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Formation and behaviour of dust particle clouds in a radio-frequency discharge: results in the laboratory and under microgravity conditions

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Abstract. In this paper we report the first observation on submicron dust particle clouds grown in a radio-frequency sputtering discharge under microgravity conditions. These results have been obtained in the PKE–Nefedov (Plasma Kristall Experiment) chamber in the framework of a French–German–Russian programme. A dust-free region, usually called the ‘void’, is observed in the laboratory and under microgravity conditions even with submicron particles. In this region, successive generations of particles can be grown, leading to the coexistence of particles with various sizes. Each generation of particles constitutes a cloud separated from the others by a definite sheath. Dynamics of these clouds have been investigated showing vortex-like motions or independent behaviour of small heaps of particles, emphasizing both attractive and repulsive effects between dust clouds. As these particles drastically influence the plasma properties, the growth kinetics is followed through the evolution of the discharge current.

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1. Introduction

Since their discovery in plasma processing reactors [1, 2], the physics of dusty (complex) plasmas is rapidly evolving [3]. These studies are performed using different methods: production of particles in the gas phase using reactive gases [4] (for example in silane [1], [5]–[7]), by sputtering from a target [8]–[12] or by injecting calibrated microparticles. A very exciting observation is the formation of a highly correlated medium, the so-called plasma crystal, where the negatively charged [13]–[15] dust particles arrange themselves in crystalline structures [14], [16]–[18].

To reveal the real interactions between dust particles and pure plasma effects we need to eliminate one of the major constraint acting in laboratory dusty plasmas: gravity force. In this way, microgravity experiments have been envisaged since the early 1990s. The first experiments under microgravity conditions have been performed recently by the Max Planck Institute for Extraterrestrial Physics (MPE) (in parabolic flights and sounding rockets [19]) and by the Institute for High Energy Densities (IHED) of the Russian Academy of Sciences (on board the MIR station [20]). These two laboratories are now involved in a joint programme called PKE–Nefedov for experiments on board the International Space Station (ISS). In early 2001, within the framework of this programme, experiments concerning the study of clouds of injected microparticles started on board the ISS. These experiments were designed to study the real three-dimensional structure of the plasma crystal.

The Research Group on Energetics of Ionized Gases (GREMI) joined IHED and MPE for an extended programme on dust particle growth. These new experiments were performed on board the ISS in late October 2001 by the French–Russian cosmonaut team of the ANDROMEDE taxi-mission. The objectives of this work were to give new insights into dust particle growth. Growth kinetics, spatial distribution and dynamics are investigated under microgravity conditions. Some

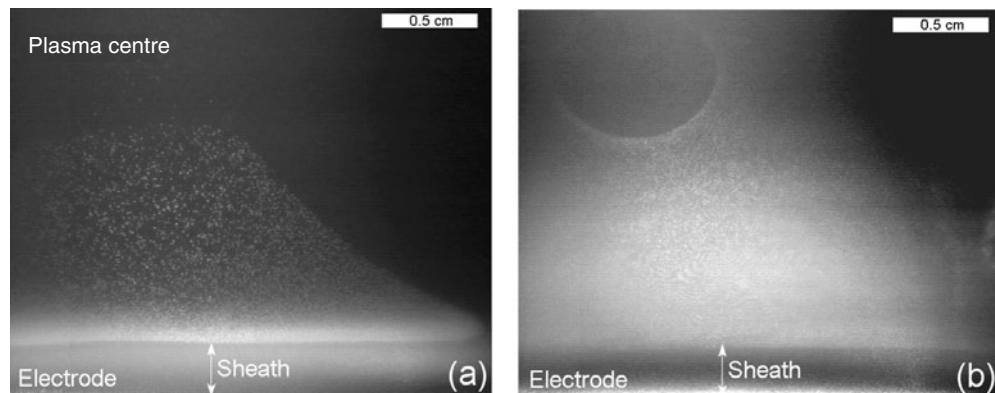


Figure 1. Different shapes of grown clouds (LR camera).

of these results are presented in section 4 and differences with on ground experiments are explored.

2. Experimental basis

2.1. Experimental set-up

The PKE–Nefedov experimental set-up consists in a parallel plate radio-frequency (RF) discharge where an argon plasma is created in push–pull excitation mode. The electrodes are separated by 3 cm and their diameter is 4 cm. Once the plasma is created, micrometre particles (melamine formaldehyde, MF ~ 3.4 and $6.8 \mu\text{m}$) can be injected into the chamber through two dispensers inserted in the centre of the electrodes. The particles are illuminated by a thin laser sheet perpendicular to the electrodes and the scattered laser light is recorded at 90° by two CCD cameras with different magnifications, a high resolution (HR) camera $\times 1$ and a low resolution camera (LR) $\times 1/3$. The laser and the cameras are installed on a motorized platform that can be moved horizontally. Thus the entire cloud can be scanned and three-dimensional information can be extracted. To ensure the more stable conditions the experiments are performed without gas flow. The behaviour and the organization of these clouds have been investigated both on the ground and in microgravity by MPE [19] (three-dimensional structure, presence of a dust-free region called the ‘void’ in the centre of the discharge).

2.2. Dust particle growth

When the plasma is switched off the injected dust particles are no longer trapped and they are evacuated through the gas flow or deposited inside the chamber. When the plasma is switched on anew, this layer of particles lying on the surfaces is subjected to ion bombardment so it can be the source of new particles through this sputtering phenomenon. This possibility has been evidenced in the GREMI laboratory by working in slightly different conditions than those used to study the clouds of injected particles.

3. Results obtained in the laboratory

In order to prepare the future microgravity experiments on board the ISS, experiments have been carried out in the GREMI laboratory to define the best conditions and parameters for dust particle growth. We present in this section the major results obtained during this period.

3.1. Clouds of grown particles

Typical discharge conditions necessary to grow particles are an argon pressure of 1.6 mbar and an RF power of about 2.5 W. In these conditions submicron dust particles can be generated in the plasma with a typical appearance time (laser light scattering observed on a screen) around 2 min. They constitute a cloud that occupies, most of the time, the whole volume of the chamber. We have observed different geometries for the particle cloud grown:

- thin layers near the electrodes
- domelike shape (figure 1(a))
- dense cloud in the whole volume (figure 1(b)).

It is clear that the number of dust particles produced depends on the amount of material deposited in the chamber, but we have observed that the most critical parameter is the purity of the plasma. Indeed, we have seen a direct correlation between the number of dust particles grown and the base pressure before an experiment. This dependence, already mentioned in similar experiments [12], can be related to different sputtering conditions due to molecular impurities (less matter sputtered) or to a modification of the chemistry in the gas phase (the same amount of matter sputtered but no coagulation between clusters in the plasma). In our experimental conditions we have noticed that a base pressure of a few 10^{-5} mbar is essential to obtain a grown particle cloud in the whole plasma chamber. Recent experiments performed with a base pressure around 10^{-6} mbar have improved the particle growth in terms of number of particles and reproducibility. This base pressure dependence is certainly amplified by the fact that we perform our experiments without gas flow leading to a quite important concentration of impurities coming from the walls or from the dust particles. The clear reasons for this dependence are actually under investigation using emission spectroscopy.

During the first minutes of each experiment, the grown particles seem to be monodisperse. After this step, a constant process of sputtering and growth leads to the coexistence of particles with various sizes. This behaviour observed in the laboratory is the most common situation but in some experiments we have observed a huge number of very small grown particles with a very weak change in size during the whole plasma run (1–2 h).

Vortex-like motions [19] are observed near the edge of the electrodes (see figure 2(b)). In these regions the particle cloud behaves like a fluid. In our experimental conditions, this collective motion can be induced by two possible phenomena: thermal convection or electrical effects due to the plasma or the electrodes. This aspect will be clarified thanks to the microgravity experiments described in section 4.

3.2. Dust-free region: void

When a sufficient number of particles is grown in the plasma we can observe the appearance of a dust-free region in the centre of the plasma [21]–[24]. We have noticed that the appearance

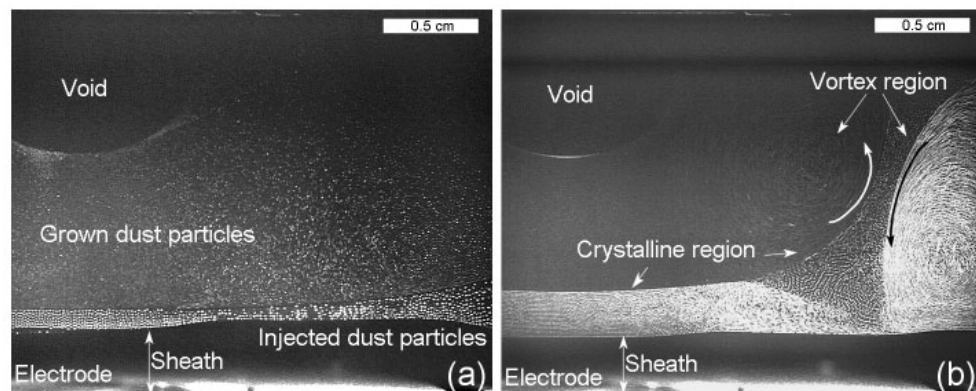


Figure 2. Void region in typical dust particle clouds: (a) mixture of grown and injected particles; (b) various sizes of grown particles with crystalline regions and vortex motions (see also [movie](#)).

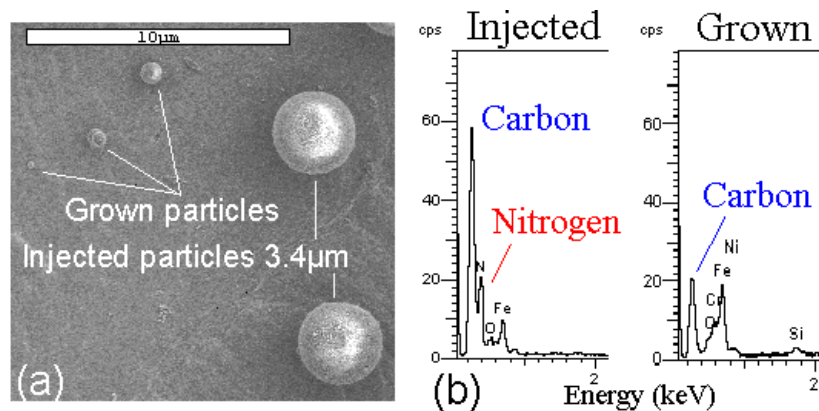


Figure 3. (a) SEM image of injected and grown particles; (b) x-ray fluorescence analysis showing the carbon structure of the grown particles.

of this region takes a few minutes. Indeed, the formation of this void is caused by an evolution of the plasma properties or an evolution of the dust particle population (in size or density). Due to these observations, the influence of impurities from sputtering or from wall outgassing is certainly predominant. Typical clouds showing a void region are shown in figure 2. Figure 2(b) shows various sizes of grown particles with smaller ones near the centre of the plasma and bigger ones near the sheath boundaries. In figure 2(a), the size of the grown particles is compared to calibrated injected particles ($3.4 \mu\text{m}$) occupying the lowest layers of the cloud. Voids with micrometre particles can also be obtained in the laboratory using thermophoretic force [25].

We can also report the frequent observation of the self-generated oscillation of the void usually called the ‘heartbeat’ instability [23]. The oscillation frequency can be controlled through the RF power; it can be increased by increasing the power.

The possibility of observing the void region and the heartbeat instability in laboratory dusty plasmas is a very interesting tool needed to investigate the physics involved in these phenomena.

3.3. Characteristics of the particles

An analysis of the dust particles grown in our experimental conditions has been performed by scanning electron microscopy (SEM) and x-ray fluorescence. These diagnostics have been used in order to characterize particles grown during successive runs and collected on a small stainless steel plate placed on the lower electrode. A more precise analysis on particles grown during the same run must be conducted. Nevertheless, in our sample we have observed spherical particles with sizes between 0.2 and 0.8 μm in diameter as shown in figure 3(a). In the same figure we can show 3.4 μm MF particles. The size of the injected particles is a little smaller than this value, certainly due to the sputtering necessary to grow new particles. X-ray fluorescence analysis is shown in figure 3(b) and allows us to see that the grown particles are principally constituted of carbon (other peaks come from the stainless steel plate used to collect the particles). Indeed, the nitrogen peak present in MF has disappeared.

3.4. Crystalline structure

Figure 2(b) also shows the possibility of obtaining crystalline structures with very small (submicron) particles. In the lower part of this figure we can see a few layers of grown particles exhibiting a crystalline structure. A detailed analysis of the structure obtained is outside the scope of this paper. We can also observe a crystallization of bigger parts of the cloud; for example, the cloud shown in figure 1(b) is nearly entirely crystalline. These situations emphasize the fact that studies of big structures can be investigated in the laboratory with dust density around 10^6 – 10^7 cm^{-3} .

3.5. Multiple generations

A very interesting result obtained in the laboratory is the multiple-generation phenomenon. This effect is characterized by successive growths of particles during the same run. In a situation where a cloud of grown particles occupies the whole volume of the chamber (except the void region), increasing the RF power leads to an increase of the size of the void region. A few minutes later (2 min approximately, the same as the typical growing time), a new generation of particles is grown in this empty central region. The new particles are grown from the first particle generation sputtered due to ion bombardment. This is deduced due to a visual decrease of the size of the first generation. The new particles continue to grow but as they are grown in a region surrounded by the first generation they cannot escape from this region. In consequence we can obtain situations where bigger particles are trapped near the centre of the plasma and surrounded by smaller particles. Successive increases and decreases of the RF power can thus lead to the coexistence of multiple generations of grown particles with different sizes as shown in figure 4. This gives a very interesting multicomponent situation with clear separations (sheaths) between the clouds.

4. Results obtained on board the ISS

The work performed in the laboratory allowed us to foresee experiments to be performed under microgravity conditions. Three experiments, each of length 90 min, have been programmed. These three experiments start with a common sequence (first 20 min) to look at the reproducibility of the phenomenon. These on-board experiments depend, obviously, on the possibility of

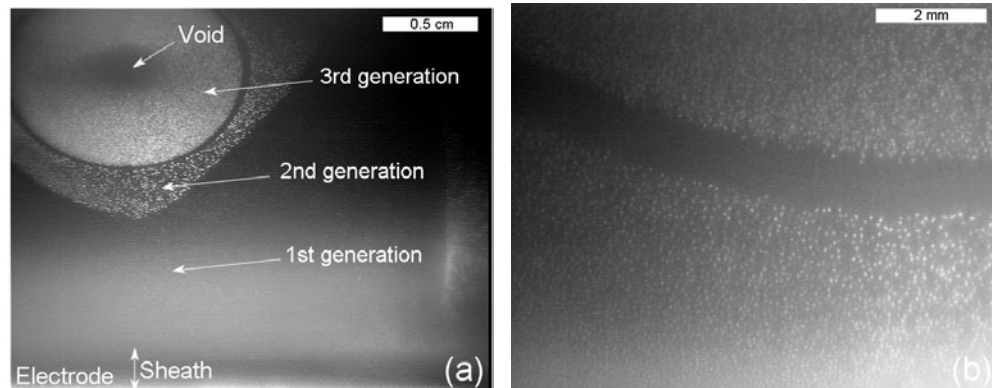


Figure 4. (a) Successive generations of grown dust particles; (b) HR camera showing the separation between a previously grown cloud (bottom) and a new growing one (top) in the void region (see also [movie](#)).

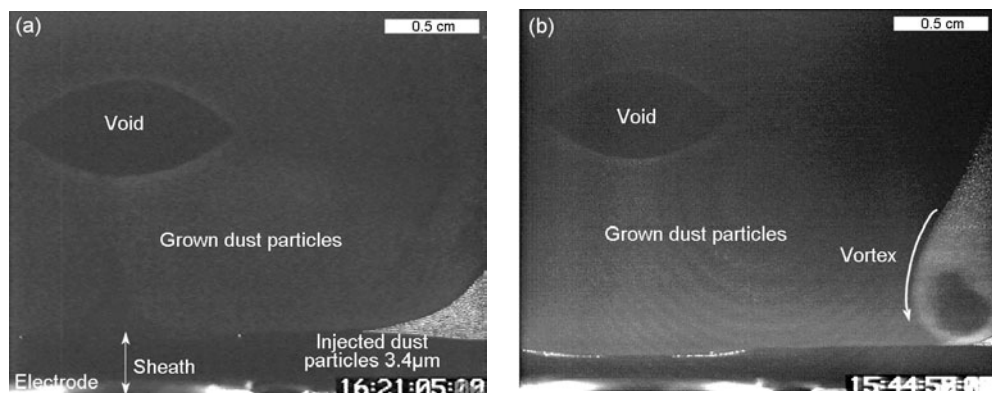


Figure 5. Dust clouds under microgravity conditions: (a) mixture of grown and $3.4 \mu\text{m}$ particles; (b) vortex-like structure near the electrode edge (25 frames superimposed).

growing dust particles. Indeed, one of the unknown parameters is the quantity of matter deposited inside the on-board chamber from previous experiments: injected particles do not fall down on the electrode when the plasma is turned off. As previously mentioned, another very important requirement is to start each run with a very low base pressure. Due to this last requirement the growth experiments have been performed with a chamber pumped by a turbomolecular pump instead of the direct connection to space already in use.

4.1. Dust particle clouds grown under microgravity conditions

Dust particle growth has been achieved on board the ISS [26]. A huge number of very small dust particles has been obtained. This situation, which has also been observed on the ground, is not the most common one and so far we cannot conclude if this effect is due to microgravity or to a high purity of the plasma and a lot of matter available. As we have seen on the ground two

different methods of growth are possible, a big number of small particles or fewer particles but with bigger sizes. Consequently the cloud obtained on board is constituted of a huge number of very small particles that cannot be individualized on images. Thus our study is focused on a global observation of the cloud.

We can notice that a first interesting result is the presence of the void region even with these small particles (figure 5(a)). On the lower right part of this figure we can see the $3.4 \mu\text{m}$ particles injected after the apparition of the grown cloud. The MF particles are pushed out to the periphery of the plasma by the grown particles. In figure 5(b) we can also observe a vortex cell of bigger grown particles. This situation emphasizes the importance of electrical effects on this phenomenon given that thermal convection does not exist under microgravity conditions. As this motion is observed near the sharp electrode edge it is certainly due to this strong electrical inhomogeneity. This fact is also confirmed by our on-ground experiments where an electrical step has been created by a small stainless steel plate lying on the lower electrode: the cloud of grown particles showed a vortex motion just above the frontier of this plate.

4.2. Influence of the dust particles on discharge characteristics

When a sufficient number of dust particles is present in a plasma discharge it is well known that the characteristics of the discharge are strongly modified [27]. In fact, as the dust particles attach free electrons of the discharge, the current reaching the electrodes is reduced. This effect has been observed during particle growth experiments (in day 1 and 2 of the on-board mission) through the evolution of the current fundamental harmonic shown in figure 6(a). This modification of the plasma resistivity is very similar to previous experiments performed in argon–silane plasma (figure 6(b)) where the fundamental and especially the second harmonic (at 40.68 Hz) are very sensitive to dust particle appearance [28]. The difference between the two situations is the typical timescale. In argon–silane the equilibrium is reached very quickly in only a few seconds while in our case it takes a few minutes. In figure 6(a) we have also plotted the evolution of the total intensity of the video signal recorded by the CCD camera. This value is composed of the laser light scattered by the growing particles and also by a part coming from the plasma itself. This last part is the enhancement of the plasma emission due to stronger excitation caused by the dust particles which modify the electron energy distribution function [27]. We can see a good agreement between the evolution of the video signal and the current harmonic.

4.3. Collective behaviour

The collective behaviour of the cloud can be studied through the excitation of the whole multicomponent cloud using a low frequency voltage (between 0.5 and 34 Hz) applied to the electrodes. This has been performed on the cloud shown on figure 5(a) constituted of grown and $3.4 \mu\text{m}$ particles. We have recorded as a function of time the position of characteristic points of this cloud (see figure 7). This analysis has been performed for frequencies between 0.5 and 5 Hz (higher values cannot be investigated due to the time resolution of the CCD camera at 25 Hz). The curves obtained are shown in figures 8(a) and (b) for two frequencies. We can see that a phase shift between the lower part of the cloud (micrometre particles near the electrode sheath) and the frontiers of the void region appears when the frequency increases. The excitation is applied to the electrodes and acts on the nearest layers. The perturbation then propagates through the cloud and reaches the void region with a time shift. The results for three different

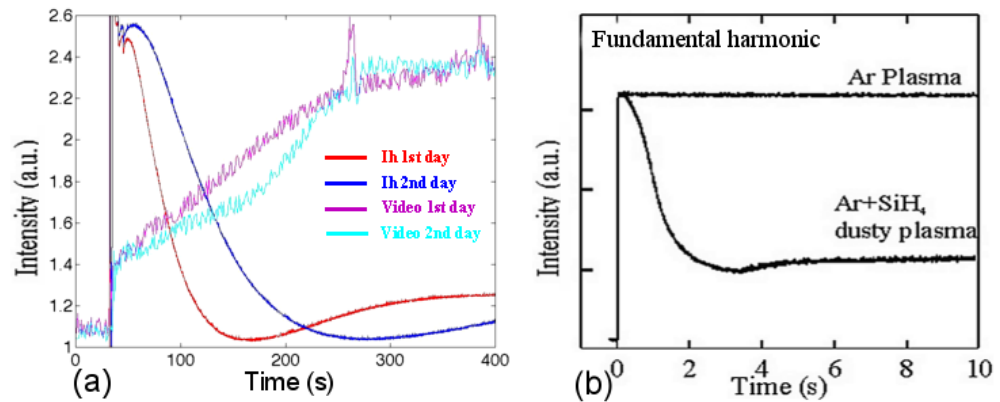


Figure 6. Evolution of the fundamental harmonic of the electrode current (a) during on board experiments correlated with the increase of the video signal intensities and (b) in argon and argon–silane plasma.

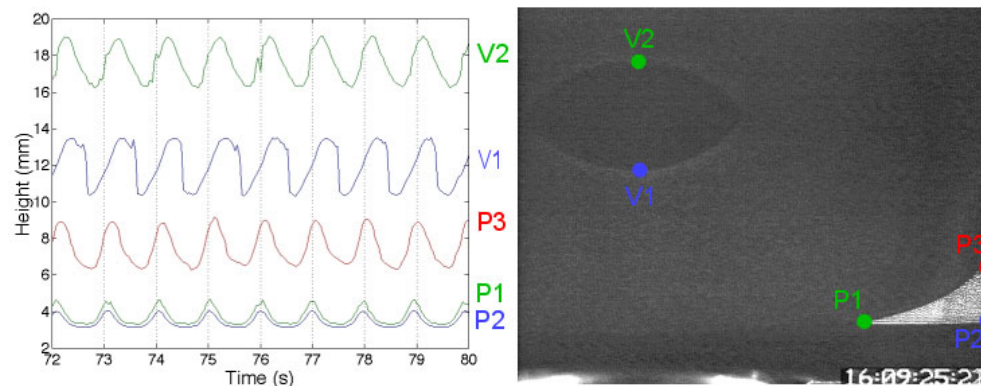


Figure 7. Time evolution of the height of characteristic points of the dust cloud under low frequency excitation (see also [movie](#)).

pressures are summarized in figure 8(c). We see that, as the pressure is increasing, the phase shift becomes higher even with small frequencies due to the fact that the wave propagation is slowed down by the gas friction. Similar experiments will be conducted on the ground to extract more information (like the dispersion relation) for comparison.

During the low frequency excitation, we have also observed an interesting behaviour traducing both attractive and repulsive interactions between dust particle clouds. Before the excitation a very small number of dust particles trapped in the upper part of the void region constitutes a small separated heap (figure 9). When the excitation is applied this small heap behaves independently of the rest of the cloud as shown in the movie. We can observe that the heap stays compact and cohesive in spite of collisions with the surrounding cloud. The collisions seem to be electrostatic, meaning that the heap is charged but the particles in the heap stay joined, perhaps emphasizing some attractive effects. Unfortunately, the video quality and the magnification are too poor to draw conclusions and new experiments will be conducted on the ground to try to reproduce this observation.

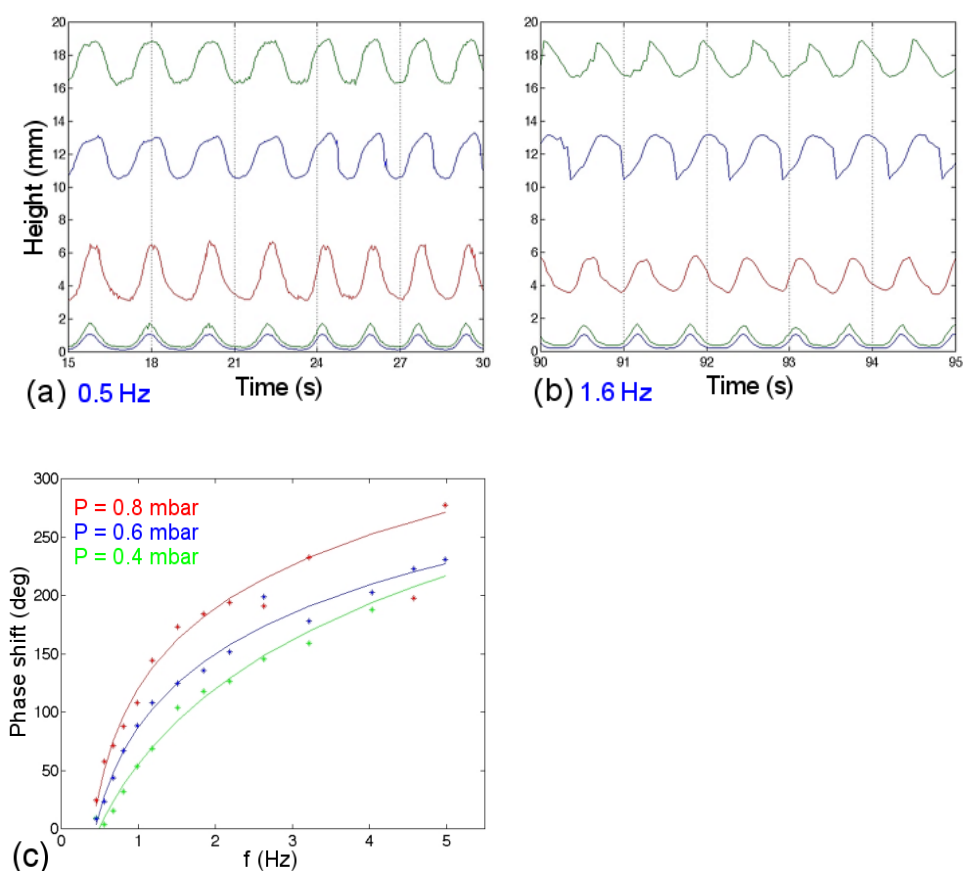


Figure 8. Evolution of the height of the characteristic points (a) for 0.5 Hz and (b) for 1.6 Hz and (c) the phase shift as a function of the frequency for three different pressures: 0.4, 0.6 and 0.8 mbar.

5. Conclusions

We have reported in this paper results concerning the growth of submicron dust particles in a sputtering discharge. These results have been obtained in the PKE–Nefedov chamber both in the laboratory and under microgravity conditions. These experiments show that a huge number of submicron carbon particles can be grown both on the ground and in space. This growth dynamics can be followed through the study of the current harmonics, especially during the first minute after the plasma ignition when the scattered laser light intensity is too small. However, a difference in the dust size kinetics has been observed. On the ground, after the first stage of growth, the particles seem to change in size more rapidly and during the whole run. In consequence, bigger particles have been grown in the laboratory. Until now it is not clear if this observation is directly related to microgravity conditions.

The void region does not exist during the first minutes of existence of the grown cloud. It appears after a few minutes and increases in size progressively even with these very small particles. This dust-free region can be the place of formation of a new generation of dust particles trapped by the surrounding cloud. The particles grown in this experiment are both sufficiently small to obtain three-dimensional clouds and sufficiently big to be observed by standard CCD cameras. This experiment gives the opportunity to study in the laboratory the formation and

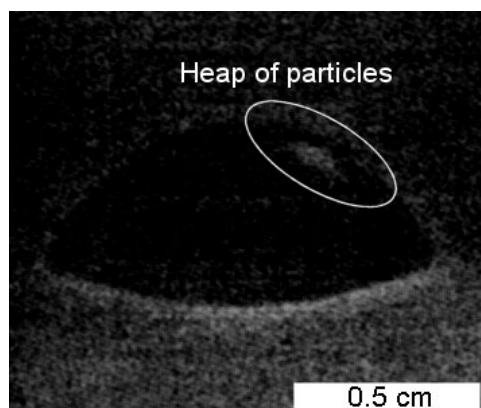


Figure 9. Small independent heap of particles generated in the upper part of the void region (see also [movie](#)).

behaviour of the void region.

The cloud of grown particles exhibits various collective effects like crystalline structure, vortex motions or formation of independent heaps of particles. All these phenomena are related to the dust particle charge and to the electrical fields existing in the plasma and near the electrodes.

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