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Particle Growth in an Experimental Dusty Plasma System *

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The coagulation and growth process of dust particles is investigated through laboratory experiment in a plasma system. A large number of dust particles with different sizes and shapes are formed. The growth process is characterized by the scattering laser intensity and fractal dimension. The comparisons of dust particles and scattering laser intensity obtained at different rf powers are presented. The three-dimensional distribution of dust particles is also given. These results provide an experimental basis for dust growth investigation.

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Dust particle contamination is an inevitable problem in semiconductor processing. Selwyn *et al.*[1,2] found that there were some particles suspended at the sheath boundaries in certain silicon etching plasmas, and the parametric conditions of the growth of these particles were consistent with the important parameters of the silicon etching process. Once the process was completed, the particles fell onto the surface of the chips, resulting in damage. This discovery provided a strong impetus for studying dusty plasmas. Theoretical models[3–5] for understanding of the growth of dust grains have been developed using the data produced by experimental observation. Based on the proposed theory, Olevanov *et al.*[3] explained the experimentally observed threshold behavior of the coagulation process and estimated the critical microparticle size for the phase transition from gas to coagulation. By comparison of experimental results with computer simulation, Hayashi and Tachibana found the growth of the particle size by coating with hydrogenated amorphous carbon.[4] Matthews and Hyde simulated the coagulation of dust aggregates under a charge-dipole interaction using a numerical model.[5]

In general, there are three stages of growth for dust grains experimentally.[3,6,7] The first is the initial growth stage. In this stage, small dust particles (∼10 nm) are produced by chemical reaction within the discharge chamber. They are almost spherical in shape, and dust clouds formed by a large number of small particles can be observed under a microscope. The second stage is the coagulation stage. In this stage, collisions between dust grains can generate new particles of different sizes and shapes. Depending on initial conditions, formation of dust chains and fractal dust grain structures can be seen. The final stage is the saturation stage. As the size of the dust grains increases continuously, the growth rate slows down and eventually shows saturation. Depending on the experimental parameters, this process can be as rapid as a few seconds or as slow as a few hours.[3] In all cases, for agglomeration to occur, the charged dust grains must have sufficient kinetic energy to overcome the electrostatic potential barrier. Praburam *et al.*[8] investigated dust growth in an ion sputtering experiment using graphite targets, where island formation through accretion of carbon atoms onto the electrode surface was observed. The overall linear growth rate was about 15 nm/min, with islands reaching 300 nm after 20 min of plasma exposure. Their experiment showed that the spherical particulates, which were too small to be imaged individually, formed as a dust cloud. Huang *et al.*[9] experimentally showed that dust particles can grow by up to about a millimeter in size. The dust particles were negatively charged and suspended by the sheath electric field and were illuminated and detected by a laser beam.[10,11] The investigations of Prabhakara and Tanna have shown that the scattered intensity could be due to either the dust density or the size of the particles, or both.[11] Although different patterns were observed during dust growth,[8,12,13] more detailed investigations are necessary to understand the mechanism of the dust growth process. In this Letter, we investigate the dust growth process using the scattering laser intensity together with the fractal dimension.

The experiment discussed here was conducted in a capacitively coupled rf discharge dusty plasma system. The powered electrode is a cylinder with a cone-shaped hole at the center, and the grounded electrode is a hollow, thin-wall cylinder surrounding the powered electrode. The dusty plasma is produced by chemical synthesis reaction of silane, argon and ethylene mixture. The background pressure is less than 5 Pa. Once the gas mixture is in the chamber, the rf

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power is adjusted to be no less than 10 W to excite discharge. The dust grains are illuminated by a He-Ne laser beam sheet. The distribution of dust grains in three dimensions can be viewed by adjusting the height of the laser beam. The growth of dust grains is monitored and digitally recorded by a microscope and CCD camera using a computer with an image-processing card and frame grabber software. In addition, the scattered light is collected with an optical fiber with the diameter of 0.5 mm and the intensity is measured by a spectrometer. The angle between the optical fiber and the horizontal plane is fixed at 60° (pointing to the center of CCD vision). The experimental setup is shown in Fig. 1.

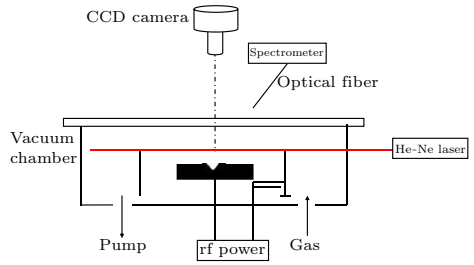

Fig. 1. Experimental setup.

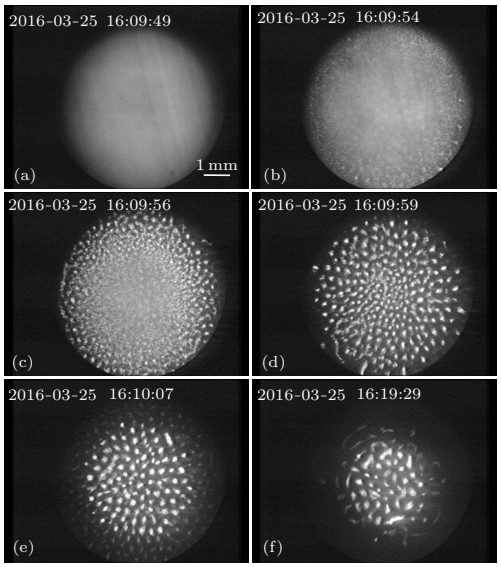

Fig. 2. Growth process of dust particles in a fixed two-dimensional plane. (a)–(f) The different snapshots after plasma discharge, started at 16:09:41.

Figure 2 shows the dust growth process for rf power of 56 W and gas pressure of 61 Pa ($SiH_4/Ar/C_2H_4$=1:19:40) where the dust particles are observed on a fixed plane 2.5 mm above inner electrode. Dense dust clouds of small size (estimated to be about nanometer size) can be seen in Fig. 2(a) after 8 s of plasma discharge. As can be seen in Fig. 2(b), after 13 s of plasma discharge, differentiating single particles is still impossible due to their small size. In Fig. 2(c), the small dust particles collide with each other frequently, growing rapidly to a size that can be resolved by a microscope while allowing single-particles to be distinguished in some areas. At this point, the number density of particles is still relatively high. As the plasma discharge time grows longer, the particle size increases and number density decreases as shown in Fig. 2(d). It is also interesting to note the high growth rate shown in Figs. 2(b)–2(d), where particles grew in only 5 s from barely recognizable to large enough for the camera to distinguish. With the growth continuous, some larger particles on the original plane will be suspended on the lower plane due to the gravity effect, leaving the original plane occupied by fewer particles, as shown in Fig. 2(f). Under the radial confinement, the larger particles will be constrained to the center part above the inner electrode. During this experiment, it can be seen that a rapid increase in the particle size before 2 min, and after 2 min the particles' sizes and shapes no longer experience significant change. In the particle growth investigation of Hayashi *et al.*,[4] the mean diameter of carbon particle growth in methane plasma linearly increased as time with the growth rate of 0.4 nm/s, and the grown particles were single spheres with nearly equal size and diameters between 2.0–2.2 μm. However, the particles in our experiment are not uniform in either size or shape, and many of the particles have much larger sizes, upto several-hundred times larger than those in the experiment of Hayashi *et al.*[4]

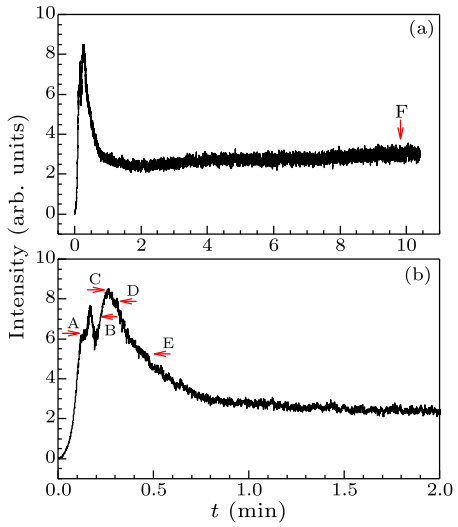

Fig. 3. The intensity of scattered light changes with the discharge time: (a) within about 10 min; and (b) the enlarged view of (a) within the first 2 min.

The growth process of dust particles can also be exhibited through detecting the intensity of laser light scattering, which is a complex convolution of particle size, shape, and concentration.[14] Figure 3(a) shows the *in situ* light scattering intensity changes vs plasma exposure time during the particle growth shown in Fig. 2. It can be clearly seen that the intensity of the scattered light increases rapidly in the initial stage of discharge and an obvious peak appears on the curve of the scattered intensity. About 2 min later, the intensity of scattered light shows no significant enhance-

ment with time. At a fixed detection angle, as the size of the particle increases, oscillations are observed in the scattering intensities. During the coagulation process, the scattered intensity can significantly increase with r_p. The end of 'fast' agglomeration occurs when electrostatic repulsion prevents agglomeration from proceeding at larger sizes.[14] Thus, after about $t = 2$ min the laser intensity falls to only 1/3 of maximum, and we can see a long plateau which is identified as the saturation stage. At the plateau, the particle sizes, shapes and number density do not change obviously. It should be pointed out that for different experimental parameters (for example, gas ratio, rf power, gas pressure) both the laser scattering intensity and the time to reach plateau are different.

The intensity and time of the snapshots shown in Figs. 2(a)–2(f) are marked in Fig. 3. Combining the growth process of Fig. 2, we can see that the end of the peak indicates the cessation of the main growth process as illustrated by a long plateau showing unremarkable increase marking the start of a saturation stage. Figure 2(c) approximately corresponds to the maximum of the peak with Figs. 2(a) and 2(b) in the ascending part of the peak, and Figs. 2(d) and 2(e) in the descending part of the peak. Figure 2(f) is located in the plateau during the saturation phase. It should be pointed out that the peak has a double-peak structure with the second peak higher than the first one, which may roughly correspond to the initial growth stage and fast coagulation stage (until now there has been no investigation distinguishing the precise separation point of these two stages).

As shown in Fig. 2, the dust particles obtained through chemical reaction in the gas discharge system tend to be non-spherical. The growth process for non-spherical dust particles can be characterized by fractal dimension obtained through a box-counting method.[9,15] The fractal dimension provides a measurement of the openness or porosity of a structure. It can range from nearly one for a linear structure to three for a compact sphere.[15] The fractal dimension is obtained by enclosing the dust region divided into square boxes with different side lengths l, and the number of boxes containing dust grains is denoted by $N(l)$. We have $\log N(l) = D\log(1/N) + C$, where D is the fractal dimension, and C is a constant. Thus the value of fractal dimension D can be obtained from the slope of the best-fit curve in the log-log plot of $N(l)$ versus $1/l$, that is, $D = \log N(l)/\log(1/l)$. Figure 4 indicates the change of fractal dimension with discharge time. The fractal dimension decreases sharply at the initial 2 min before remaining almost invariant (or with only small changes). This indicates that the initial growth and coagulation stages occur about at $0 < t < 2$ min. After 2 min, dust growth is in the saturation stage. This result is also roughly in agreement with the scattering laser intensity of Fig. 3. Thus, the fractal dimension of the system can also be used to qualitatively characterize the growth process of the dust particles.

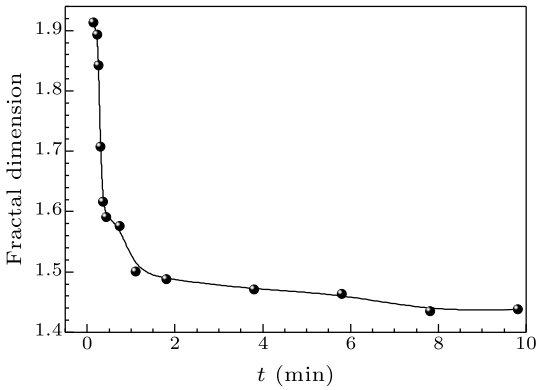

Fig. 4. Fractal dimension changes as discharge time.

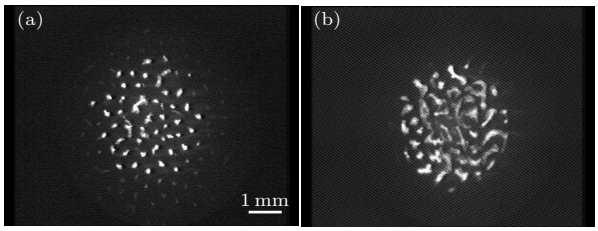

Fig. 5. Dust particles obtained with (a) 42 W and (b) 75 W.

The rf power directly impacts overall dust growth. Increasing the rf power causes significant changes in both the shape and size of the particles. Figure 5 shows a comparison of the snapshot of dust grains in a fixed two-dimensional plane of 3.5 mm above the inner electrode growth at 40 Pa ($SiH_4/Ar/C_2H_4$=1:19:50) with different rf powers. Dust particles with sphere-like shapes and smaller sizes are generally obtained at a lower rf power; for example, see Fig. 5(a). As the rf power is increased, the dust particles show different topological shapes and sizes, and non-spherical large particles can be obtained at a higher rf power; as shown in Fig. 5(b). It is known that higher rf power causes a higher electron temperature, leading to the increased dissociation of the hydrocarbon compounds,[16] contributing to the faster growth rate and the formation of larger dust particles. Since dust particles are generally negatively charged, the agglomeration requires that the reactants in an agglomerating collision have sufficient kinetic energy to overcome the mutual electrostatic repulsion. Thus, agglomeration of larger particles preferentially occurs at higher discharge powers.[17] In addition, in our experiment the electric polarization effect also exists in the uneven charged surface of particles by the influence of electric field of the plasma sheath.[18] The existence of polarization also implies the existence of an attraction between dust particles. Thus, at higher rf power, the particles have more chances to coagulate (or attract) together to form larger size nonspherical particles, such as rod-like and Y shapes in our experiment.[9,18]

The intensity of the scattered light of the dust par-

ticles obtained under different rf powers (in Fig. 5) with discharge time is shown in Fig. 6. Since the scattering laser light is related to the number density or the size of dust particles, or both,[11] which depend on discharge parameters; for example, rf power. As shown in Fig. 6, a higher rf power will cause a stronger scattering laser intensity. At the same time, the higher rf power will cause the faster growth rate, which reaches the saturation state earlier. In Fig. 6, the phenomena of double-peak structure are observed again. In short, the comparison on the particle growth behavior can also be reflected through comparing the intensity of scattered light.

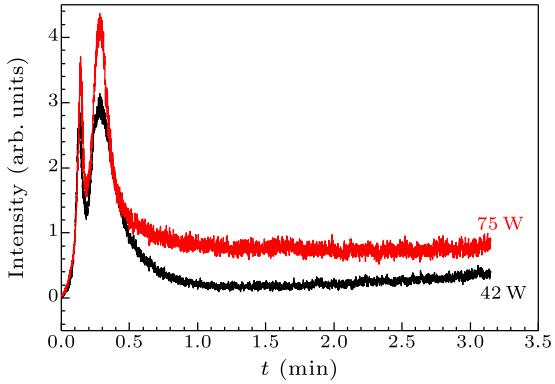

Fig. 6. The intensity of scattered light with different rf powers.

We investigate the dust particles' growth process not only in a fixed plane but also in different planes by adjusting the laser height up and down above the inner electrode. The 3D distribution can be obtained by combining the observations in different 2D planes. Figure 7 shows an example of the distribution for dust particles grown at 32 W and 40 Pa ($SiH_4/Ar/C_2H_4$=1:19:50) in the different planes with different heights above the inner electrode. As the height increases, the dust particles have a larger distribution. It is easy to understand that the 3D distribution of the dust particles forms a cone-like shape, which is related to the cone-shaped confinement in the inner electrode. In other words, the shape of the discharge electrode can influence the spatial distribution of dust particles in three dimensions. In addition, although at present the rf power dust particles are dominantly spherical-like shapes, the unequal size and shape of dust particles in each plane still can be observed, indicating the characteristic of the grown particles in our experiment.

Due to the effect of gravity, the average size of dust particles decreases from lower plane to upper plane. During particle growth, dust particles are levitated at different heights as a result of the equilibrium of gravity and electrostatic force. Small particles with small charge-to-mass-ratio can be easily levitated in the small electric field such as those in the bulk plasma.[19] Large particles require a strong field to levitate and, therefore, are suspended at the lower height, which is near the inner electrode in our experiment.

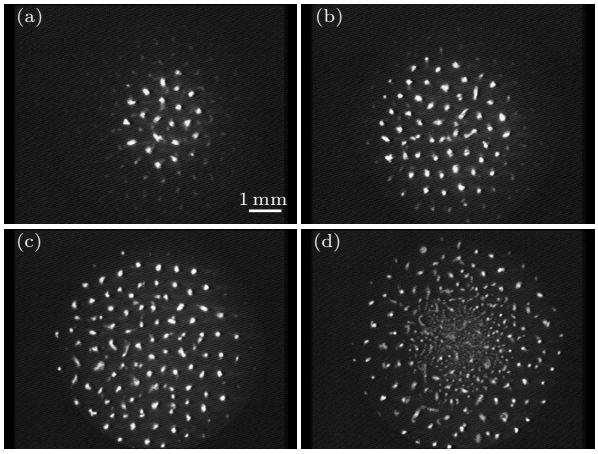

Fig. 7. Particles in different planes with different heights above the inner electrode: (a) $h = 3.5$ mm, (b) $h = 4.0$ mm, (c) $h = 4.5$ mm, and (d) $h = 5$ mm.

In conclusion, the growth process of the dust particles in a plasma system has been investigated. The *in situ* intensity of scattering laser light from dust particles and fractal dimension have been used to characterize the growth process. The intensity of scattering laser light shows an obvious peak with the double-peak structure. In addition, the fractal dimension decreases as the dust particles grow, until they remain almost invariant. The results of the comparison also show that spherical-like small dust particles are generally obtained at lower rf power and non-spherical large particles are obtained at higher rf power. Meanwhile, the scattering light intensity is stronger with higher rf power than with lower rf power. The particle distribution in three-dimensional space is also shown, which is related to the shape of the inner electrode and gravity effect.

References

[1] Selwyn G S 1994 *Plasma Sources Sci. Technol.* **3** 340
[2] Selwyn G S et al 1989 *J. Vac. Sci. Technol.* A **7** 2758
[3] Olevanov M A et al 2004 *J. Exp. Theor. Phys.* **98** 287
[4] Hayashi Y et al 1994 *Jpn. J. Appl. Phys.* **33** 4208
[5] Matthews L S et al 2004 *IEEE Trans. Plasma Sci.* **32** 586
[6] Kawasaki H et al 1998 *J. Appl. Phys.* **83** 5665
[7] Watanabe Y et al 1996 *J. Vac. Sci. Technol.* A **14** 540
[8] Praburam G et al 1996 *Phys. Plasmas* **3** 1212
[9] Huang F et al 2012 *Phys. Plasmas* **19** 093708
[10] Barkan A et al 1994 *Phys. Rev. Lett.* **73** 3093
[11] Prabhakara H R et al 1996 *Phys. Plasmas* **3** 3176
[12] Yoo W J et al 1993 *J. Vac. Sci. Technol.* A **11** 1258
[13] Dahiya R P et al 2002 *Phys. Rev. Lett.* **89** 125001
[14] Bouchoule A 1999 *Dusty Plasmas, John wiley & Sons Canada Limited* (New York: Academic) p 186–198
[15] Perry J D et al 2010 *IEEE Trans. Plasma Sci.* **38** 792
[16] Hong S et al 2003 *Plasma Sources Sci. Technol.* **12** 46
[17] Huang F Y et al 1996 *J. Vac. Sci. Technol.* A **14** 562
[18] Hou X N et al 2016 *Phys. Plasmas* **23** 093702
[19] Samsnov D et al 2003 *New J. Phys.* **5** 24