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Formation, Growth, and Behavior of Dust Particles in a Sputtering Discharge

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Abstract. Dust particle growth instability and residual dust charges in a rf discharge have been studied. Dust particles were grown by sputtering of a polymer material previously deposited on the electrodes. We show that two regimes of the dust particle growth instability exist and are characterized by their frequencies. An empirical explanation based on the assumption of different dust particle growth rates is proposed for the existence of these regimes. We also show that dust particles do keep residual charges for a long time in afterglow plasma.

Introduction

Dusty or complex plasmas are partially ionized gases composed of neutral species, ions, electrons and charged dust particles. Dust particles are electrically charged due to their interactions with ions and electrons of the surrounding plasma [Havnes *et al.*, 1990; Matsoukas and Russell, 1995; Arnas *et al.*, 1999]. In laboratory plasmas, dust particles are negatively charged due to a higher mobility of electrons.

Dust particles in plasma were discovered in the 1920's by Langmuir *et al.* [1924] in a streamer discharge. These particles came from sputtered tungsten cathode. Until the late 1980's, dusty plasmas were not strongly studied except few astrophysical articles (see for example Havnes and Morfill [1984]; Havnes [1984]). In 1989, Selwyn discovered dust particles in IBM plasma processing reactor by using laser light scattering [Selwyn *et al.*, 1989]. These particles were directly grown in the plasma and were sources of wafer contamination. Dust particle growth in plasmas became thus of major concern and a better understanding of physical phenomena involved in dusty plasmas such as growing and charge processes was necessary.

In laboratory experiments, dust particles can be either injected or grown directly in the plasma. Injected dust particles are generally micron-size particles and due to their masses, they are confined near the bottom electrode where the electric force counterbalance the gravity. Microgravity conditions are necessary to study dust cloud of micrometer size particles filling the whole plasma chamber [Nefedov *et al.*, 2003]. In laboratory, dense clouds of submicron particles light enough to fill the gap between the electrodes can be obtained using reactive gases such as silane [Bouchoule and Boufendi, 1993; Cavarroc *et al.*, 2006] or using a target sputtered with ions from plasma [Praburam and Goree, 1996; Samsonov and Goree, 1999b,a; Mikikian and Boufendi, 2004; Mikikian *et al.*, 2003b].

During the growth of dense clouds of particles, plasma instabilities can occur. In argon-silane rf discharge, instabilities have been encountered during the growth of nanoparticles and can be observed on both amplitude of the third harmonics (40.68 MHz) of the discharge current and the self-bias voltage (Vdc) [Cavarroc *et al.*, 2006]. A proposed explanation was an attachment induced ionization instability as observed in electronegative plasmas. Instability during dust particle growth has also been found in sputtering discharge [Praburam and Goree, 1996; Samsonov and Goree, 1999b,a]. This instability can be divided in modes appearing consecutively: the filamentary mode and the great void mode. It was explained by a spontaneous fluctuation in the dust number density leading to a lower depletion of electrons in the region

of reduced dust particle density. This leads to a higher ionization rate in this region inducing two forces applied on the negatively charged dust particles: an inward electric force induced by the positive space charge (and thus electric field) and an outward ion drag force. The threshold for the instability is determined by particle size (charge) and the electric field strength. Similar instability in the PKE-Nefedov reactor where dust particles were grown by sputtering of a polymer previously deposited on the electrodes has also been observed by *Mikikian et al.* [2006] but the separation in modes was more complicated. In this paper, we report complementary measurements to *Mikikian et al.* [2006] experiments. We focused on studies at an argon pressure $P = 1.6 \text{ mbar}$ and injected power $P_W = 3.25 \text{ W}$. It has been found that the dust particle growth instability (DPGI) has two different regimes: the fast regime and the slow regime. These two regimes are characterized by their frequencies and can be early identified by looking at the amplitude of the first harmonic of the current.

Behavior of dust particles in dusty plasmas is determined by dust particle charge as it determines the interaction between a dust particle and electrons, ions, its neighboring dust particles, and electric field. Many articles are so devoted to charging processes in both space and laboratory plasmas (see for example [*Arnas et al.*, 1999; *Havnes et al.*, 1990; *Pavlu et al.*, 2005; *Samaritan and Vladimirov*, 2003]). Nevertheless, only few studies are devoted to decharging process [*Ivlev et al.*, 2003; *Couëdel et al.*, 2006]. In this paper, we expose new results we obtained [*Couëdel et al.*, 2006].

Experimental set-up and conditions

The work presented here is performed in the PKE-Nefedov (Plasma Kristall Experiment) chamber designed for microgravity experiments [*Nefedov et al.*, 2003]. It is a rf discharge operating in push-pull excitation mode. It consists of 4 cm diameter parallel electrodes separated by 3 cm. The injected power varies in the range 0 – 4 W.

Dust particles are grown in an argon plasma (0.2 – 2 mbar) from a sputtered polymer layer deposited on the electrodes and coming from previously injected dust particles (3.4 μm , melamine formaldehyde). A detailed description of this experiment and previous results are presented in Ref [*Nefedov et al.*, 2003; *Mikikian and Boufendi*, 2004; *Mikikian et al.*, 2003b, 2006].

The dust particle cloud is illuminated by a thin laser sheet and the light scattered by the dust particles is recorded by three standard CCD cameras at 25 images per second. Two of them record the light at 90° while the third one records at an angle of roughly 20° – 30°. This third camera permits to see the dust cloud in first stages of the growth when particles are very small.

For the study concerning the DPGI, two signals were measured: First the amplitude of the fundamental harmonic of the discharge current and second, the total emitted plasma light using five horizontally aligned optical fibers each one coupled with photomultiplier tube (PM). Each fiber was focused in a specified region of the plasma and has a spatial resolution about 3 mm. Electrical and optical signals were recorded simultaneously using an oscilloscope with 5kS/s sampling rate.

The measurements have been performed by the following routine. First the chamber was pumped down to the lowest possible pressure (base pressure $\sim 2.5 \cdot 10^{-6} \text{ mbar}$). Then, argon was injected up to the operating pressure $P = 1.6 \text{ mbar}$ and the discharge was turned on with an injected power $P_W = 3.25 \text{ W}$. Electrical and optical signals were recorded from the plasma lighting to plasma extinction.

For the study concerning residual charges, the top electrode was cooled down. An upward thermophoretic force was thus applied to dust particles in order to balance gravity [*Rothermel et al.*, 2002] when the plasma is off. To study particle charges, a sinusoidal voltage produced by a function generator with amplitude $\pm 30 \text{ V}$ and frequency of 1 Hz was applied to the bottom electrode. Induced low frequency sinusoidal electric field $E(r, t)$ generated dust oscillations if

they kept a residual electric charge. By superimposition of video frames, particle trajectories have been obtained. From oscillation amplitudes, the residual dust charges can be deduced

The dust particle growth instability

During dust particle growth, the amplitude of the first harmonic of the discharge current is decreasing (Fig.1) due to electron attachment on dust particles. Tens of second after plasma ignition, DPGI appears and can be well observed on both electrical and optical measurements [Mikikian *et al.*, 2006]. In electrical measurement, the beginning of the DPGI is characterized by a strong amplitude oscillation of the discharge current (Fig.1). Mikikian *et al.* [2006] showed that the light emitted by the plasma is also modulated and that the DPGI evolves following defined pattern: 3 ordered phases (P1, P2, P3) followed by a chaotic phase and other phases.

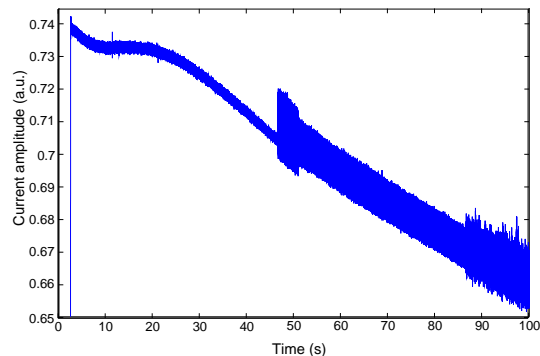


Figure 1. Discharge current amplitude during the first 100 s of the discharge

As it can be seen in fig.2, Fourier transforms of both electrical and optical signals exhibit these phases. It can be noticed that the frequency of the P3 phase (between 50 s and 90 s in the left diagrams of figure 2 or between 80 s and 140 s in the right diagrams) is lower on optical signals. Two DPGI regimes have been observed: a slow regime and a fast one with typical frequencies few times higher than the slow one. Nevertheless, these regimes are not strictly separated and DPGI can occur in a wide range of frequencies.

It has also been found that DPGI phase duration times are much longer for slow regimes than for fast ones. Figure 3 shows that the higher is the frequency of the P3 phase, the shorter is its duration.

As the first phase P1 can be very short (less than 0.1 s), it can not be used to build statistics. For this reason, the appearance time as a function of P2 frequency (phase around 48 s in the left diagrams of figure 2 or around 60 s in the right diagrams) has been plotted (fig.3). It shows that for high frequencies the DPGI appears faster than for low frequencies.

Looking at the first 30 s of the discharge (before DPGI), the current amplitude exhibits also different pattern (fig.4). In the case of fast regime DPGI, the current amplitude firstly decreases during ~ 5 s then stabilizes for ~ 10 s and finally decreases again. In the case of slow regime DPGI, the current amplitude do not decrease immediately after plasma ignition. After ~ 1 s, it starts to decrease but very slowly compared to fast regime DPGI and after ~ 5 s starts to increase. Finally after ~ 15 s, the current amplitude decreases again but slowly compared to fast regime DPGI.

A possible explanation for the differences between the two regimes is a variation of the dust density (n_d)/dust size (r_d) ratio in the plasma from the first instant of the discharge. Indeed, it has been shown that dust particle growth is very sensitive to gas purity [Mikikian *et al.*, 2003a]. Thus slight differences in gas purity from one measurement to another can induce big change in dust density. In case of a slow regime DPGI, a reduced dust growth rate is assumed. Electron attachment on dust particle is smaller (smaller dust particles density and/or smaller particles) and thus the decrease of current amplitude is not as important as in case of a fast regime DPGI in which the growth rate is bigger. Another fact indicates that the dust density for the slow regime is lower: the small frequencies of the DPGI phases. As these frequencies are less than 100 Hz, they should be related to dust plasma frequency which is proportionnal to $\sqrt{n_d/r_d}$. Thus if this ratio is small, the dust frequency is also small. Finally, Samsonov and Goree [1999a] and Mikikian *et al.* [2006] proposed that the DPGI begins when the ratio n_d/r_d reaches a critical value. Thus, for reduced growth rates it takes a longer time to reach this critical value.

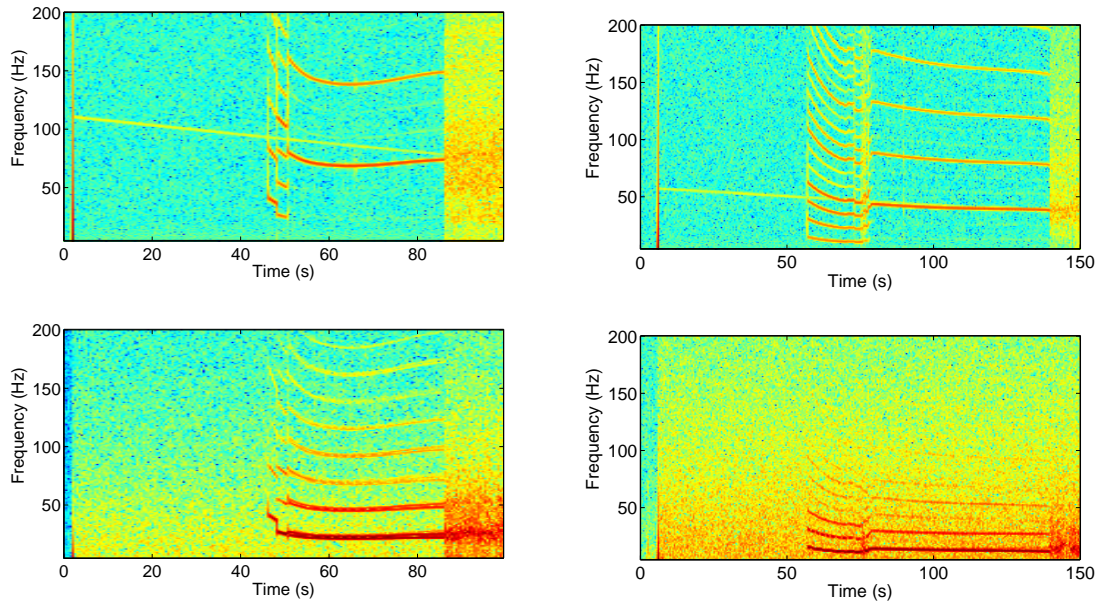


Figure 2. Left: Electrical signal Fourier spectrum of the current amplitude (Top) and Optical signal Fourier spectrum (Bottom) for fast regime DPGI. Right: Electrical signal Fourier spectrum of the current amplitude (Top) and Optical signal Fourier spectrum (Bottom) for slow regime DPGI.

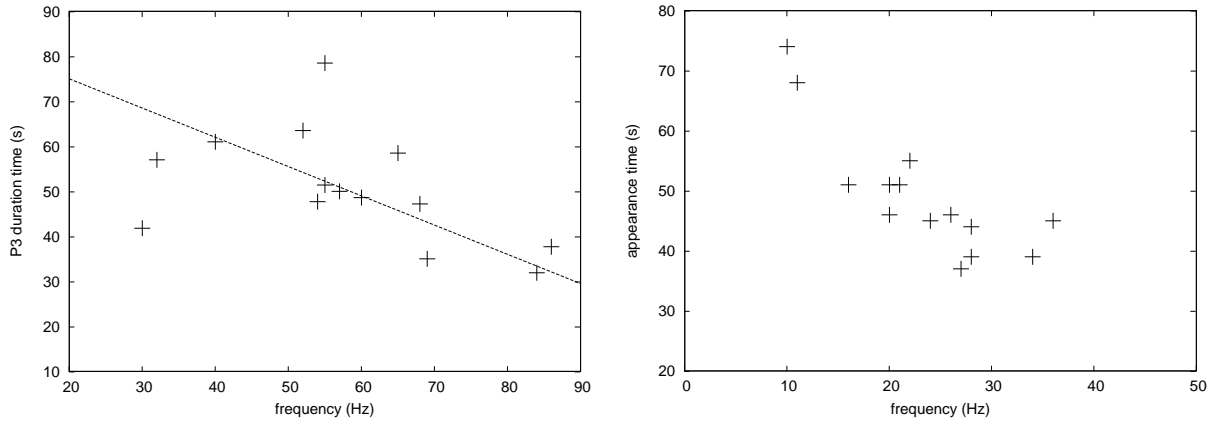


Figure 3. Left: P3 duration as a function of its mean frequency. Right: Growth instability appearance time as a function of P2 mean frequency

This agrees with the fact that lower frequencies correspond to longer DPGI appearance time. *Mikikian et al.* [2006] also reported that for low argon pressure when dust particles are difficult to grow [*Mikikian et al.*, 2003a], DPGI appearance time is longer, confirming our hypothesis.

Residual electric charge on dust particle

After the discharge was switched off, some dust particles were sustained in the discharge by the thermophoretic force which compensate their weight. A thermal gradient of 6 K between the electrodes permitted to sustain $r_d \sim 200$ nm radius particles with mass $m_d \sim 7 \cdot 10^{-17}$ kg. As it can be seen in Fig.5, the low frequency sinusoidal electric field created by the low frequency sinusoidal voltage applied to the bottom electrode by a function generator induced oscillations

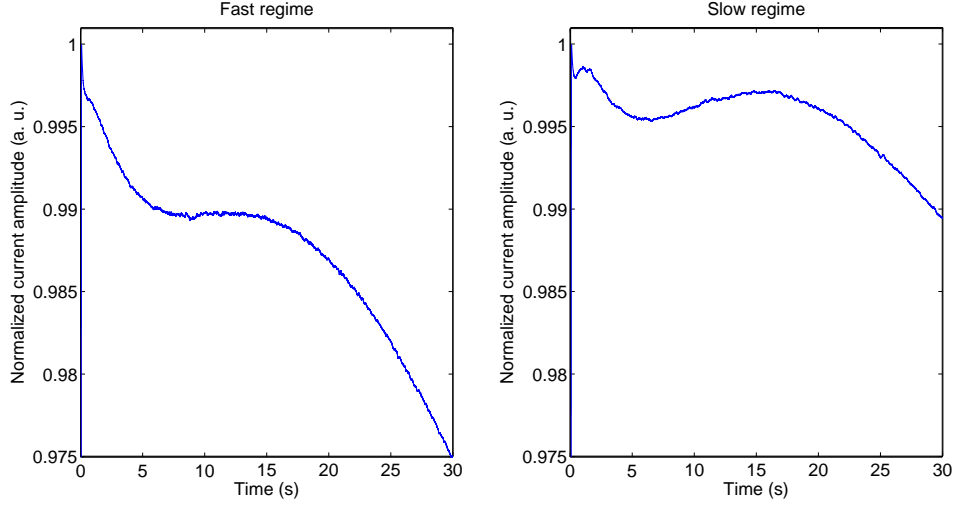


Figure 4. Left: Normalized fast regime current amplitude. Right: Normalized slow regime current amplitude

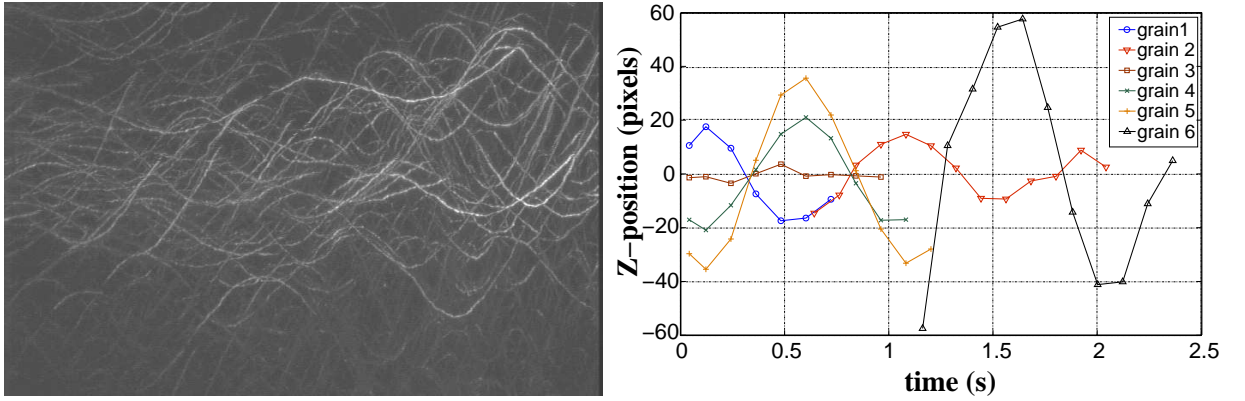


Figure 5. Left: Superimposition of video frames. Right: Reconstructed particle trajectories

of dust particles. This indicates that dust particles keep a residual electric charge in discharge afterglow. From oscillation amplitude b of a dust particle, its residual charge Q_d can be obtained [Couëdel et al., 2006]:

$$Q_{d_{res}} = \frac{m_d b(\omega, Q_d, E_0(z_{mean})) \omega \sqrt{\omega^2 + 4\gamma^2/m_d^2}}{E_0(z_{mean})} \quad (1)$$

where γ is the damping coefficient, $E_0(z_{mean})$ the electric field at the dust particle mean height z_{mean} and $\omega = 2\pi f$ where f is the frequency imposed by the function generator.

It has been found that positively charged, negatively charged and non charged dust particles coexist during more than one minute in the discharge chamber after plasma extinction. Measured residual charges were in the range $[-12e : +2e]$ depending on experimental conditions. Residual charge exists in both cases when the function generator was switched on during the discharge or after a few seconds the discharge is turned off. We proposed in Couëdel et al. [2006] that residual charges on dust particles are due to the influence of dust particles on plasma loss process.

Conclusion

Measurements of discharge current amplitude and total light emitted by the plasma have been performed for dust particle growth instability in the PKE-Nefedov reactor at an argon pressure $P = 1.6 \text{ mbar}$ and an injected power $P_W = 3.25 \text{ W}$. We show that the DPGI can exist in two regimes characterized by their frequencies. We proposed that these regimes are related to the growth rate which is strongly related to gas purity which can vary from one experiment to another and change the ration n_d/r_d which is a key parameter in DPGI appearance.

Measurements in discharge afterglow also permit us to observe and measure dust residual charges after plasma extinction. These residual charges can be either positive or negative and are of few electrons.

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