ELECTRICAL DIAGNOSIS OF RADIOFREQUENCY DUST-FORMING PLASMAS

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Abstract
The aim of this paper is to present a powerful diagnostic tool which allows the detection of dust-particles formed in a plasma when their size is of the order of 2-3 nm. It is based on the analysis of the discharge impedance. The method involves the determination of the current up to the fifth harmonic, the voltage and the current - voltage phase-shift, using a non-perturbative system for the electrical measurements. Here the first results obtained in an argon-silane discharge are presented. This method has a temporal resolution in the microsecond range and is very easy to implement and can thus be used for industrial reactors.

Introduction
Much work has been carried out on dust particles nucleation and growth in low pressure plasmas used in microelectronic devices fabrication. They have been recognized as the main source of reactor contamination and the principal limiting factor for a-Si:H deposition especially when attempting to increase deposition rate. However, in the last few year it has been shown that if nanometer size crystalline particles are incorporated in an amorphous matrix, they provide very interesting opto-electronic properties to the grown layers[1,2]. The properties of such layers strongly depend on the size and concentration of these crystallites. In order to correlates the film characteristics it is very important to understand the nucleation and growth process and to in-situ detect the appearance of these entities.

Many techniques have already been used for this purpose such as laser induced explosive evaporation (LIPEE)[3] or LLS coupled with a Photon counting system[4]. These methods provide useful information about the dust appearance, size and concentration and also insight on the different growth phases. However, they require view ports in the reactor, not often available in industrial equipment, and they are quite difficult to implement.

In this paper we present a method based on the analysis of the radio-frequency (RF) discharge impedance which enables detection of dust particles appearance when their size is in the range of 2-3 nm. In the last ten years much work has focussed on the impedance of the RF discharges in order to measure the electrical parameters of the plasma[5,6]. The discharges considered in this work are non-symmetrical and capacitive-like. They present two or more sheaths where the relation between the RF voltage and the RF current is non-linear. This non-
linearity induces the appearance of harmonics in the discharge current which can give significant information about the electron and ion dynamics[7,8]. This dynamics is affected by the dust particles appearing in the plasma bulk. In this study, this effect is used to develop this diagnostic which is robust, non perturbative and well adapted to be implemented on the industrial reactors.

**Experimental set-up**

The experimental set-up used in this work has already been described in detail elsewhere[9]. The radio-frequency (13.56 MHz) discharge is produced in a grounded cylindrical box (13 cm inner diameter) equipped with a showerhead type RF powered electrode. The bottom of the box is closed with a 20 % transparency grid. This allows a vertical laminar gas flow. The discharge structure is surrounded by a cylindrical oven which allows the gas temperature to be varied from room temperature up to 200 °C. The gas temperature is measured in the gas flow below the bottom grid by means of a J-type thermocouple. Three vertical slits (2 mm width, 4 cm height) allow the optical access to the plasma at 0°, 90° and 180° around the chamber. The whole system is enclosed in a vacuum vessel of 30 cm height and inner diameter. Three optical windows on the vacuum vessel (5 cm in diameter, placed 90° apart) are aligned with the slits. The experimental conditions used in this work were as follow : 30 sccm of Ar, 1.2 sccm of SiH₄, total pressure of 12 Pa and an RF power of 10 W corresponding to 600 V peak to peak.

The RF voltage \( V(t) \) is measured using a capacitive probe placed at the connection between the coaxial cable and the RF electrode. The RF current is measured with a 250 MHz bandwidth current probe. The two signals are then processed using a home-made electronic board which allows to record the time evolution of the amplitudes of the voltage, the fundamental and third harmonics of the current and also voltage-current phase shift. The slopes are visualized on a Lecroy scope LC684DXL (1.5 GHz bandwidth and 8 GSs).

![Harmonics of the RF Current](image)

**Fig. 1 : Harmonics of the RF Current**

**Results and discussion**

Figure 2 shows the time evolution of the third harmonic for a pure argon and dust forming discharges over 0.4 second. It clearly that these two situations are quite different. In the first situation there are no particles occurring in the plasma and then the amplitude of this component of the current remains constant. However, in the second situation the amplitude is
affected by the appearance of the dust particles in the earlier stage of their formation. This behavior has been recorded at room temperature and the time $t_1$ at which we start to observe a modification of the amplitude is about 70 ms. The same situation was observed for the first harmonic. Nevertheless, we know from previous works on the nucleation and growth of the dust particles that at that time the coalescence phenomenon has not yet started and the particle size is about 2 nm. The typical time for the formation of such particles has been shown to be of the order of 1 ms at room temperature[10].

The amplitude decreases linearly from $t_1$ to $t_2$, where slightly increases before a faster decrease. This can be related to the modification of the electron collision frequency induced by the appearance of the dust particles. In fact, when the particles appear in the plasma, in the electron collision frequency equation it is necessary to add a term which takes into account the electron-particle collisions and that can be expressed as

$$N_p \sigma_{e-p} v_e$$

where $N_p$ is the particle concentration, $\sigma_{e-p}$ the collision cross section and $v_e$ the mean thermal electron velocity. From this we can conclude that the concentration of the particles increases linearly. During this phase, less than 1% of the particles are charged and trapped through a charging-recombination process[10,11].

The second time $t_2$ corresponds to the moment where the dust particle concentration reaches a critical value of about $10^{11}$ cm$^3$ from which the coagulation phenomenon starts[3]. By knowing the total number of argon atoms and the electron temperature $T_e = 2$ eV we can estimate the contribution of the powders to the electron collision frequency. It represents about 3% of the total frequency. That is why the first decrease, between $t_1$ and $t_2$, is so small.

When the coalescence process starts, the particle number density decreases and then their contribution to the collision frequency decreases. This explain the increase observed after $t_2$. As the particles grow up in the plasma bulk, they can attach more than one electron. Thus, the drastic decrease of the amplitude after $t_2$ corresponds in fact to the drop of the electron density[12]. In this last situation, all the plasma characteristics are strongly modified. This is well known and called the $\alpha \rightarrow \gamma'$ transition.

However, during the first phase, from $t_1$ to $t_2$, the sheath thickness remains constant and thus the related electrical capacitance. This can be easily followed on Figure 3 which also shows the time evolution of the voltage – current phase shift (curve b). This parameter is mainly
determined by the capacitance of the sheaths. Up to $t_2$, the phase shift remains constant. This means that the first nanoparticles have no appreciable effect on the argument of the electrical impedance of the plasma before the coalescence phase. After $t_2$, it means after the beginning of the coalescence phase, the current-voltage phase shift drastically decreases as already observed in previous studies[9].

If the gas temperature is increased, while keeping constant the total number of molecules and atoms which are injected in the discharge box, the particle formation is known to be delayed[3]. The time evolutions of the amplitude of the third harmonic for different gas temperatures have been recorded (fig.4). This result is in good agreement with the one obtained by LIPEE[3] where it has been shown that the particle nucleation and growth are very sensitive to the gas temperature. It is clear from figure 4 that even at high temperature, condition where we expect to grow amorphous layers we can have nanoparticles formed in the plasma. However their concentration never reaches the critical concentration to give bigger dust particles.

Figures 5a and 5b give respectively the evolution of $t_1$ and $t_2$ with respect to the gas temperature. The evolution of $t_1$ indicates that the effect of the temperature on the first phase of particle nucleation and growth is linear. Studies are now under way to explain clearly this observation. However, figure 5b gives the same existence diagram presented in the reference[9]. It defines the conditions where we can avoid the formation of particles with respect to the gas temperature.

![Fig. 5](image)

Conclusion

In this paper we presented a method to detect the dust particle formation in a plasma. Here we studied a typical silane based discharge used to grow amorphous and polymorphous silicon thin films. It is clearly shown that this detection concerns particles in the earlier phase of their formation having a mean size of about 2 nm. It emphasizes the different growth phases. This robust diagnostic has a time resolution of less than 1 μs and is very easy to be implemented on industrial reactors.
References