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Study on interaction between plasma fluctuation and nucleation of nanoparticles in plasma CVD
プラズマCVDにおけるプラズマ揺動と
ナノ粒子の核形成間の相互作用に関する研究

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We have investigated effects of plasma fluctuation on the growth of nanoparticles in capacitively-coupled rf discharges with amplitude modulation. Nanoparticles grow more slowly for higher AM levels, that is they have 7 times higher density and 23% smaller size. This AM method is useful for the production of a large amount of nanoparticles with a small size.

1. Introduction

Studies focused on reducing the dimensions of semiconductor structures [1] to the nanometer regime have led to a fascinating class of novel materials that exhibit quantum confinement characteristics. These materials are referred to as nanoparticles or quantum dots. Although such nano-composite materials with quantum dots allows us to fabricate high-efficiency optoelectronic devices [2-4], efficient methods for synthesizing large numbers of small non-agglomerated nanoparticles with a narrow size distribution are required. Plasma processing can be a good candidate method for synthesizing such nanoparticles in the gas phase of reactive plasmas [5-7]. For faster development of fabrication methods for nano-materials, effects of plasma fluctuations on the growth of nanoparticles in reactive plasmas must be taken into account because plasma fluctuations affect nanoparticles in dust plasmas [8]. Investigation of the effects leads to control the growth of nanoparticles and to improve the fabrication technology of nano-materials. Moreover, effects of plasma fluctuations on the growth of nanoparticles in reactive plasmas have not been reported yet. In order to study the effects of plasma fluctuations on the growth of nanoparticles, we use the amplitude modulation (AM) method [9], which can control the plasma fluctuation level in reactive plasmas [10].

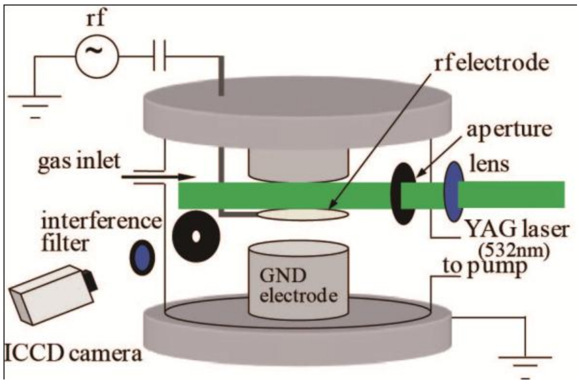

Fig.1 Experimental setup. In order to monitor the growth of nanoparticles, the nanoparticles are illuminated by SHG of CW YAG laser light (λ = 532 nm, 2.0 W), and the scattering light is detected using an ICCD camera.

In the present paper, we report observation results of the effects of plasma fluctuations on the growth of nanoparticles in reactive plasmas using an in-situ laser-light scattering (LLS) method. Moreover, we succeed in the production of a higher density of smaller nanoparticles than the conventional method without using AM [7], and we will discuss the mechanism of the effects.

2. Experimental setup

Experiments were performed using a capacitively coupled rf discharge reactor with an in-situ LLS system, as shown in Fig. 1[10]. The inner diameter and height of this reactor were 260 mm and 230 mm, respectively. In the reactor, a powered disc electrode with a diameter of 60 mm and a thickness of 1 mm was placed 20 mm below an upper grounded electrode with a diameter of 60 mm located 20 mm above a lower grounded electrode with a diameter of 60 mm. The reactor was evacuated to a base pressure of less than 10^{-3} Pa with a rotary pump and a diffusion pump. The flow rates of $Si(CH_3)_2(OCH_3)_2$ and Ar were 0.2 and 40 sccm, respectively. The total gas pressure in the reactor was 133 Pa. The temperature of the reactor wall was maintained at 373 K in order to avoid liquefaction of $Si(CH_3)_2(OCH_3)_2$. In order to generate nanoparticles, we sustained a discharge by applying 150 peak-to-peak voltage V_{pp} of 60 MHz to the powered electrode for a discharge period of T = 8 s. The corresponding discharge power was 30 W. The self-bias voltage V_{dc} was 0.4 V. The plasma parameters of electron density n_e and floating potential V_f were measured using an rf-compensated Langmuir probe. The typical electron density n_e was approximately 10^{10} cm^{-3}.

3. Experimental results and discussion

Plasma fluctuations can be synchronized with the AM of rf discharge voltage [19]. Figure 2 shows that AM level dependence on LLS intensities at t = 8s at plasma/sheath boundary region near the upper grounded electrode. Here, a modulation frequency f_{AM} is 10 kHz. The LLS intensity with AM is smaller than ones without that. The LLS intensity decreases by 60 % with increasing the AM level up to 30%. This result shows AM of rf discharge voltage suppresses the growth of nano-particles.

In order to clarify the growth suppression mechanism, the size and density of nanoparticles were determined based on the LLS intensity [11]. Figure 3 shows the dependence of the size and density of nanoparticles on the AM level. The nanoparticle density increases by 7 times from 2.3×10^9 cm^{-3} for the 0% AM level to 1.6×10^{10} cm^{-3} for the 30% AM level, whereas the nanoparticle size decreases by 23% from 12.5 nm for the 0% AM level to 9.7 nm for the 30% AM level. In Ref. [9], Y. Watanabe *et al.* controlled the size and density of nanoparticles by varying the rf discharge power and/or discharge duration. Here, we show that amplitude modulation of the rf discharge voltage is another tuning knob for controlling nanoparticle size and density.

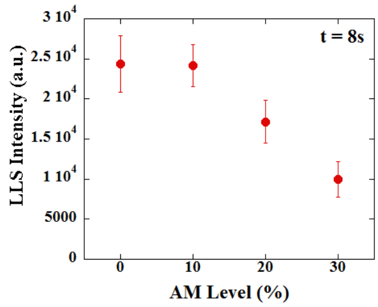

Fig.2 AM level dependence on LLS intensities at t = 8s at plasma/sheath boundary region near the upper grounded electrode. AM was carried out with a sine wave of 10 kHz.

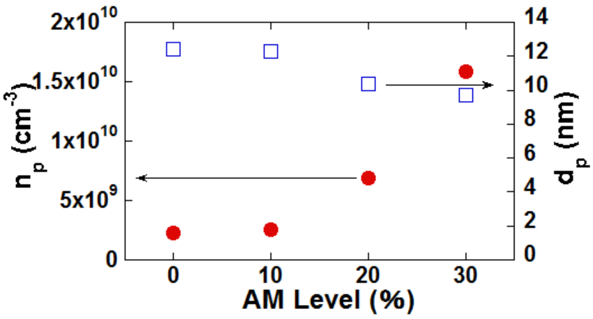

Fig.3 Amplitude modulation level dependence of the density and size of nanoparticles.

Acknowledgements

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