma by balancing, on an effective surface, the electron (ion) charging with the electron (ion) desorption flux. The number of electrons bound in the polarization potential determines the charge of the particle. Using the grain temperature as an adjustable parameter we obtained far better agreement with measurements, in particular, with respect to the radius dependence of the charge, then approaches based on balancing at the grain surface the total charging fluxes which we argue is the wrong condition. It neglects the microscopic processes determining the charge of the particle: sticking and desorption of electrons at the grain surface.

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