Laser-filamentation-induced condensation and snow formation in a cloud chamber

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Received November 10, 2011; revised January 16, 2012; accepted January 31, 2012; posted January 31, 2012 (Doc. ID 157795); published March 26, 2012

Using 1 kHz, 9 mJ femtosecond laser pulses, we demonstrate laser-filamentation-induced spectacular snow formation in a cloud chamber. An intense updraft of warm moist air is generated owing to the continuous heating by the high-repetition filamentation. As it encounters the cold air above, water condensation and large-sized particles spread unevenly across the whole cloud chamber via convection and cyclone like action on a macroscopic scale. This indicates that high-repetition filamentation plays a significant role in macroscopic laser-induced water condensation and snow formation. © 2012 Optical Society of America

OCIS codes: 120.3940, 140.3450, 260.5130.

In the past few years, the Teramobile group demonstrated laser-assisted water condensation both in a cloud chamber and the atmosphere [1–3]. The ideas are different from classical cloud seeding by firing massive amounts of carbonic ice, silver iodide, etc., into the atmosphere [4–6]. They relied on the generation of high-density charged particles, which initiated condensation nuclei [1–3,7]. The latter further grew in size to form ice crystals in the low-temperature clouds. Due to the high electron density of ~1016 cm−3 in the self-guided ionized filaments [8–12], water condensation around the filaments has been observed in both saturated and subsaturated conditions [1]. It was proposed that the photo-oxidative chemistry of nitrogen induces the binary H2O-HNO3 nucleation, on which water droplets could grow [1–3].

However, a significant rainmaking would not take place if the cloud seeding via the laser-induced binary H2O-HNO3 nucleation was only limited in and around the filament’s active volume, which has a diameter of ~100 μm [9]. A 100 TW femtosecond laser pulse was used to induce water condensation on a macroscopic scale by generating several hundreds of multifilaments in a beam size of ~10 cm [13]. It was found that nanoparticle generation increased faster than linearly with the power and with the number of the filaments at laser power beyond 50 TW. Contrary to using a high-energy laser pulse, in this Letter, we demonstrated laser-induced water condensation and spectacular snow formation on a macroscopic scale by firing the filaments of low-energy femtosecond laser pulses at 1 kHz repetition rate into a cloud chamber.

The experiments were performed using a femtosecond Ti:sapphire laser, which delivered 9 mJ/50 fs pulses with a repetition rate of 1 kHz [Fig. 1(a)]. The laser pulses were focused by an f/70 concave mirror and launched into a diffusion cloud chamber filled with ambient air to generate filaments with a length of ~10 cm [top of Fig. 1(b)]. A 532 nm probe beam of 16 mm (height) × 5 mm (width), with 10 W output power, copropagated with the femtosecond laser beam into the chamber. The side Mie scattering was recorded by a digital camera. The cloud chamber had a size of 0.5 m × 0.5 m × 0.2 m, a vertical temperature gradient

![Fig. 1. (Color online) (a) Schematic experimental setup. (b) (Top) Fluorescence of the laser filament in the cloud chamber without the probe beam. (Bottom) Recorded side Mie scattering around the filament induced by the probe beam with an exposure time of 1/4000 s. The heap of snowpack below the filament center was illuminated by the stray light of the green laser beam. (c) Close-up shot for the snowpack in (b). (d) Laser-induced snow formation on the whole bottom base plate after firing the 1 kHz laser pulses for 2 h. (e) Close-up shot for the snowpack in (d).](image-url)
was maintained in the chamber by using a refrigerating machine to cool the bottom base plate at a temperature of −46 °C, while the top of the chamber was kept naturally cooled.

The height of the laser axis relative to the bottom base plate of the cloud chamber was set to be 10 mm, where the temperature and supersaturation ratio (S) were −29 °C and 1.27 ± 0.16, respectively. (The saturation ratio was derived from the measured temperature distribution inside the chamber [14].) It was found that, as shown in Fig. 1(b), large-sized particles in the range of 40–300 μm in diameters were produced around the filament. After an irradiation for 30 min by the kilohertz pulses, a heap of snow confined in an area of ∼2.0 × 1.5 cm² on the bottom base plate was observed just below the laser filament center. It was carefully shoveled out and weighed ∼13 mg. The acidity of the snow was measured to be pH ∼2 by using the pH test paper, which indicated that an acid with a concentration of ∼0.01 mol/L was produced. In addition, the melted snow was analyzed by an ion chromatograph and HNO₃ was confirmed with a concentration of 0.032 mol/L. The generation of HNO₃ in the laser-induced snowpack confirmed the pathway for the efficient H₂O-HNO₃ ice nucleation owing to the photo-oxidative chemistry of nitrogen triggered by filamentation [1–3].

A close-up photograph of the snowpack showed that the shape of the snowflake exhibited branched or dendritic features with a size of ∼1.45 mm × 1.25 mm, as shown in Fig. 1(c). By increasing the irradiation duration to 2 h, the weight of the laser-induced snowpack below the filament center increased to ∼55 mg. Besides this heap of snow, the whole bottom base plate was covered by a light layer of snow as shown in Fig. 1(d). However, as compared with the dendritic snowflake in the snowpack at the center, the snowflakes elsewhere took the form of ice particles and had much smaller sizes as shown in Fig. 1(e). The measured acidity of the snow outside the snowpack showed that the acid concentration (of HNO₃) was at the comparable level of ∼0.01 mol/L as in the snowpack. This result indicated that the binary H₂O-HNO₃ nuclei generated by the laser filaments moved far away from the filament and stimulated the snow formation through cloud seeding on a macroscopic scale.

The intense updraft, convection, and even cyclone induced by the 1 kHz laser filamentation were observed with naked eyes and recorded by the digital camera. Figure 2(a) shows the side scattering of the probe laser beam before the femtosecond pulses were fired into the cloud chamber. Only small particles with a uniform distribution were observed. After firing the filaments of the 1 kHz femtosecond pulses, an intense air current moving upward (updraft) was induced near the filament center. Figure 2(b) was just one video capture. Obvious trailing smear due to the high-speed motion of particles can be seen because only 25 frames could be captured per second with an exposure time of ∼1/50 s for each frame. Nevertheless, the maximum motion speed of the air current could roughly be estimated as >10 cm/s. Owing to the high clamped intensity of 5 × 10⁵ W/cm² in the filament [15,16], cold moist air containing water molecules and small-sized droplets was ionized by the 1 kHz laser pulses through multiphoton absorption. Generation of high-density charged particles of ∼10¹⁶ cm⁻³ in the self-guided ionized filaments such as N⁺, O⁺, H⁺, and OH⁺ would trigger the binary H₂O-HNO₃ nucleation [2]. However, the upward moving warm moist air would encounter the cold air above resulting in further water condensation and hence the release of latent heat. This causes the rising air to accelerate and form convection due to the decrease of the pressure in the zone above the filament center. Video captures [e.g., Fig. 2(b)] showed that the surrounding cold moist air was sucked to the low-pressure zone near the filament center and replenished the water vapor loss there. The firing of the filaments with a repetition rate of 1 kHz provided a source to generate a continuous updraft accompanied by turbulence and convection, which occurred on a much larger scale than the filament active volume. This led to a spectacular water condensation and snow formation over the entire cloud chamber as shown in Fig. 1(d) after 2 h of firing the laser. However, because the intense updraft and convection were concentrated above the filament center, the observed heap of snow was confined in an area of 2.0 cm × 1.5 cm below the filament center on the bottom base plate as shown in Fig. 1(d). As the water droplets or ice particles grew in size by a process known as the Wegener–Bergeron–Findeisen process [17], they would fall down due to their mass. But they might get caught by the intense updraft into another cycle to increase their sizes. That seemed to explain why the snowflakes in the snowpack had larger sizes as shown in Fig. 1(e).

Much clearer scattering images were recorded at different instants of time as shown in Figs. 2(c) and 2(d), respectively. It was seen that scattered water droplets

![Fig. 2](https://example.com/fig2.png)

Fig. 2. (Color online) (a) Side Mie scattering image before firing the laser filament. (b) Video capture for side Mie scattering by the laser-induced water condensation. (c)–(d) Side Mie scattering images at different instants of time. The exposure times were 1/4000 s and 1/2000 s, respectively. The figures A, B, C below (d) corresponded to the zoomed in pictures of regions A, B, C in (d), respectively. Heaps of snow are evident at the bottom of (b)–(d).
or large-sized particles with several tens to hundreds of micrometers in size were generated above the filament. The velocities of some particles might be estimated by the trailing smear of the particle in the scattering images as shown in B and C below Fig. 2(d), which were the zoomed in pictures corresponding to the regions B and C in Fig. 2(d), respectively. For example, dividing the trailing smear length by the exposure time, we could estimate that one particle with the size of ~23 μm in region B moved with a velocity of ~23 cm/s while the particle in region C moved with the velocity of ~60 cm/s. Some particle as shown in A for the region A of Fig. 2(d) had inner structures, which implied that large-sized ice particles beyond 200 μm might be generated through the laser-induced intense updraft and convection.

The effect of repeatedly chopping the 1 kHz beam with a chopper, as shown in the inset of Fig. 3, was investigated. The amount of the laser-induced snow formation below the filament center at different chopping rates such as 150, 15, 5, and 1 Hz, were measured respectively and compared with the result using the full 1 kHz beam, as shown in Fig. 2 (square symbols). Note that the total number of pulses is constant in the chopped beams over any interval. The firing duration of the femtosecond pulse trains was fixed to be 40 min. Hardly any snow was induced by the laser filaments at 1 Hz chopping rate. The dynamic motion of the updraft induced by the laser filaments was also recorded by the camera at the same time. It was found that the upward velocity of the laser-induced updraft was highly dependent on the chopping rate as shown in Fig. 3 (inverted triangle symbols). It was highly correlated with the amount of the laser-induced snow formation. At 1 kHz, the detected updraft’s velocity could be as high as ~60 cm/s while only intermittent updraft with the velocity of ~3.6 cm/s was observed at 1 Hz. These results indicated that a continuous heating of cold moist air by firing high-repetition laser pulses in the cloud chamber was essential for laser-induced water condensation and snow formation on a macroscopic scale.

In conclusion, we demonstrated large-scale laser-filamentation-induced water condensation and snow formation by firing 1 kHz femtosecond laser pulses into a laboratory diffusion cloud chamber. The generation of hygroscopic HNO₃ in the snowpack confirmed the pathway for the efficient ice nucleation owing to the photo-oxidative chemistry of nitrogen triggered by the filamentation [1–3]. Furthermore, a continuous and intense updraft of warm moist air induced by the high-repetition laser filamentation would speed up the particle–particle collisions and large-sized particles generation as it encountered the cold moist air above and via convection and cyclone-like action on a macroscopic scale. The demonstrated results indicated high-repetition laser pulses would be a better choice to induce large-scale water condensation and snow formation more efficiently.

This work was supported by the National Basic Research Program of China (contract no. 2011CB808100, 2010CB923203), National Natural Science Foundation of China (contract nos. 60921004, 10974214, 61008011), and the State Key Laboratory Program of Chinese Ministry of Science and Technology. See Leang Chin acknowledges the support of Canada Research Chairs, Natural Science and Engineering Research Council, and Quebec Fund for Nature and Technology Research.

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