A new diagnostic to characterize a plasma crystal

B M Annaratone\textsuperscript{1}, P Bandyopadhyay\textsuperscript{2}, M Chaudhuri\textsuperscript{1} and G E Morfill\textsuperscript{1}

\textsuperscript{1} Max-Planck Institut für Extraterrestrische Physik, D-85740 Garching, Germany
\textsuperscript{2} Institute for Plasma Research, Bhat, Gandhinagar 382428, India
E-mail: bma@mpe.mpg.de

\textit{New Journal of Physics} 8 (2006) 306
Received 18 September 2006
Published 6 December 2006
Online at \url{http://www.njp.org/}
doi:10.1088/1367-2630/8/12/306

\textbf{Abstract.} We describe experiments aimed at characterizing complex plasma crystals and their interaction with the space charge surrounding an RF discharge. By analysing the complex plasma radiofrequency impedance, we were able to measure the density of the free electrons of the crystal. The density of the ions was derived by Langmuir probe measurements in the bulk plasma combined with a model of the RF plasma sheath. Surprisingly the full characterization of the crystal reveals that the 3D ensemble is quasi-neutral, i.e. a three component plasma and the particles levitate in their self-built electric field, much lower than the sheath field. This result is inherent to the 3D geometry.

\textbf{Contents}

1. Introduction 2
2. Experimental set-up 2
3. Derivation of the electron density 2
4. The RF sheath and derivation of the ion density 7
5. Characterization of the plasma crystal 8
6. Conclusion 9
Acknowledgments 9
References 9
1. Introduction

Plasma technology plays a very important role in many aspects of our daily lives—from computer chips, telecommunication chips, solar cells to flat TV screens, to name but a few. This technology will become more important still, as plasma control of surface modifications advances to even smaller scales and new developments, such as ‘smart plasmas’ and ‘microplasmas’ need to be perfected. In all these devices, knowledge of the detailed structure of the plasma sheath will become progressively more necessary—yet currently sheaths are the effects that are physically least well characterized. The problem is compounded by the fact that plasma in such devices may contain fine (nano or micro) powders either by design [1], or as a pollutant [2], which affect the detailed plasma structure and introduce temporal evolution. Apart from technology, microparticles in plasmas—so called complex plasmas—have become an important topic for fundamental research, ever since it was discovered that these plasmas may exist in strong coupling (liquid or crystalline) states [3]–[5]. Complex plasmas have since been used to experimentally study the kinetics of phase transitions, crystallization front dynamics [6], shear flows, cluster stability [7], new waves/phonon modes [8], shocks and solitons [9], and the kinetic of turbulent flows [10], to name but a few of these topics. For a recent and comprehensive review see [11]. In addition there is the growing topic of dust hazard in fusion reactors [12] and astrophysics motivations [13].

In this study, we address both issues: the characterization of the plasma sheath and the fundamental interaction between plasma and microparticles. We present a new diagnostics based on the physical concept of the electron plasma resonance and of the plasma sheath resonance. This diagnostic has been applied to a three component complex plasma to provide the density of the free electrons (bound electrons constitute the negative charge of the particles). The density of the free electrons reduces the indetermination of the system and, together with more conventional diagnostics and the modelling of the RF sheath, we have been able to fully characterize the particle assembly.

This study was originally motivated by the observation of the effect shown in figure 1. The crystal height above the lower electrode does not scale as the dimension of the sheath. Surprisingly the thickness of the crystal is almost independent of the pressure.

2. Experimental set-up

The experiments were performed in a standard RF reactor in which the upper electrode was RF driven and the lower electrode was adaptive and mainly grounded. Only a small, central part of the lower electrode was driven to manipulate the particle as in [14, 15], where the conventional diagnostics are also described. To be absolutely sure that the effects observed did not depend on any dc current we covered the lower electrode with a 0.55 mm thick glass. Spherical particles (Melamine-formaldehyde with diameters 3.42, 6.87 and $8.77 \mu m$) were injected in the chamber and levitated in the argon plasma-sheath above the glass, forming a crystal. This study concerns the sheath fully loaded with particles, that does not support any particular layer.

3. Derivation of the electron density

The new diagnostic consists of applying an RF voltage, of variable frequency and constant amplitude, to the central small electrode. The RF voltage induces an RF current circulating
Figure 1. The position of the crystal as a function of the pressure. The upper line represents the plasma boundary as derived from the integrated visible emission. Particle radius 3.4 \( \mu \text{m} \), \( V_{\text{RF-electrode}} = 300 \ V_p-p \).

Figure 2. Particles \( \Phi = 6.87 \ \mu \text{m} \) levitated above the central electrode driven in non-resonant frequency. Dimensions: 21.5 mm \( \times \) 9.88 mm.

through the sheath and the plasma. In our case, with a complex plasma facing the electrode, the displacement of the particles induced by the dc bias of the glass is a clear proof of this RF current. As the RF frequency is increased the displacement is first independent of the frequency over a large range, see figure 2, with respect to the dc case, figure 3, is then amplified, figure 4, and later reduces almost to zero, figure 5. This cut-off occurs because the electron plasma resonance of the crystal is reached, see the experimental curves in figure 6 for three particle sizes. At frequencies
Figure 3. Particles $\Phi = 6.87 \, \mu m$ levitated above the central electrode driven in the dc case. Dimensions: 21.5 mm $\times$ 9.88 mm.

Figure 4. Particles $\Phi = 6.87 \, \mu m$ levitated above the central electrode driven in resonant frequency, 23 MHz. Dimensions: 21.5 mm $\times$ 9.88 mm.

approaching $f_{pe}$, as for the plasma sheath resonance [16] and the resonance probe [17], the region next to the central electrode, the plasma sheath, can be described in terms of positive permittivity, while the complex plasma above has a negative permittivity. The peak in the curves is not always evident. It is clear when the crystal is low and the geometry is, as in figures 2–5, almost a hemisphere. The radii are all equal and the resonance is narrow. The peak broadens when the crystal is high above the electrode because the current spreads in many possible paths, each with a different resonant frequency. Our geometry is ‘hybrid’, and the ‘series’ resonance is sometimes broadened; however there is a substantial coherence between the maximum, $f_{res}$, and the cut-off, as for planar geometry, [16]:

$$f_{res} = f_{pe} \sqrt{\frac{s}{2s + p}}$$

with $s$ the thickness of the
Figure 5. Particles $\Phi = 6.87 \mu m$ levitated above the central electrode driven at a frequency approaching $\omega_{pe}$, 50 MHz. Dimensions: 21.5 mm $\times$ 9.88 mm.

Figure 6. The normalized displacement of the particles at $P = 19$ Pa as a function of the frequency. $V_{\text{RF–electrode}} = 300 V_{\text{p–p}}$.

sheath below the crystal and $p$ the path of the current in the crystal (figure 7). Using these values $f_{pe}/f_{res}$ are given in table 1 column 9 while the experimental values are in column 8. The electron density in the crystal has been obtained from the definition of electron plasma frequency $f_{pe}$, a cut-off unique to any geometry, using the frequency of the last experimental point, at which the displacement of the particles is comparable with one particles’ layer thickness, our ultimate error for the displacement. It is clear that only the free electrons participate in the resonances in our frequency range. In two component plasma physics the presence of ions does not much modify the electron plasma frequency, although the opposite is not true. In complex plasmas the lighter species, electrons, has the same resonances with or without dust (see also the study on the propagation of Langmuir waves in dusty plasmas [8]). Electron losses are negligible because
Figure 7. Schematic of the RF current path in the sheath and in the crystal for frequency below 100 MHz.

Table 1. Data corresponding to figure 6. Column: 1, diameter of the particles; 2, average distance between grains; 3, relative variation of the distance between grains from the top and bottom layers; 4, vertical dimension of the crystal; 5, density of the grains, derived from column 2; 6, plasma-sheath resonance; 7, cut-off frequency; 8, ratio \(f_{pe}/f_{res}\) from columns 5 and 6; 9, theoretical ratio \(f_{pe}/f_{res}\); 10, density of free electrons in the crystal as derived from column 6; 11, \(T_e\) from Langmuir probe; 12, \(n_e\) in the main plasma from Langmuir probe; 13, space potential of the main plasma with respect to ground, from Langmuir probe; 14, RF fluctuation of the main plasma, from the driven Langmuir probe; 15, dc potential of the crystal with respect to the plasma edge, derived from equation (1); 16, \(kT_e/eL\), the field sustainable in a quasi-neutral plasma.

| \(a (\mu m)\) | \(\Delta (\mu m)\) | \(\Delta \Delta (\mu m)\) | \(L (mm)\) | \(n_e (10^{13} m^{-3})\) | \(f_{res} (MHz)\) | \(f_{pe} (MHz)\) | \(k\) | \(k (n_e)\) | \(T_e (eV)\) | \(V_p (V)\) | \(V_{RF} (V)\) | \(V_{c} (V)\) | \(kT_e/eL (V mm^{-1})\) |
|-------------|-------------|-----------------|--------|-----------------|-----------|-----------|---|--------|--------|--------|-------|-------|-----------------|
| 3.42        | 214         | 0.167           | 1.13   | 1.02            | 14.7      | 46.7      | 3.16 | 1.93   | 2.69   | 3.29   | 4.9    | 23.0  | 32.0            | 13.4  | 2.90            |
| 6.87        | 464         | 0.284           | 1.91   | 0.1             | 33.8      | 79.4      | 2.34 | 1.38   | 7.78   | 3.88   | 6.3    | 29.2  | 18.4            | 8.4   | 1.05            |
| 8.77        | 358         | 0.289           | 1.83   | 0.2             | 58.8      | 119.9     | 2.03 | 1.37   | 17.7   | 3.31   | 11.7   | 28.1  | 21.4            | 6.5   | 1.80            |

the electron current taken away by the particles is less than 1% of the electron flux to the electrode, equal to the collision-modified Bohm flux. Moreover this resistive element would not shift the resonance, it would only reduce the quality factor. Some very small particles, on the top of the crystal, visible in figures 2–5, do not shift their position at all, showing that the RF current does not reach there. For frequencies above the cut-off, \(\approx 100\) MHz, the RF current approaches the plasma sheath resonance of the main two component plasma above the crystal and the resonant path closes to ground through the opposite electrode. In this case the assembly of particles is liquid in the lower part, due to a ‘shortage’ of electrons no longer in equilibrium, and the main plasma becomes brighter showing power deposition.
Table 2. Column: 1, diameter of the particles; 2, ion density in the crystal, collisional sheath (CS); 3, charge of the grains, CS; 4, electric field derived from levitation, CS; 5, floating potential of the grain with respect to the local space potential, CS; 6, floating potential from OML, with energetic ions, CS; 7, combined Debye length, CS; 8, Havnes parameter, CS; 9, ion density in the crystal, collisionless sheath (CL); 10, charge of the grains, CL; 11, electric field derived from levitation, CL; 12, floating potential of the grain with respect to the local space potential, CL; 13, floating potential from OML, with energetic ions, CL; 14, combined Debye length, CL; 15, Havnes parameter, CL.

| a  | (n_i)CS | Z_g(e) | E | (V_i)_g | (V_i)_OML | λ_d | (n_i)CL | Z_g(e) | E | (V_i)_g | (V_i)_OML | λ_d | P_H |
|----|--------|--------|---|--------|-----------|-----|--------|--------|---|--------|-----------|-----|-----|
|    | 10^14 m^-3 | (×10^3) | kT_e/e | kT_e/e | kT_e/e | (V_m f.p.) | (V_m f.p.) | kT_e/e | kT_e/e | kT_e/e | kT_e/e | kT_e/e | kT_e/e | kT_e/e | kT_e/e |
| 3.42 | 2.54 | 2.23 | 0.861 | 0.571 | 2.07 | 1318 | 8.46 | 1.0 | 0.75 | 2.56 | 0.2 | 3.00 | 1862 | 2.83 |
| 6.87 | 3.27 | 24.9 | 0.625 | 2.69 | 2.67 | 1148 | 3.21 | 1.8 | 10.4 | 1.49 | 1.1 | 3.42 | 1148 | 1.34 |
| 8.77 | 6.08 | 19.7 | 1.64 | 1.96 | 2.82 | 711 | 2.42 | 3.5 | 8.1 | 3.99 | 0.8 | 3.54 | 714 | 0.99 |

4. The RF sheath and derivation of the ion density

In modelling the boundary of the main plasma above the crystal we safely assume that this is not modified by the presence of the crystal downstream because of its small dimension with respect to all the other areas surrounding the plasma. The m.f.p. of the electrons is larger than the overall bulk-plasma electrode sheath and the electron losses are negligible. Therefore $n_e$ can be approximated by a Maxwellian distribution enhanced by the presence of RF [15]:

$$n_e = n_e(0) \exp \left( \frac{eV_{DC}}{kT_e} \right) I_0 \left( \frac{eV_{RF}}{kT_e} \right),$$

where $V_{DC}$ and $V_{RF}$ are the potentials in the sheath (they have a constant ratio as measured by Langmuir probe in the bulk plasma), $T_e$ is the electron temperature and $I_0$ is the zeroth order modified Bessel function of the first kind. Inserting $n_e$ provided by the plasma sheath resonance in equation (1), we can derive the potential difference between the main plasma and the plasma crystal, penultimate column in table 1. Following the weakly collisional calculations by Riemann [18] this potential difference includes a quasi-neutral, dimensionless, pre-sheath, $V_x = \sqrt{(x_0 - x)/\lambda_{m f.p.}}$ from which ions exit with the Bohm speed, and a transition region with an electric field of the order of $kT_e/e/\lambda_{m f.p.}^2/5$. If we take a typical m.f.p. of 0.3 mm for slow ions, the pre-sheath voltage is: $V_0 = 1kT_e/e$. Then the flux of ions out of the plasma can be estimated:

$$J = n_i(0) \exp \left( eV_0/kT_e \right) \sqrt{kT_e/M},$$

where $n_i(0) = n_e(0)$. The density of the ions in the crystal is obtained dividing the flux by the ion drift velocity in the transition region field, see column 2 in table 2.

We now compare the above calculation of the ion density with the collisionless model which would provide the lower limit for the ion density. In our case, we assume the absence of collisions only in the upper part of the sheath; this should still be valid at somewhat higher pressures than for the full collisionless sheath as in [19]. Consequently the ion density, $n_i$, is derived from the
continuity equation:

\[ n_i = n_i(0) \exp \left( -\frac{V_0}{kT_e} \right) (1 - \frac{V}{V_0})^{-1/2}, \]

where \( V_0 = \frac{1}{2} kT_e/e \), according to the Bohm criterion (see column 9 in table 2). On average the collisional model gives a density of the ions about twice that using collisionless calculation; nevertheless the main results of this paper are basically unchanged. The latter model does not assume any spatial distribution for the potential and would be applicable also in the presence of a double layer, of thickness \( \lambda_D \) above the crystal [20]. The energy acquired in the double layer is much larger (and the m.f.p. longer) than the energy acquired outside, where ions may have suffered collisions.

5. Characterization of the plasma crystal

The physical quantities measured or derived so far allow us to characterize the plasma crystal. Let us assume for now that the crystal is quasi-neutral (we shall come back to this later). If this is the case \( n_i/n_e = n_g z_g/n_e + 1 \), with \( n_g \) and \( z_g \) respectively the density and the charge number of the grains. With the experimental densities for the three components of the plasma, \( z_g \) is derived (see table 2). Charge and mass of the particles give us the minimum electric field \( |E| \) required for levitation (\( E \) may exceed this value if ion drag is relevant, it may not be important in our case, as in [21]). The derived field is comparable or smaller (see table 2) than \( kT_e/L \) with \( L \) the vertical dimension of the crystal. This latter quantity is about the maximum field sustainable by a quasi-neutral plasma; in this way the above statement of quasi-neutrality is self-consistently validated. The field of the crystal is also much smaller than the electric field derived by the solution of the unperturbed Poisson equation with the RF enhanced electron density at the same position. Apparently the maximum dimension of the crystal (and the maximum number of particles that can be levitated) is limited by the electric field required for levitation, at a given electron temperature. This statement is further validated by the observation that the thickness of the crystal is constant in the range of pressure 10–80 Pa, where the electron temperature does not vary significantly.

Using the vacuum approximation, which is reasonable for small particles, the floating potential of the grains, \( V_{f-grain} \) has been derived. It is consistently lower, in absolute value, than the floating potential predicted by the OML theory [22] for isolated particles in a supersonic ion stream (see \( V_{f-OML} \) table 2). Two counteracting effects may modify the charge on a grain in a crystal with respect to its charge when isolated. The neighbouring grains limit the access of ions and this, in the floating condition, would reduce the electron flow. The negative potential of the grain is consequently more repelling and higher in absolute value. However this effect is not applicable in our case because the Bohm flow out of the plasma, \( \simeq 0.5 \text{ A m}^{-2} \) would sustain \( 10^5 \) layers. In contrast, in a plasma crystal, neighbouring particles ‘compete’ to absorb free charges. For interacting grains the space charge is, at a certain distance from the grain, more positive than for the isolated grain. The ion flow increases leading to a net charge reduction per grain, which implies a lowering of the repulsive potential with respect to OML (see e.g. Havnes et al [23]) in agreement with our data.

The Havnes parameter represents the ratio between the negative charge bound to the grains and the free electrons per unit volume. \( P_H = n_g z_g/n_e \). Our new diagnostic provides directly the denominator, even in the case of strongly interacting particle systems. It should be noted that
estimates of the Havnes parameter have been, until now, only possible in weakly interactive systems where the average density of the electrons approaches the density of the electrons in the inter-particle volume.

6. Conclusion

The analysis of the radiofrequency impedance is a powerful method to study plasmas in a variety of different situations in general and small plasma crystals in particular. The interpretation of the spectra will form the basis for future experiments with high frequency and pulses in charged dusty environments (to give an example pollution in the ionosphere and mesosphere and other).

We have demonstrated that the levitation of each particle of the crystal is due to a balance of gravity and electrostatic forces in the local pre-sheath field of the crystal. Small inequalities in the levitation balance are compensated by elastic forces, i.e. the crystal ‘rests’ partly on the lower layer while it ‘hangs’, in part, from the upper one (see $\delta \Delta / \Delta$ in table 1). From another point of view, however, we may consider the crystal like a macroscopic assembly levitating in the strong field of the sheath. The sheath electric field is screened by the presence of this ‘porous body’ with double layers, see also [20], and a Bohm point, taking into account the supersonic ion drift, towards the electrode. In this second interpretation individual particles are all supported by collective interaction.

The main result of this paper is the difference between 2D layers, which can exist in a sheath if the curvature of the potential is in a direction normal to the layer, and 3D assemblies, where layers interact and quasi-neutrality is the energetically favourable state. The capacity of a very small quantity of mass in screening high electric fields in charged environments is astonishing. The particles act as a selective sink for electron-ion couples, ions being limited by the Bohm flow. The mechanism always reduces the charge separation and therefore the energy of the system. We can foresee possible applications in astrophysics.

Acknowledgments

We would like to thank Herr Steffes for continuous assistance. This research was founded by: Das Bundesministerium für Bildung und Forschung durch das Zentrum für Luft- und Raumfahrt e.V. (DLR) unter dem Förderkennzeichen 50 RT 0207.

References

[1] Roca P, Cabarocas I, Gay P and Hadjadj 1996 J. Vac. Sci. Technol. A 14 655
[2] Selwyn G S, Singh J and Bennet R S 1989 J. Vac. Sci. Technol. A 7 2758
[3] Thomas H M, Morfill G E, Dumel G V, Goree J, Feuerbacher B and Mhlmann B 1994 Phys. Rev. Lett. 73 652
[4] Chu J H and I L 1994 Phys Rev. Lett. 72 4009
[5] Thomas H M and Morfill G E 1996 Nature 379 806
[6] Rubin-Zuzic M, Morfill G E, Ivley A V, Pompl R, Klimov B A, Bunk W, Thomas H M, Rothermel H, Havnes O and Fouquet A 2006 Nature Phys. 2 181
[7] Antonova T, Annaratone B M, Yaroshenko V, Thomas H M and Morfill G E 2005 Phys. Rev. Lett.
[8] Shukla P K and Mamun A A 2002 Introduction to Dusty Plasma Physics (Bristol: IOP Publishing) ISBN: 0-7503-0653

New Journal of Physics 8 (2006) 306 (http://www.njp.org/)
[9] Samsonov D, Zhdanov S K, Quinn R A, Popel S I and Morfill G E 2004 Phys. Rev. Lett. 92 255004
[10] Morfill G E, Rubin-Zuzic M, Rothermel H, Ivley A v, Klumov B A, Thomas H M and Konopka U 2004 Phys. Rev. Lett. 92 175004
[11] Fortov V E, Ivley A V, Khrapak S A, Khrapak A G and Morfill G E 2005 Phys. Rep. 421 1
[12] Winter J 2004 Plasma Phys. Control. Fusion 46 B583
[13] Verheest F 2000 Plasma Phys. Control. Fusion 41 (Suppl. 3A) A445
Verheest F 2000 Waves in Dusty Space Plasmas (Dordrecht: Kluwer)
[14] Annaratone B M, Glier M, Stuffer T, Thomas H, Raif M and Morfill G E 2003 New. J. Phys. 5 92
[15] Annaratone B M, Antonova T, Goldbeck D D, Thomas H M and Morfill G E 2004 Plasma Phys. Control. Fusion 46 B495–B509
[16] Annaratone B M, Ku V P T and Allen J E 1995 J. Appl. Phys. 77 5455
[17] Blackwell D D, Walker D N and Amatucci W E 2005 Rev. Sci. Instrum. 76 23503
[18] Riemann K U 1997 Phys. Plasmas 4 4158
[19] Ingram S G and Braithwaite N St J 1988 J. Phys. D: Appl. Phys. 21
[20] Annaratone B M, Khrapak S A, Bryant P, Morfill G E, Rothermel H, Thomas H M, Zuzic M, Fortov V E, Molotkov V I and Nefedov A P 2002 Phys. Rev. E 66 056411
[21] Tomme E B, Annaratone B M and Allen J E 2000 Plasma Sources Sci. Technol. 9 87–96
[22] Allen J E, Annaratone B M and de Angelis U 1999 Plasma J. Phys. 63A 299
[23] Havnes O, Goertz C K, Morfill G E, Gruen E and Ip W 1987 J. Geophys. Res. 92 2281