Dust cloud dynamics during particle successive generations

Marjorie Cavarroc, Maxime Mikikian, Yves Tessier, Laïfa Boufendi

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M. Cavarroc, M. Mikikian, Y. Tessier, L. Boufendi

GREMI, 14 Rue d'Issoudun, BP 6744, 45067 Orléans cedex 2, France

marjorie.cavarroc@univ-orleans.fr

Silane-based plasmas are widely used to deposit nanostructured silicon thin films or to synthesize silicon nanoparticles. Dust particle formation in Ar/SiH_4 plasmas is a continuous phenomenon: as long as silane precursors are provided, new dust generations are formed. Successive generations can be monitored thanks to various electrical (V_{dc}/3H) and optical (OES, video imaging) diagnostics. Experiments presented in this paper have been performed in a capacitively-coupled radiofrequency discharge, at low pressure (12 Pa) in an Argon/Silane mixture (92:8).

Evidence of dust particle successive generations

Thanks to correlation between 3H (current third harmonic), Laser Light Scattering (LLS) and Optical Emission Spectroscopy (OES), dust particle successive generations can be evidenced. They appear as a low frequency oscillation in the signals, with a period of typically one minute (see fig.1). In order to explain the behavior of the curves, the existence of a void region (dust-free region) is assumed and four parts are identified. In part I, the increase in $I_{750.38}$ corresponds to the opening of a void region, where the ionization rate is enhanced and where a new dust particle generation grows. New dust particles are neither big enough nor numerous enough to be detected by LLS. However, they push away bigger (highly charged) ones from the discharge center. It causes a net increase of the free electron density in the discharge, leading to 3H increase. In part II, newly created dust particles grow in the void region and become detectable by LLS. They also start to attach free electrons due to their increasing size. They keep on pushing bigger dust particles away, and tend to fill the whole void. There is still an increasing amount of free electrons in the plasma, but the net increase is lower than in part I, and explains the slope modification in 3H. In part III, the new generation is still growing and now fills the void region. It is now clearly detected thanks to LLS. The net increase in the free electron density

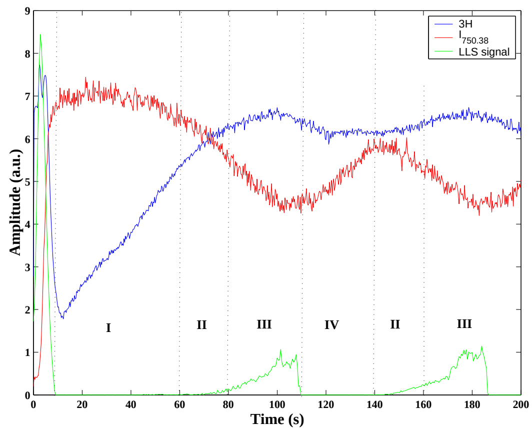
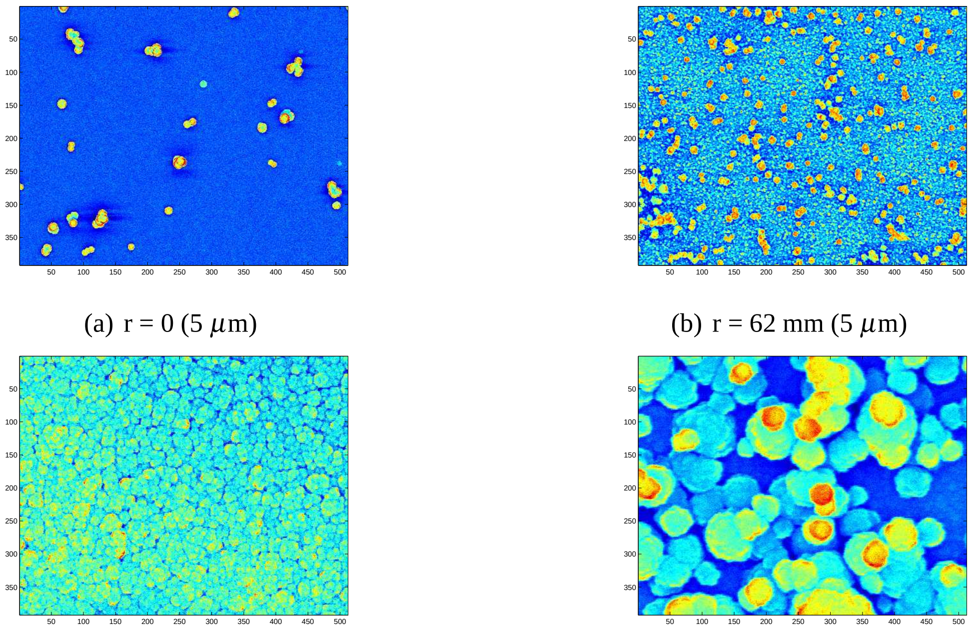


Figure 1: 3H, LLS and OES versus time during dust particle successive generations



(a) r = 0 (5 μm) (b) r = 62 mm (5 μm)

(c) r = 0 (200 nm) (d) r = 62 mm (200 nm)

Figure 2: SEM images of the spatially resolved depositions. Electrode center is located at r = 0, and edge at r = 62 mm. The image scale is indicated in brackets.

is less and less important due to the strong attachment on the growing particles. In part IV, a new void is opening in the discharge center. An equilibrium is reached between free electrons recovered from dust expellation and those lost by attachment. A new generation starts to grow in the void region. The phenomenon is cyclic and starts again with part II, III and IV, and so on...

Evidence of a void region

The first correlation between 3H, the LLS signal and the OES one lets think that a void region could appear in the dust cloud. Indeed, the increase in the ionization, before a new generation is detected by LLS, is typical of a void region. Thanks to video imaging, this region of higher ionization can be evidenced, confirming the results obtained by OES. Some spatially resolved depositions have been performed during the successive generations. Figure 2 gives an overview of the results (SEM images). From figures 2(a) and 2(b), we conclude that few "big" dust particles are present in the discharge center (r = 0), while more and more are found when the discharge radius is scanned towards the edge (r = 62). From figures 2(c) and 2(d), we conclude that lots of "small" particles (around 10 nm in diameter) are deposited in the discharge center, while the deposition becomes less and less dense toward the plasma edge, with nanoparticles bigger and less numerous.These results are totally consistent with a void region, where new dust particle generations would grow.

Successive generation instability (SGI)

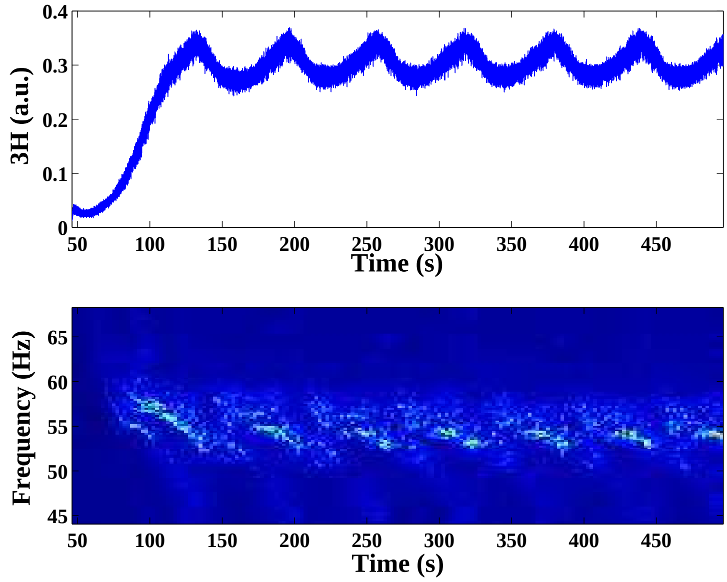


Figure 3: (a)Time-evolution of the third harmonic of the discharge current during dust particle successive generations in an argon-silane plasma (b) Spectrogram of 3H during the successive generation instability.

If we have a look to the inner structure of the 3H signal during the oscillation due to dust particle growth, we can bring to the light the unstable behavior of the signal (see figure 3).Tis instability is self-excited (no external excitation) and lasts as long as the plasma is on. Its beginning occurs generally a few seconds after the plasma ignition. The setting-up of this instability appears as an increase in the oscillation amplitude at the beginning of the phenomenon. The onset of the instability exhibits a very complex scheme with several frequency-branches in the spectrogram. Actually, we can determine that the instability is mainly characterized by two very close frequencies that become closer and closer. These two close frequencies could explain the noticeable modulation of the electrical signal that we observe. This complex scheme does not depend on experimental parameters. It is the same whatever the conditions and is very reproducible. When performing a time-resolved FFT of the 3H signal, we obtain a spectrogram as the one in figure 3. Typical frequencies are comprised between 40 and 60 Hz. This spectrogram shows an alternance of very ordered phases and less ordered ones. The ordered phases seem to correspond to dust particle formation for each new generation. During theses phases, the instability frequency linearly decreases on an interval of about 10 Hz. The less ordered phases seem to correspond to dust expelling from the plasma. During these phases, the frequency is not well-defined, but is globally higher than the one of the ordered phases. Using a high-speed video camera to record the plasma glow, we found quite similar results in the optical measurements. Moreover, some strong intensity decreases (over one or two frames) of the central region can even been evidenced. The SGI is globally not very sensitive to experimental parameters compared to the aggregation instability previously studied in [1, 2]. As a matter of fact, no case of disappearance of the instability has been observed by modifying the experimental conditions. As soon as a second dust particle generation starts to grow in the plasma, the instability arises. It seems to be an intrinsic characteristic of dust successive generations in Ar/SiH_4 low-pressure plasmas. Nevertheless, some slight modifications in the instability frequencies (always lower than 100 Hz) have been observed, depending on the experimental conditions. The frequency of the instability is quite difficult to determine.

When the injected rf power is increased, the frequencies of the instability tend to increase. We can also notice that the instability begins earlier for higher injected powers as expected from our experience. On the contrary, these same frequencies tend to decrease when the silane flow rate in the discharge is increased. The gas temperature effects are even more difficult to border. It seems to have no effect on the frequency of the high-ordered phases while it seems that the frequency of the less-ordered phases increases when decreasing gas temperature. Furthermore, pressure does not seem to have any outstanding effect on the instability behavior.

Discussion

We evidenced the presence of a void region in the dust cloud, where new dust generations grow, pushing previously formed and bigger dust particles toward the plasma edge. The SGI are closely linked to dust particle successive generations. The high-ordered phases correspond to new dust formation, while less-ordered ones correspond to the expulsion of bigger dust particles toward the plasma edge.

The formation of a new dust generation highly modifies the ionization rate, and thus the ion drag force, in the void region. The equilibrium between the ion drag and the electric force becomes then unbalanced, leading to an oscillation of the void region as already observed in other dusty plasmas [3]. A very similar phenomenon has also been observed in the PKE experiment, where the void region tends to be unstable when new dust generations grow inside.

References

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