Study on morphology and growth of water-ice grains spontaneously generated in a laboratory plasma

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Abstract

An apparatus has been developed to study the nucleation, growth, and morphology of water-ice grains spontaneously generated in a weakly ionized plasma having very cold neutral particles. Nucleation of water-ice grains in the lab experiment occurs only when plasma exists but the plasma density is not too high. Nonspherical, fast growth occurs when the mean free path of water molecules exceeds the screening length for the ice grain in which case molecules incident on the ice grain can be considered to have collisionless trajectories. High water vapor pressure enhances this nonspherical, fast growth provided the collisionless condition is satisfied. Magnetic field impedes nonspherical growth by reducing the charge residing on water-ice grains if the field is sufficiently strong to make the electron gyro radius smaller than the ice grain screening length.

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1. Introduction

Water-ice dusty plasmas are composed of electrons, ions, neutrals, and water-ice grains. These plasmas exist in many natural contexts, most notable examples being terrestrial polar mesospheric clouds, certain of Saturn's rings, and astrophysical molecular clouds. The water-ice grains constituting terrestrial polar mesospheric clouds have 10-100 nm nominal size [Havnes *et al.*, 1996] and exist during the summer at ~85 km altitude and polar latitudes in the mesopause. Because of the high altitude of these clouds, they are visible at night and so are also called noctilucent clouds. The water-ice grains are negatively charged with a few elementary charges. Occasionally positively charged grains are observed and it is presumed that this positive charging occurs by the photoelectric effect [Havnes *et al.*, 1996]. Strong reflection of 50 MHz – 1.3 GHz radar has long been observed from these clouds [Ecklund and Balsley, 1981] and Bragg reflection by electrons is thought to be responsible for the radar echo [Rapp and Lubken, 2004]. However, sounding rocket measurements sometimes show radar echoes occur from bite-out regions where most of the electrons reside on the water dust grains so there are no free electrons in the plasma [Havnes *et al.*, 2001; Rapp *et al.*, 2003]. The reason for this remains controversial; Bellan [2010] suggested that elongated morphology of water-ice grains might explain this bite-out mystery.

Saturn's E-, F-, and G-rings are also water-ice dusty plasmas and are composed of micron size water-ice grains [Goertz, 1989]. Since the number density of water-ice grains is quite high in the F-ring, a large fraction of the negative charge resides on the dust grains and collective behaviors of the ice grains such as waves are observed [Goertz, 1989; Murray *et al.*, 2008]. On the other hand, the number densities of water-ice grains in E- and G-rings are so small that most electrons are free in the plasmas and the dust grains do not have strong mutual interactions. Saturn's E-ring and Enceladus have attracted much recent attention because Cassini spacecraft images reveal that Enceladus emits water vapor and water-ice

1 grains into the E-ring [*Porco et al.*, 2006].

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3 In astrophysical molecular clouds, water-ice grains play several important roles including
4 forming planetesimals, cooling the cloud, and storing oxygen molecules. These astrophysical
5 water-ice grains have been observed indirectly via infrared spectroscopy, and their density
6 structure has been used to estimate the evolution stage of molecular clouds [*McClure et al.*,
7 2012].

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9 Probably for reasons of mathematical convenience, it has generally been assumed that
10 water-ice grains are spherical in all the contexts mentioned above. However there is no strong
11 evidence that this is so. We believe that non-spherical geometry is more likely in many
12 situations because water molecules have a large dipole moment and because water-ice grains
13 in a plasma environment become electrically charged. Furthermore, it has been reported that
14 if water-ice grains were non-spherical, certain observed features might be better explained
15 [*Baumgarten and Fricke*, 2002; *Rapp et al.*, 2007; *Bellan*, 2010]. In order to investigate the
16 possibility of non-spherical ice grains, we built a laboratory apparatus and used this apparatus
17 to investigate the morphology of water-ice grains in a plasma. Initial results show that water-
18 ice grains are spontaneously generated in a plasma environment and grow nonspherically if
19 the neutral pressure of the background weakly ionized plasma is low [*Chai and Bellan*, 2013].
20 We report in this paper measurements made to provide an improved understanding of the
21 underlying physics of nonspherical growth. Besides morphology, we also investigated the
22 nucleation and the growth rate of water-ice grains.

23 24 25 26 27 28 29 30 31 32 33 34 35 36 37 38 39 40 41 42 43 44 45 46 47 48 49 50 51 52 2. Experimental setup

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54 Figure 1 shows a photo of the water-ice dusty plasma; a detailed description is given
55 elsewhere [*Chai and Bellan*, 2013]. The apparatus incorporates design features of a
56 previously existing experiment at the Max-Planck-Institute for Extraterrestrial Physics

[Shimizu *et al.*, 2010] but differs in two important aspects: the inter-electrode separation is adjustable and water vapor is directly injected into an argon plasma instead of forming water molecules in a plasma created from a fill of D₂ and O₂. Adjustable electrodes provide a wider range of pressures for plasma breakdown and direct injection of water vapor into a pre-existing argon plasma provides independent means for controlling and measuring the water vapor pressure. The plasma is created in the space between two 6 cm diameter aluminum electrodes in the vacuum chamber. The electrodes are thermally connected via cold fingers to containers holding liquid nitrogen.

In normal operation, the electrodes are cooled by first pouring liquid nitrogen into the containers. We then wait 45 minutes to obtain suitably low equilibrium temperature in the chamber. The vacuum chamber is then filled with 100-600 mTorr Ar gas (13-80 Pa) and a plasma is ignited with 0.5-2 W of 13.56 MHz rf power applied across the electrodes. Immediately after 0.5-2 mTorr (0.07-0.27 Pa) water vapor has been introduced into the plasma, water-ice grains form spontaneously and levitate between the electrodes. Sufficient cooling time and plasma are necessary for formation of the water-ice grains – if there is not enough cooling time or no plasma, water-ice forms only on the surface of the electrodes.

The water-ice grain size and shape are measured using the method shown in Fig. 2(a). In this method, relatively large water-ice grains levitating near bottom electrode are illuminated by a 5 mW, 632.8 nm He-Ne laser with 0.5 mm beam diameter (focused by lens). Magnified images of ice grains are then obtained by a long distance microscope lens (Questar QM-100) mounted on a digital SLR camera (Nikon D5300); the lens and camera view at a right angle to the laser beam. The distance between the water-ice grains and the entrance of the long distance microscope is about 20 cm. The magnification of this microscope-camera system is 16 and the depth of field is 30 μ m. This imaging system shows the water-ice grain size and shape directly provided the water-ice grain dimension exceeds the resolution threshold of a

few microns. This imaging system also enables measuring the water-ice grain number density by simply counting how many water-ice grains exist in the laser beam path.

A Langmuir probe is used to measure ion density as shown in Fig. 2(b). The Langmuir probe is biased with a large negative voltage (-30 V) and the ion saturation current flowing through a 100 k Ω resistor is measured. Ion density is calculated from the measured ion saturation current, using the relation $n_i = I_{is} / (\alpha e A_p u_s)$ where n_i is the ion density, I_{is} is the ion saturation current, α is the ratio of ion density at sheath to bulk, A_p is the area of probe tip, and u_s is the ion sound velocity. Ion sound velocity is given by $u_s = (kT_e / m_i)^{1/2}$ where T_e is the electron temperature and m_i is the ion mass. It is assumed that $\alpha = 0.5$ and that $T_e = 3$ eV to obtain $u_s = 2700$ m/s. The probe tip diameter is 0.5 mm and the tip length is 20 mm.

3. Results

3.1. Nucleation of water-ice grains

Because no smoke particles are injected, the nucleation is homogeneous. Nucleation of water-ice grains is observed to occur when three requirements are satisfied: (1) plasma is present, (2) the water vapor pressure exceeds some specific threshold value, and (3) input rf power, i.e. plasma density, is not higher than a certain value. It is also found that if the input rf power is lower than a certain value, the plasma becomes unstable after nucleation occurs and sometimes terminates, probably because the water-ice grains scavenge too many electrons so a plasma cannot be sustained. Our results suggest that charged plasma particles contribute to the initiation of water-ice nucleation but there should not be too high a density of charged particles.

Since the nucleation of water-ice does not take place when the plasma is ignited with high rf power, we ignited plasmas with low rf power (0.5 W) and then increased rf power (0.5 - 2 W) in this work. By doing so, we were able to obtain large water-ice grains, which will be

discussed later. Besides being of interest for their own sake, larger grains have more clearly resolved images because they are well above the resolution threshold of the imaging system.

The summarized results of this work are shown in Figs. 3(a) and (b). Figure 3(a) is obtained in argon plasmas having 1.5 mTorr water vapor while Fig. 3(b) is based on the results from argon plasmas having 2 mTorr water vapor. In both water vapor pressures, water-ice grains become large and more elongated as the background gas pressure decreases or the input rf power increases (i.e., larger and more elongated toward upper left corner of figure). Also, water-ice grains, generated in the plasmas with 2 mTorr water vapor are larger and more elongated than those generated in the plasmas with 1.5 mTorr water vapor pressure if all other parameters are the same.

3.2 Pressure effect

We investigated how the shape and growth rate of water-ice grains are affected by the pressure of the Ar background neutral gas. Plasmas were ignited with 0.5 W of rf power at various Ar background pressures (100-600 mTorr) and 2 mTorr water vapor pressure. The rf power was then increased to 2 W at 20 s after plasma ignition and the camera system was used to image the water-ice grains (see upper-most row in Fig. 3(b)).

The images of water-ice grains shown in Figs. 4(a)-(d) reveal that ice grains generated in low Ar pressure plasma are large and vertically elongated whereas ice grains generated in high Ar pressure plasma are small and spherical. Figure 4(a) is a composite of several different grains due to the low number density of ice grains at 60 mTorr while Figs. 4(b)-(d) are single photos. Because it was not possible to ignite plasma at 60 mTorr Ar pressure, we ignited plasma at 100 mTorr first and then reduced the Ar pressure to 60 mTorr after water-ice grain growth finished to obtain images of water-ice grains.

The aspect ratio and maximum ice grain major radius obtained from Fig. 4 are shown in

Figs. 5(a) and (b). The aspect ratio and maximum major radius increase respectively from 1 to 5 and from 2.6 μm to 70 μm as Ar background pressure decreases from 600 mTorr to 60 mTorr. The number density of ice grains decreases from $1.2 \times 10^5 \text{ cm}^{-3}$ to $1.8 \times 10^4 \text{ cm}^{-3}$ as the Ar pressure decreases from 600 mTorr to 100 mTorr.

It is unclear why all the water-ice grains are aligned in the vertical direction but it is surmised that this vertical alignment is related to a sheath/presheath electric field in the plasma. Since the plasma potential is higher than the wall potential in typical laboratory plasmas, an electric field exists in the sheath and presheath regions between the bulk plasma and the electrodes which define the wall. Ice grains seem to be aligned along this electric field.

In order to study water-ice grain growth, we obtained images of water-ice grains at a sequence of times as shown in Figs. 6(a)-(d). These images were obtained using a plasma sustained by 2 W rf power at 100 mTorr Ar pressure and 2 mTorr water vapor pressure. It is clearly seen that ice grains become larger and more elongated with increasing time. Figure 7(a) shows the time evolution of the major radius of water-ice grains at different background pressures. In all cases, water-ice grains first grow fast and then stop growing at around 60-80 s. We think this saturation is caused by either depletion of the water molecules that are the source for water-ice grain growth or by sublimation of water molecules from the water-ice grain surface. Figure 7(b) reveals that the fastest growth (0.27 $\mu\text{m/s}$) occurs at the lowest background pressure indicating that collision of water molecules with background gas impedes water-ice grain growth.

3.3 RF power effect

The effect of rf power on water-ice grain shape and growth rate was investigated by igniting plasmas with 0.5 W rf power at 100 mTorr Ar and 1.5 mTorr water vapor pressures. At 20 s,

the rf input power was varied from 0.5 to 2 W and images of the water-ice grains were made (see left-most column in Fig. 3(a)). Photos of water-ice grains obtained for a series of rf power plasmas are shown in Figs. 8(a)-(d) and reveal that the ice grains generated in the high input power plasmas are larger and more elongated.

Ion density, measured by a Langmuir probe, increases from $4.7 \times 10^9 \text{ cm}^{-3}$ to $1.5 \times 10^{10} \text{ cm}^{-3}$ as rf power varies from 0.5 W to 2 W. Using these density values, aspect ratio and maximum major radius of water-ice grains are displayed as functions of ion density in Figs. 9(a) and (b). As the plasma ion density increases, the aspect ratio increases from 1.1 to 3 and the maximum major radius increases from $3.3 \text{ }\mu\text{m}$ to $14 \text{ }\mu\text{m}$. The number density of ice grains remains approximately constant at $2.2 \times 10^4 \text{ cm}^{-3}$ as rf power is varied from 1 W to 2 W.

Figure 9(c) shows the time evolution of water-ice grain major radius for the various ion density plasmas obtained from the time-resolved ice grain images. As for the sequence of different background pressures, the ice grains first grow fast and then stop growing at around 60 s. Fastest growth rate ($0.23 \text{ }\mu\text{m/s}$) is observed when the ion density is highest ($1.5 \times 10^{10} \text{ cm}^{-3}$) as shown in the Fig. 9(d), indicating that increasing ion density has the same effect as lowering background gas pressure.

3.4 Water vapor pressure effect

The water vapor pressure was adjusted to investigate how this pressure affects the shape and growth rate of water-ice grains. Plasmas were ignited with 0.5 W rf power at 100 mTorr Ar and 1, 1.5, and 2 mTorr water vapor pressures. After plasma ignition the rf power was increased to 2 W and the water-ice grains were photographed (upper left points in Figs. 3(a) and (b)). As shown in Figs. 10(a)-(c), more elongated and larger ice grains are generated in the higher water vapor pressure plasma. As the water vapor pressure increases, the ice grain aspect ratio increases from 2.0 to 3.5 and the ice grain maximum major radius increases from

11 μm to 21 μm (see Figs. 11(a) and (b)). The number density of ice grains decreases slightly from $3.2 \times 10^4 \text{ cm}^{-3}$ to $2.2 \times 10^4 \text{ cm}^{-3}$ as water vapor pressure increases.

As in the previously shown results, water-ice grains first grow fast and then stop growing in all water vapor pressure cases as seen in the Fig. 11(c). Figure 11(d) shows that fastest growth (0.33 $\mu\text{m/s}$) occurs at the highest water vapor pressure (2 mTorr), indicating ice grains can grow large and nonspherical when the source water molecules are plentiful.

The effect on ice grain shape and growth rate of water vapor pressure was also investigated for low density plasma (0.5 W rf power case). Unlike the high density plasma, water-ice grain aspect ratio does not change significantly as water vapor pressure increases for low density plasma although the water-ice grain growth rate increases. We conclude that water-ice grain morphology is only affected by the water vapor pressure when the condition of nonspherical growth is satisfied (i.e., the high rf power, low pressure case).

3.5 Magnetic field effect

The role of magnetic field in water-ice grain growth and morphology was studied. Plasmas were ignited with 0.5 W of rf power at 100 mTorr Ar pressure and 2 mTorr water vapor pressure with and without a vertical magnetic field. The magnetic field was turned on from the beginning of plasma ignition. At 20 s, the input rf power was increased to 1.5 W. The images of water-ice grains captured at 120 s in these plasmas show that the ice grains change from being large and elongated (major radius of 9.0 μm , aspect ratio of 2.0) to small and spherical (major radius of 3.1 μm , aspect ratio of 1.1) when the magnetic field increases from 0 to 190 G (see Fig. 12).

We also tried to apply magnetic field to the high density plasma (2 W rf power), but the ice grain shape and size were not affected significantly even by 190 G magnetic field. This indicates that a stronger magnetic field is needed to change water-ice grain size and shape in

the higher density plasma.

4. Discussion

It is found that water-ice grains grow larger, more elongated, and faster at low background Ar pressure, i.e., when the mean free path of water molecules is long. The mean free path of water molecules is expressed as $\lambda_{\text{mfp}}=1/(\pi d^2 n_g)$, where d is the summation of incident and target molecules' radii (here, summation of water molecule radius and Ar atom radius), n_g is the target neutral gas number density (here, Ar). The calculated mean free paths are 321 μm , 161 μm , and 80 μm for 100 mTorr, 200 mTorr, and 400 mTorr pressures, respectively.

Larger and more elongated water-ice grains are observed to be generated in higher ion density plasmas, i.e., in plasmas where the screening length of water-ice grains is short. In our experiments, the major radius of water-ice grain becomes comparable to or larger than the ion Debye length, $\lambda_{\text{Di}}=(\epsilon_0 k T_i / n_i e^2)^{1/2}$ where T_i is the ion temperature and n_i is the ion density. The water ice grain behaves like a floating Langmuir probe so the screening length (sheath width) of water-ice grains is expected to be of the order of the electron Debye length, $\lambda_{\text{De}}=(\epsilon_0 k T_e / n_e e^2)^{1/2}$ where n_e is the electron density [Daugherty *et al.*, 1992]. With the assumption that $T_e=3$ eV and $n_e \sim n_i$, the electron Debye lengths are calculated to be 181 μm , 128 μm , and 104 μm for plasmas having ion and electron densities of $5 \times 10^9 \text{ cm}^{-3}$, $1.0 \times 10^{10} \text{ cm}^{-3}$, and $1.5 \times 10^{10} \text{ cm}^{-3}$, respectively.

The presumed mechanism for nonspherical growth has some similarities to that suggested by Stark *et al.* [2006] but also differs in important aspects. In the theory of Stark *et al.* [2006], positively ionized incident molecules are accelerated along the electric field lines near a dust grain and accreted so long as the initial kinetic energy is lower than Coulomb potential energy. The incident charged molecules are preferentially attracted to the sharp edges of the grain because strong electric fields exist there. The water ice situation differs because incident

1 water molecules are uncharged but highly polarized. Consequently, the water molecules are
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3 attracted to regions of large field gradient in analogy to iron filings being attracted to the
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5 poles of a bar magnet. Since the location of large field gradients is also at the sharp edges, the
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7 neutral dipolar water molecules are similarly attracted to the sharp edges. Another difference
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9 between this situation and that proposed by Stark *et al.* [2006] is that accumulation of neutral
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11 water molecules will not change the charge of the dust grain whereas accumulation of
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13 positive ions will reduce the charge.
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17 This nonspherical growth mechanism requires that molecules should not collide with
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19 background molecules before they reach the ice grain surface. If a molecule does suffer a
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21 collision before reaching the ice grain surface, it will not follow the field gradient, and
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23 therefore, only uniform, spherical growth can occur. Our results strongly support this idea.
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25 Nonspherical growth of water-ice grains is observed to occur when the mean free path of
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27 water molecules exceeds the electron Debye length.
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32 Another possible explanation of nonspherical growth is the directional, collisional
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34 coagulation of ice grains as described in *Du et al.* [2010]. According to *Du et al.* [2010], the
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36 dust grains can obtain large enough kinetic energy by the dust-acoustic wave to overcome the
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38 Coulomb repulsive potential energy so they agglomerate with each other to grow. We do not
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40 believe this mechanism causes the nonspherical growth occurring in our experiment because a
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42 high speed movie camera (1000-2000 fps) does not reveal any collisions between water-ice
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44 grains; the grains always remain well-separated. Furthermore the dust-acoustic wave is
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46 observed in the plasma both when ice grains are small and spherical (high pressure and low rf
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48 power cases) and when ice grains are large and non-spherical (low pressure, high rf power).
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54 The role, if any, of the strong electric field existing in the plasma-sheath region, in
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56 nonspherical growth is not well understood at this time. Because there is always a strong
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58 sheath electric field no matter what the plasma parameters are, we believe that the extremely
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1 localized electric field gradient established just exterior to the nonspherical ice and attracting
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3 the dipole moment of nearby water molecules is more important than the electric field in the
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5 plasma-sheath region. While the sheath electric field does not affect grain growth, it likely is
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7 responsible for the vertical alignment of non-spherical ice grains.
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10 Growth rate of water-ice grain is observed to be proportional to water vapor partial pressure
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12 as shown in Fig. 11(d). This corresponds to one aspect of the supersaturation growth theory
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14 given by Hesstvedt [1961], but unlike Hesstvedt, here the growth rate also depends on the
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16 non-water background gas pressure because this background controls the collision mean free
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18 path.
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22 It is noticeable that the aspect ratio of water-ice grains is only affected by water vapor
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24 pressure when the nonspherical growth condition is satisfied (collisionless regime). The
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26 morphology of water-ice grains is not changed by water vapor pressure when the plasma is in
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28 the collisional regime, e.g. low rf power, high pressure case.
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32 The reason why the size and aspect ratio of water-ice grains are affected by magnetic field is
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34 related to the charging of a water-ice grain. It is generally known that magnetic field reduces
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36 the number of charges residing on dust grain if the magnetic field is strong enough to make
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38 the electron gyro radius comparable to the dust grain screening length [*Tsytoovich et al.* 2003].
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40 The electron gyro radius in a 190 G magnetic field is 220 μm while the electron Debye length
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42 is 128 μm (with $T_e=3$ eV, $n_e=1.0\times 10^{10}$ cm^{-3}), indicating charge reduction is likely to happen.
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44 Because a smaller dust grain charge number implies a weaker interaction between water-ice
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46 grains and incident water molecules, a magnetic field causes the water-ice grains to remain
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48 small and less elongated just as for low water vapor pressure.
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54 Our plasma parameters such as ambient gas pressure, water vapor partial pressure, and
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56 plasma density are quite different from those of naturally occurring plasmas such as polar
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58 mesospheric clouds, Saturn's rings, and astrophysical molecular clouds: a critical question is
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whether the findings presented here can be extrapolated to natural plasmas. In order to address this question, we calculate the ratio of mean free path to screening length as given in Table 1. In the calculation, nominal plasma and water-ice grain parameters are used. The screening length of water-ice grains is the electron Debye length for Caltech plasma (because water-ice grain size is larger than ion Debye length) while the ion Debye length is the screening length for other natural plasmas (because water-ice grains are much smaller than ion Debye length). As seen in Table 1, the mean free path to screening length ratios of polar mesospheric clouds (PMC), Saturn's ring, and astrophysical molecular clouds are all similar to or larger than that of Caltech plasma, suggesting it is likely that nonspherical water-ice grains exist in these natural plasmas.

5. Conclusion

It is found that the plasma is necessary for the nucleation of water-ice grains to take place in our lab experiment and that the plasma density should not be too high. We do not now know the reason why there is an upper limit of plasma density and plan to investigate this in the future. Water-ice grains grow more elongated, larger, and faster in plasmas where the mean free path of water molecules is longer than the screening length of water-ice grains. Aspect ratio, final size, and growth rate of water-ice grains can be enhanced by high water vapor pressure when the collisionless condition is satisfied. A magnetic field that magnetizes electrons impedes nonspherical growth by reducing the charge on an ice grain. It is expected that non-spherical water-ice grains exist in natural plasmas such as polar mesospheric clouds, Saturn's ring, and astrophysical molecular clouds because they are in the collisionless regime.

Our future plans include igniting plasma with other gases such as hydrogen and nitrogen to better mimic the actual atmospheric environment. We will try to produce ice grains made of molecules that have no dipole moment such as CO_2 to ensure the nonspherical growth is

1 induced by dipole moment and the electric field gradient established near nonspherical
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3 geometry. Why ice grain growth stops and why the grains are all lined up vertically are also
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5 targets of future investigation. We also have plans to adopt more sophisticated diagnostics for
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7 small ice grains, for investigating nucleation and plasma chemistry, and to push our
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9 experimental parameters such as densities toward actual mesospheric cloud conditions.
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Table 1. Nominal plasma parameters and the ratios of $\lambda_{\text{mfp}}/\lambda_{\text{s}}$ of Caltech and natural plasmas

	n_{g}	T_{g}	λ_{mfp}	T_{e}	n_{e}	λ_{De}	T_{i}	n_{i}	λ_{Di}	r_{p}	$\lambda_{\text{mfp}}/\lambda_{\text{s}}$
Caltech	10^{22}	10^{-2}	10^{-4}	1	10^{16}	10^{-4}	10^{-2}	10^{16}	10^{-6}	10^{-6}	1
PMC	10^{20}	10^{-2}	10^{-2}	10^{-2}	10^9	10^{-2}	10^{-2}	10^9	10^{-2}	10^{-7}	1
E-, F-, G-rings	10^9	10^{-3}	10^8	5	10^7	1	5	10^7	1	10^{-6}	10^8
Molecular clouds	10^{15}	10^{-3}	10^2	1	10^6	1	10^{-3}	10^6	10^{-1}	10^{-7}	10^3

¹ T_{g} : background gas temperature, λ_{s} : screening length of water-ice grains

² All in SI units except temperature in eV.

³ References:[*Rapp and Lubken*, 2004; *Goertz*, 1989; *Ip* 2000;*Shu et al.* 1987; *Zubko et al.* 2004]

Figure captions

Fig. 1. Photo of Caltech water-ice dusty plasma apparatus. Two parallel, cold fingered electrodes are used to ignite and cool down the plasma. Ar gas is used for making background plasma and water vapor is used to form water-ice grains. Laser-aided long distance microscope camera and single Langmuir probe are shown at the bottom right.

Fig. 2. Schematic sketches of (a) laser-aided long distance microscope camera and (b) Langmuir probe.

Fig. 3. Summarized sketches of water-ice grain shape and size generated (a) in the 1.5 mTorr water vapor pressure and (b) in the 2 mTorr water vapor pressure plasmas.

Fig. 4. Images of water-ice grains captured at Ar pressure of (a) 60 mTorr (composite image), (b) 100 mTorr, (c) 200 mTorr, and (d) 600 mTorr. Input rf power was 2 W and water vapor pressure was 2 mTorr. Maximum major radius (r_p), aspect ratio (a), and number density (n_p) are indicated as white in each image.

Fig. 5. (a) Aspect ratio and (b) maximum major radius of water-ice grains as functions of Ar background pressure.

Fig. 6. Time-resolved images of water-ice grains captured at (a) 20 s, (b) 40 s, (c) 60 s, and (d) 80 s. Plasma was sustained by 2 W rf power at 100 mTorr Ar pressure and 2 mTorr water vapor pressure.

Fig. 7. (a) Time evolution of major radius of water-ice grains at various pressures. Squares, circles, triangles, and inverted triangles represent Ar background pressures of 100 mTorr, 200 mTorr, 400 mTorr, and 600 mTorr, respectively. (b) Growth rate, defined as maximum major radius divided by time to reach that size, as a function of Ar background pressure.

Fig. 8. Images of water-ice grains spontaneously formed in the plasma sustained by rf power of (a) 0.5 W, (b) 1 W, (c) 1.5 W, and (d) 2 W. Ar and water vapor pressures were 100 mTorr and 2 mTorr, respectively.

Fig. 9. (a) Aspect ratio and (b) maximum major radius of water-ice grains as functions of ion density of plasma measured by Langmuir probe. (c) Time evolution of major radius of water-ice grains generated in various ion density plasmas. Squares, circles, triangles, and inverted triangles represent rf power of 0.5 W, 1 W, 1.5 W, and 2 W, respectively. (d) Growth rate as a function of ion density of plasma.

Fig. 10. Water-ice grain images captured at the water vapor pressure of (a) 1 mTorr, (b) 1.5 mTorr, and (c) 2 mTorr. Input rf power was 2 W and Ar pressure was 100 mTorr.

Fig. 11. (a) Aspect ratio and (b) maximum major radius of water-ice grains as functions of water vapor pressure. (c) Time evolution of major radius of water-ice grains generated at various water vapor pressure. Squares, circles, and triangles represent water vapor pressures of 1 mTorr, 1.5 mTorr, and 2 mTorr, respectively. (d) Growth rate as a function of water vapor pressure.

Fig. 12. Images of water-ice grains captured in the plasma with (a) no magnetic field, (b) 92

1 G magnetic field, and (c) 190 G magnetic field. Ar and water vapor pressures were 100 mTorr
2
3 and 2 mTorr, respectively. Input rf power was 1.5 W.
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Fig. 1

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Fig. 11

Fig. 12