Experimental Determination of Dust-Particle Charge in a Discharge Plasma at Elevated Pressures

S. Ratynskaia,¹ S. Khrapak,¹ A. Zobnin,² M. H. Thoma,¹ M. Kretschmer,¹ A. Usachev,² V. Yaroshenko,¹ R. A. Quinn,¹ G. E. Morfill,¹ O. Petrov,² and V. Fortov²

¹Centre for Interdisciplinary Plasma Science, Max-Planck-Institut für extraterrestrische Physik, D-85741 Garching, Germany ²Institute for High Energy Densities, Russian Academy of Sciences, Izhorskaya 13/19, 125412, Moscow, Russia

(Received 12 March 2004; published 17 August 2004)

The charge of dust particles is determined experimentally in a bulk dc discharge plasma in the pressure range 20–100 Pa. The charge is obtained by two independent methods: one based on an analysis of the particle motion in a stable particle flow and another on an analysis of the transition of the flow to an unstable regime. Molecular-dynamics simulations of the particle charging for conditions similar to those of the experiment are also performed. The results of both experimental methods and the simulations demonstrate good agreement. The charge obtained is several times smaller than predicted by the collisionless orbital motion theory, and thus the results serve as an experimental indication that ion-neutral collisions significantly affect particle charging.

DOI: 10.1103/PhysRevLett.93.085001

PACS numbers: 52.27.Lw, 52.20.-j, 52.35.-g

There is currently considerable interest in understanding the properties of dusty (complex) plasmas—plasmas containing charged micron-size particles (grains). This interest was originally driven by astrophysical topics [1] and industrial plasma applications [2]. It is also recognized that complex plasmas open up the possibility to study a variety of phenomena (e.g., phase transitions, transport, waves, etc.) at the most elementary kinetic level [3,4].

The particle charge is one of the most important parameters of complex plasmas. In gas discharges the (negative) charge on a particle is determined by the balance of electron and ion fluxes to its surface. To calculate these fluxes the collisionless orbital motion limited (OML) theory [5] is typically used on the basis that the electron and ion mean free paths $l_{e(i)}$ are long compared to the plasma screening length $\lambda_{\rm D}$. However, theory has shown that ion-neutral charge exchange collisions in the vicinity of a small probe or dust grain can lead to a substantial increase in the ion current to their surfaces [5–9]. It has been demonstrated that the ion collisions can suppress the particle charge even when l_i is considerably greater than $\lambda_{\rm D}$. Another effect for charge reduction is that of "closely packed" grains [10].

So far most of the experimental particle charge determinations were performed in sheath or striation regions of discharges [11–15]. The comparison with theory is complicated here due to the strong plasmas anisotropy and non-neutrality, the presence of "suprathermal" ions and electrons, etc. In addition to the charging model one needs to choose an appropriate model for the sheath, which is itself a sophisticated task. Hence, there is clearly a lack of direct measurements of particle charge in bulk plasmas. In this Letter, we present experimental results on the dust-particle charge in a plasma at elevated neutral gas pressures. The experiment is performed with particles of radius $a = 0.6 \ \mu m$ in a horizontal dc discharge tube. For these particles the weak ambipolar radial electric field is sufficient to compensate gravity allowing us to study dust charging in the quasineutral plasma. Highly space and time-resolved measurements of the particle flow and comprehensive probe measurements of plasma parameters make it possible to use theoretical models where the only unknown parameter is the particle charge. This enables us to determine the charge experimentally by two independent methods. The results are then compared with those of molecular-dynamics (MD) simulations.

The experiment is performed in a dc discharge generated in an U-shaped glass tube, the PK-4 facility (see sketch in Fig. 1), and operated in neon at pressures 20–100 Pa and current of 1 mA (voltage of 1 kV). The plasma parameters are measured in the absence of dust



FIG. 1. Sketch of the experimental setup.

using a probe with a length of 4.5 mm and a radius of 25 μ m. For our conditions, the electrons are in the collisionless regime and, thus, the Druyvesteyn formula is used to calculate the electron density n_e and the electron temperature T_e [16]. The maximum of the first derivative of the probe current is used to estimate the plasma potential [16]. Results of the probe measurements of averaged axial values of n_e , T_e , and axial electric field *E* are presented in Fig. 2. The ion temperature is assumed to be close to the neutral gas temperature, $T_i \simeq T_n \simeq 0.03$ eV, for the pressure range used.

When the dust particles (plastic spheres with mass density of 1.51 g/cm³) are injected into the discharge, they charge negatively and drift against the discharge electric field in the horizontal part of the tube (see Fig. 1). The particle flow is illuminated by a laser sheet of a width $100 \pm 30 \ \mu$ m. The particle motion is recorded by a video camera with a field of view of $6.4 \times 4.8 \ \text{mm}^2$ and a rate of 120 frames per second. The measurements are taken with the camera positioned as shown in Fig. 1; the particle dynamics is essentially the same along the tube. From an analysis of the digitized video the dust number density n_d can be estimated by counting the number of particles in single snapshots. The particle velocities, V_d , can be obtained by tracking individual particles through the video sequence.

We study the particle dynamics varying neutral gas pressure p and the number of injected particles N_d (controlled by settings of the particle dispenser). This allows us to determine the dust-particle charge by two methods. For a sufficiently low number of particles the flow is stable for all pressures studied. The charge is estimated from the force balance condition using the experimentally found particle velocities at different pressures.



FIG. 2. Results of probe measurements; Electron temperature (circles), electron density (squares) and electric field (triangles). Dashed lines correspond to linear fits used in the calculations.

For larger N_d the transition to unstable flow with a clear wave behavior occurs at a certain threshold pressure p_* (see Fig. 3). The transition is a manifestation of the ion-dust streaming instability, caused by the relative drift between the dust and the ion components. The transition can be found experimentally with an accuracy of ~ 1 Pa. The value of p_* depends strongly on N_d (shifting towards higher pressures when N_d is increased). An upper value of N_d , corresponding to $n_d \simeq 4 \times 10^5$ cm⁻³ gives an upper limit of p_* of approximately 60 Pa. This value of n_d is chosen to ensure that the discharge parameters are not strongly modified by the presence of dust and we can use the results of the probe measurements [17]. The images of particle motion just above (1–4 Pa) and at the threshold are recorded and analyzed. In this case the charge can be estimated from a linear dispersion relation describing the transition of the particle flow to the unstable regime at p_* . In addition, the force balance condition for pressures

Below we present theoretical models for the two methods of charge estimation.

above p_* is used to estimate the charge.

(i) Force balance.— The particle velocity in a stable flow is determined by the balance of the forces acting on the particles: The electric force, $F_{el} = QE$, the neutral drag force, $F_n = -m_d \nu_{dn} V_d$, and the ion drag force, $F_i = m_d \nu_{di} (u_i - V_d) \simeq m_d \nu_{di} u_i$. Here m_d is the dustparticle mass, u_i is the ion drift velocity, ν_{dn} and ν_{di} are the momentum transfer frequencies in dust-neutral and dust-ion collisions, respectively. The force balance is

$$F_{el} + F_i + F_n = 0. (1)$$

For the momentum transfer in dust-ion collisions we use



FIG. 3. Particle flow just above the threshold at pressure $p \approx 56$ Pa (upper figure) and at the threshold pressure $p_* \approx 53$ Pa (lower figure) for the particle number density $n_d = 2.3 \times 10^5$ cm⁻³. Both pictures have a field of view of 6.4×4.8 mm². The center of the tube is located at the lower edge of the pictures.

$$m_d \nu_{di} = \frac{8\sqrt{2\pi}}{3} a^2 n_i m_i \upsilon_{T_i} \left(1 + \frac{1}{2} \frac{R_{\rm C}}{a} + \frac{1}{4} \frac{R_{\rm C}^2}{a^2} \Lambda \right), \quad (2)$$

where $R_{\rm C} = |Q|e/T_i$ is the Coulomb radius for ion-dust collisions, m_i is the ion mass, $v_{T_i} = \sqrt{T_i/m_i}$ is the ion thermal velocity, and Λ is the *modified* Coulomb logarithm [Eq. (12) of Ref. [18]]. In deriving Eq. (2) a subthermal ion drift ($u_i \leq v_{T_i}$) is assumed, which means that the effective screening length is close to the ion Debye radius, $\lambda_{\rm D} \simeq \lambda_{\rm Di} = \sqrt{T_i/4\pi e^2 n_i}$ [19]. From momentum conservation we have $\nu_{id} = \nu_{di}(m_d n_d/m_i n_i)$, where ν_{id} is the momentum loss frequency of the ions in ion-dust collisions. The momentum transfer in dust-neutral collisions is $m_d \nu_{dn} = (8\sqrt{2\pi}/3)\delta a^2 n_n m_n v_{T_n}$, where m_n , n_n , and v_{T_n} are the mass, density, and thermal velocity of neutrals, respectively. The numerical factor $\delta =$ $1 + \pi/8 \simeq 1.4$, corresponding to diffuse scattering with full accommodation is chosen in accordance with recent experimental results [20]. The ion drift velocity is determined by ion-neutral and ion-dust collisions, $u_i \simeq$ $eE/m_i \nu_i^{\text{eff}}$, where $\nu_i^{\text{eff}} = \nu_{in} + \nu_{id}$. The frequency ν_{id} is given above. For ν_{in} we use an estimate $\nu_{in} = n_n \sigma_{in} \nu_{T_i}$, with the effective momentum transfer cross section taking into account both charge exchange and polarization interaction, $\sigma_{in} \simeq 10^{-14} \text{ cm}^2$ [21,22].

(ii) Linear dispersion relation.—Though linear theory might not be applicable to describe the wave mode observed in the experiment (e.g., wave number and frequency), it should be adequate to predict the onset of self-excited waves at p_* . In the derivation of a dispersion relation the following effects are taken into account: Electron, ion, and dust collisions with neutrals, ion-dust collisions, and drifts of the electron, ion, and dust components relative to the stationary neutral gas. We also assume "warm" electrons and ions and "cold" dust. Using the hydrodynamic approach we get

$$1 + \frac{\omega_{pe}^{2}}{k^{2}v_{T_{e}}^{2} - ku_{e}(i\nu_{en} + ku_{e})} + \frac{\omega_{pi}^{2}}{k^{2}v_{T_{i}}^{2} + ku_{i}(i\nu_{i}^{\text{eff}} - ku_{i})} - \frac{\omega_{pd}^{2}}{\omega(\omega + i\nu_{dn})} = 0, \quad (3)$$

where k and ω are the wave number and frequency, $\omega_{pi(e)} = v_{T_{i(e)}}/\lambda_{\text{D}i(e)}$ is the ion (electron) plasma frequency, and $\omega_{pd} = \sqrt{4\pi Q^2 n_d/m_d}$ is the dust plasma frequency. The electron-neutral collision frequency and electron drift velocity are given by $\nu_{en} = n_n \sigma_{en} v_{T_e}$ and $u_e = eE/m_e \nu_{en}$, respectively. In a neon plasma with $T_e \gtrsim$ 1 eV, we have $\sigma_{en} \simeq 2 \times 10^{-16}$ cm² [23].

We solve Eqs. (1) and (3) numerically for the plasma parameters taken from probe measurements. The ion density is obtained from the quasineutrality condition $n_e + Z_d n_d = n_i$, where $Z_d = |Q|/e$ is the particle charge number. As discussed above, we assume that n_e is unaf-

fected by the presence of dust [17], but n_i is increased. Hence, in our calculations n_i , ν_{di} , ν_{id} , u_i , ω_{pi} , and ω_{pd} are functions of the particle charge only. Equation (1) is solved directly, yielding the particle charge. Solution of the dispersion relation (3) gives the dependence of $\omega =$ $\omega_r + i\omega_i$ on the wave number k for a given particle charge. The charge is then determined by matching the experimental observations: Stable mode ($\omega_i < 0$ for all k) above the threshold pressure p_* and unstable mode ($\omega_i >$ 0 for a range of k, corresponding to experimentally found wavelengths) below p_* . An illustration of such a solution is shown in Fig. 4. The results of both methods are presented in Fig. 5 and demonstrate good agreement. The error bars correspond to the uncertainties in n_d $(50\%), n_e (30\%), E (10\%), and V_d (15\%)$. Both methods are quite insensitive to the value of T_e .

To have an independent verification of the charge estimates described above, MD simulations of particle charging have been carried out for conditions similar to those of the experiment. The simulations are performed using a code originally developed by Zobnin *et al.* [6] to study the effect of ion-neutral collisions on the particle charging. As seen from Fig. 5 the charge found from the simulations is in good agreement with the results of both experimental methods. Some discrepancies can be attributed to the fact that the conditions used in the simulations are not completely identical to those of the experiment (e.g., weak plasma anisotropy, ion losses to the tube walls, ion-neutral polarization interaction are not taken into account).



FIG. 4 (color online). Numerical solution of the linear dispersion relation (3). The dust number density and threshold pressure correspond to those of Fig. 3. Real and imaginary parts of wave frequency ω are given by solid and dashed lines, respectively (note that the imaginary parts are multiplied by a factor 10). Red/blue (light gray/dark gray) lines correspond to p = 53 (56) Pa. Particle charge number in this calculation is $Z_d = 2 \times 10^3$.



FIG. 5 (color online). The particle charge obtained from experiments [force balance for low number of injected particles (open circles); force balance for pressures above the threshold (open squares), solution of dispersion relation (solid squares)] and from MD simulations (red diamonds). The area between the two dotted lines corresponds to the charge given by the OML model for Havnes parameters between P = 0.2 (upper line) and P = 3 (lower line).

Figure 5 shows the results of charge calculations using the OML theory for the range of the Havnes parameter, $0.2 \leq P \equiv Z_d n_d / n_e \leq 3$, estimated from experimentally determined charges. We use the OML expression modified to take into account the contribution of dust to the qua $v_{T_e} \exp(-Z_d e^2/aT_e) =$ sineutrality condition, $v_{T_i}(1 + Z_d e^2 / aT_i)(1 + P)$ [4]. The difference between OML theory and the charges found from experiments and MD simulations is most significant at higher pressures. At $p \sim 100$ Pa we have $P \simeq 0.2$, $\Delta/\lambda_D \simeq 3.6$ (where Δ is the intergrain distance), and $l_i/\lambda_D \simeq 0.6$. This means that the quasineutrality is weakly affected by the dust, the effect of "closely packed" grains is insignificant, and, therefore, we attribute the dramatic charge suppression (up to 5 times) at higher pressures to the effect of ion-neutral collisions. For the lowest pressures investigated, $l_i/\lambda_D \simeq 3$ and the charge tends towards the OML values, as expected from theory [5,6]. Some difference still exists and can be caused by a combination of all the effects mentioned above. Experiments with a smaller N_d (low dust density) as well as MD simulations show a decrease of charge with pressure. This can be mainly due to an increase in ion collisionality [6] (it is unreasonable to expect more than a 30% effect due to decrease in T_e , see Fig. 2). Experiments with a larger N_d reveal the opposite tendency which might be explained by some modifications of the plasma parameters at high dust number densities. These tendencies are not strongly pronounced, however, especially taking into account the experimental uncertainties, and $Z_d \simeq 1500 \pm 500$ is a reasonable estimate for the whole pressure range investigated.

In conclusion, we have determined the charge of dust particles in a bulk dc discharge plasma under the conditions when the ion mean free path is comparable to the plasma screening length. Two independent experimental methods and MD simulations agree well with each other and yield a charge which is considerably smaller than that predicted by OML. Thus our results prove experimentally the significant effect of ion-neutral collisions on particle charging in plasmas.

The authors highly appreciate valuable comments of Professor O. Havnes and Dr. P. Bryant. This work was supported by DLR under Grant No. 50 WP 0204.

- [1] C. K. Goertz, Rev. Geophys. 27, 271 (1989).
- [2] L. Boufendi and A. Bouchoule, Plasma Sources Sci. Technol. **11**, A211 (2002).
- [3] H. M. Thomas and G. E. Morfill, Nature (London) 379, 806 (1996).
- [4] V.E. Fortov et al., Phys. Usp. 47, 447 (2004).
- [5] R.V. Kennedy and J.E. Allen, J. Plasma Phys. 69, 485 (2003).
- [6] A.V. Zobnin et al., JETP 91, 483 (2000).
- [7] M. Lampe et al., Phys. Rev. Lett. 86, 5278 (2001).
- [8] M. Lampe et al., Phys. Plasmas 10, 1500 (2003).
- [9] Z. Sternovsky, S. Robertson, and M. Lampe, J. Appl. Phys. 94, 1374 (2003).
- [10] A. Barkan, N. D'Angelo, and R. L. Merlino, Phys. Rev. Lett. 73, 3093 (1994).
- [11] A. Melzer, A. Trottenberg, and A. Piel, Phys. Lett. A 191, 301 (1994).
- [12] A. Homann, A. Melzer, and A. Piel, Phys. Rev. E 59, 3835(R) (1999).
- [13] E. B. Tomme et al., Phys. Rev. Lett. 85, 2518 (2000).
- [14] V.E. Fortov et al., Phys. Rev. Lett. 87, 205002 (2001).
- [15] A. A. Samarian and S.V. Vladimirov, Phys. Rev. E 67, 066404 (2003).
- [16] V. Demidov, S. Ratynskaia, and K. Rypdal, Rev. Sci. Instrum. 73, 3409 (2002).
- [17] This assumption is supported by our estimates showing that below this particle density the electron mobility is unaffected by the dust, and electron loss to the particles is smaller than that due to recombination on the walls of the tube. The latter means that the ionization/recombination balance of electrons is not changed by the presence of the dust and neither are T_e and E. For a dc discharge at constant current, $I \propto n_e E$, we conclude that the average value of n_e is also unaffected by the dust.
- [18] S. A. Khrapak et al., Phys. Rev. E 66, 046414 (2002).
- [19] S. A. Khrapak et al., Phys. Plasmas 10, 4579 (2003).
- [20] B. Liu et al., Phys. Plasmas 10, 9 (2003).
- [21] M. A. Biondi and L. M. Chanin, Phys. Rev. 94, 910 (1954).
- [22] R. N. Varney, Phys. Rev. 88, 362 (1952).
- [23] A. Gilardini, Low Energy Electron Collisions in Gases (Wiley, New York, 1972).