Surface states and the charge of a dust particle in a plasma

F. X. Bronold1, H. Felske1, H. Kersten2, and H. Deutsch1

1 Institut für Physik, Ernst-Moritz-Arndt-Universität Greifswald, D-17489 Greifswald, Germany
2 Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, D-24098 Kiel, Germany

(Dated: May 15, 2008)

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.

PACS numbers: 52.27.Lw, 52.40.Hf, 73.20.-r, 68.43.-h

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 spacebound 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, 6, 7, 8, 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 quasi-stationary, are too high. Usually, the approximations for the fluxes, mostly orbital motion limited (OML) [10, 11, 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 scale. Once these processes are incorporated it is clear that the charge of the particle is not determined by the quasi-stationarity of the net charge but by the individual quasi-stationarity 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 quasi-stationary 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 \( \epsilon \), 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 \( Z_p \). Defining \( \xi = (\epsilon - 1)/2(\epsilon + 1)Z_p \), measuring distances from the grain surface in units of \( R \) and energies in units of \( U_p = Z_p^2/2R \), the interaction energy at \( x = r/R - 1 > x_b \), where \( x_b \) is a cut-off below which the grain surface is not perfect anymore, reads

\[
V_{e,i}(x) = \pm \frac{1}{1 + x} - \frac{\xi}{(1 + x)^2(2 + x)} \approx \begin{cases} 1 - \xi/2x & \text{electron} \\ -1/(1 + x) & \text{ion}. \end{cases}
\] (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 \( U \sim o(eV) \). These are the electrons which may get bound in the polarization-induced short-range part of the potential, well described by the approximate expression. Ions, on the other hand, having a finite radius \( r_{i\text{size}}^i/R = x_{i\text{size}}^i \approx 10^{-4} \), 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 wavefunctions \( u_{e,i}(x) \) have to vanish for \( x \to \infty \), irrespective of...
FIG. 1: Left panel: Potential energy for an electron (ion) in the field of a MF particle \((R = 4.7 \, \mu m, Z = 6800)\) 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 quasi-stationarity, surface charges \(\sigma_{e,i}\) bound at \(x_s \approx R\) and \(x_r \approx (2\pi \sigma_e n_e)^{-1}\), respectively, balance charging fluxes \(s_{e,i,j} = \sigma_{e,i}\) with desorption fluxes \(\tau^{-1}_{e,i}\).

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 \((R = 4.7 \, \mu m, \epsilon = 8,\) and \(Z_p = 6800)\) in a helium discharge with plasma density \(n_e \approx n_i = 0.62 \times 10^{10} \, cm^{-3}\), ion temperature \(k_B T_i = 0.04 \, eV\), and electron temperature \(k_B T_e = 2.2 \, eV\) \([13]\). 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: \(\lambda_{e,R}^B \sim |V_e|/V_e| \sim 10^{-4}\). 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 \(k(20000)\) state. That ions behave classically is not unexpected. Their \(s\)-de-Broglie wavelength is much smaller than the scale on which the potential varies: \(\lambda_{e,R}^B \sim 10^{-5} \ll |V_i|/V_i| \sim 1\) for \(x \gtrsim 10^{-3}\).

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 quasi-stationary charge (in units of \(-e\) is thus approximately

\[
Z(x) = 4\pi R^3 \int_{x_s}^{x} dx' (1 + x')^2 \left[ n_b^b(x') - n_i^b(x') \right] \tag{2}
\]

with \(x < \lambda_R^B = \sqrt{kT_i/e}\), the ion Debye length, which we take as an upper cut-off, and \(n_e^b\) the density of bound electrons and ions. The results presented above suggest to express the density of bound electrons by an electron density surface density: \(n_e^b(x) \approx \sigma_e \delta(x - x_e)/R\) with \(x_e \approx x_b \approx 0\) and \(\sigma_e\) the quasi-stationary solution of

\[
d\sigma_e/dt = s_e j_e - \tau_{e}^{-1} \sigma_e , \tag{3}
\]

where \(j_e\) is the electron charging flux from the plasma and \(s_e\) and \(\tau_e\) 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 quasi-stationary 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

\[
Z_p \equiv Z(x_e < x < x_i) = 4\pi R^2 (s\tau)_e j_e . \tag{4}
\]

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 \(\tilde{U}\). Assuming this group of electrons to be in quasi-equilibrium with the surface electrons after overcoming the Coulomb barrier, absolute reaction rate theory \([13]\) allows us to estimate

\[
(s\tau)_e \approx \frac{h}{k_B T_p} \exp \left[ \frac{E_{e}^d}{k_B T_p} \right] , \tag{5}
\]

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. (4) 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

\[ Z_p = 4\pi R^2 \frac{h}{k_B T_p} e^{E_{eOML}/k_B T_p} j_e^{OML}(Z_p) \]  

with \( j_e^{OML} = n_e \sqrt{k_B T_e/2 \pi m_e} \exp[-Z_p e^2/R k_B T_e]. \)

Thus, \( Z_p \) depends on the radius \( R \), the plasma parameters \( n_e \) and \( T_e \), and the surface parameters \( T_p \) and \( E_{e}^{OML}. \)

**Results.**—To estimate \( E_{e}^{OML} \) we imagine an electron with energy just above \( U \) approaching the grain. By necessity, it comes very close to the surface (see left panel in Fig. 1). For any realistic surface barrier, the wavefunction 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, \( E_{e}^{OML} \approx R_0 (\epsilon - 1)/16(\epsilon + 1)^2, \)

where \( R_0 \) is the Rydberg energy. \( T_p \) cannot be determined as simply. It depends on the heating and cooling fluxes to-and-fro 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 \( E_{e}^{OML} \approx 0.51 \) eV and \( (n) \approx 0.4 \times 10^{-6} \) s.

In Fig. 2 we analyze within our approach the pressure dependence of the charge of a MF particle with \( R = 1 \) \( \mu \)m in the bulk of the neon discharge of Ref. \( [2]. \) Since the plasma parameters entering Eq. (6) are known \( [3], T_p \) is again the only free parameter. Fixing \( T_p \) at a particular value gives the isothermal particle charges \( Z_p(T_p). \) From \( Z_p(T_p) = Z_{\text{exp}} \) follows then the \( T_p \) required to reproduce the data. The predicted increase of \( T_p \) with pressure is realistic. Indeed, assuming \( T_p \sim T_g, \) 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 \( j_e^{OML} = j_i^{OML} + j_i^{CX} \) with \( j_i^{OML} = n_i \sqrt{k_B T_i/2 \pi m_i}[1 + Z_p e^2/R k_B T_i] \) the OML ion charging flux and \( j_i^{CX} = n_i (0.1 \lambda_{D}^2/l_{cx}) \sqrt{k_B T_i/2 \pi m_i} (Z_p e^2/R k_B T_i)^2 \) the ion flux due to charge-exchange (CX), where \( l_{cx} = (\sigma_{cx} n_g)^{-1} \) is the scattering length, \( \sigma_{cx} \) is the cross section, and \( n_g = p/k_B T_g \) is the gas density \([6]. \)

Although for \( \sigma_{cx} \approx 2 \times 10^{-14} \) 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 non-linear \( R \) dependence obtained from the surface model than to the linear one resulting from \( j_e^{OML} = j_i^{OML} + j_i^{CX}. \)

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 \( n_{e0} \exp[\Phi(z_{\text{eq}}(R))/k_B T_e] \) with \( \Phi(z) \) the sheath potential and \( z_{\text{eq}}(R) \) the equilibrium position of the particle with radius \( R \) \([7]\). When the grains are not too deep in the sheath \((R < 5 \) \( \mu \)m), we find excellent agreement with the data for \( T_p \approx 420 \) K. Our approach fails, however, for \( R > 5 \) \( \mu \)m (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.
we then obtain $Z(x_i < x < \lambda_B^i) = Z_p - Z_i$ with

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

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, $E_i^B(Z_p) = -V_i(x_i)U(Z_p) = 4\pi \sigma_{x_0} a g n_y Z_p R_0$, and depends strongly on $Z_p$ and $x_i$. For the situation shown in Fig. 1 we obtain $E_i^B \approx 0.37$ eV and $(st)_i \approx 0.6 \times 10^{-8}$ 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 $Z_i \approx 148 < Z_p$, which is the order of magnitude expected from molecular dynamics simulations [21]. Thus, even when the particle charge is defined by $Z(x_i < x < \lambda_B^i)$ 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.

Support from the SFB-TR 24 is greatly acknowledged. F. X. B. is funded by MV 0770/461.01.

[1] E. C. Whipple, Rep. Prog. Phys. 44, 1197 (1981).
[2] M. Horány, Annu. Rev. Astron. Astrophys. 34, 383 (1996).
[3] O. Ishihara, J. Phys. D: Appl. Phys. 40, R121 (2007).
[4] V. E. Fortov, A. V. Ivlev, S. A. Khrapak, A. G. Khrapak, and G. E. Morfill, Phys. Rep. 421, 1 (2005).
[5] S. A. Khrapak, S. V. Ratynskaja, A. V. Zobnin, A. D. Ushachev, V. V. Yaroshenko, M. H. Thoma, M. Kretschmer, H. Hoefnag, G. E. Morfill, O. F. Petrov, et al., Phys. Rev. E 72, 016406 (2005).
[6] A. A. Samarian and S. V. Vladimirov, Phys. Rev. E 67, 066404 (2003).
[7] E. B. Tomme, B. M. Annaratone, and J. E. Allen, Plasma Sources Sci. Technol. 9, 87 (2000).
[8] E. B. Tomme, D. A. Law, B. M. Annaratone, and J. E. Allen, Phys. Rev. Lett. 85, 2518 (2000).
[9] B. Walch, M. Horány, and S. Robertson, Phys. Rev. Lett. 75, 838 (1995).
[10] R. V. Kennedy and J. E. Allen, J. Plasma Phys. 69, 485 (2003).
[11] J. E. Allen, B. M. Annaratone, and U. de Angelis, J. Plasma Phys. 63, 299 (2000).
[12] I. B. Bernstein and I. N. Rabinowitz, Phys. Fluids 2, 112 (1959).
[13] C. F. Boettcher, Theory of electric polarization (Elsevier Publishing Company, Amsterdam, 1952).
[14] H. Kersten, H. Deutsch, and G. M. Kroesen, Int. J. Mass Spectrometry 233, 51 (2004).
[15] H. J. Kreuzer and Z. W. Gortel, Physassorption Kinetics (Springer Verlag, Berlin, 1986), pp. 13.
[16] G. H. P. M. Swinkels, H. Kersten, H. Deutsch, and G. M. W. Kroesen, J. Appl. Phys. 88, 1747 (2000).
[17] R. Hegerberg, M. T. Elford, and H. R. Skullerud, J. Phys. B 15, 797 (1982).
[18] M. Lampe, R. Goswami, Z. Sternovsky, S. Robertson, V. Gavrishchaka, G. Ganguli, and G. Joyce, Phys. Plasma 10, 1500 (2003).
[19] M. Lampe, V. Gavrishchaka, G. Ganguli, and G. Joyce, Phys. Rev. Lett. 86, 5275 (2001).
[20] B. L. Moiseiwitsch, Proc. Phys. Soc. London A 69, 653 (1956).
[21] S. J. Choi and M. J. Kushner, IEEE Trans. Plasma Science 22, 138 (1994).