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LETTER

Laser-filament-induced condensation in an inverted cloud chamber

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Abstract

We have demonstrated experimentally that firing a laser filament in a subsaturated zone of a cloud chamber of inverted temperature profile could induce water condensation. Initially, through ion chromatography performed on samples collected from a receptacle placed under the laser, the presence of HNO3 was observed. The existence of this HNO3, as well as digital camera images of very tiny droplets close to the laser axis viewed at an angle perpendicular to the chamber, suggest that the condensation could be due to the laser. A second experiment measuring the growth of the water droplets using a cooled receptacle confirmed the existence of laser-induced condensation in these conditions.

(Some figures may appear in colour only in the online journal)

1. Introduction

The recent past has shown a number of interesting developments in the field of femtosecond laser filament [1–10] induced condensation [11–21]. The Teramobile group in particular has proved that condensation can occur due to femtosecond laser filaments both inside a cloud chamber and also in the outside atmosphere [11–17]. Subsequently, more attention has been attracted to the physical and chemical interactions that underlie this newly discovered phenomenon. It was shown later that photodissociation, ionization, or electron impacts on H2O, O2 and N2 could open up chemical pathways towards the generation of up to \( \sim 10^{14} \) molecules cm\(^{-3}\) s\(^{-1}\) of HNO3 (multi-ppm range) in air [13], while in nature the molecule can only be found in the ppt range [22]. The generation of HNO3 is followed by the formation of binary H2O–HNO3 clusters that grow into condensation nuclei [11–21]. These seeds are then propelled into the surrounding environment, resulting in droplet formation. This pathway to efficient nucleation, being the most reasonable model existing currently, has also been observed more recently by the SIOM (Shanghai Institute of Optics and Fine Mechanics) group [19–21], who demonstrated that the binary H2O–HNO3 clusters can nucleate directly into ice and snow in a cloud chamber with a temperature gradient between \(-46^\circ\text{C}\) and room temperature over a vertical distance of about 20 cm. This was done by confirming the presence of HNO3 in the generated snow and ice using pH paper and ion chromatography. It was also found that nucleation in the active filament volume alone is not sufficient to sustain efficient droplet/ice/snow formation. By changing the chopping rate of the 1 kHz laser beam, they noticed that efficient condensation also depended on the strong turbulence surrounding the filament. This turbulence helps propagate the condensation nuclei to zones farther away from the filament, where the water vapor concentration could be more significant, leading to even greater condensation. Also, more recently, outdoor experiments done by the Teramobile group have confirmed that it is possible to induce condensation using a laser in subsaturated conditions at a relative humidity as low as 75% [15].

The cloud chambers in all the previous experiments until now were based on a design in which the cold plate is placed at the bottom; hence the temperature increases from...
bottom to top. In those previous experiments, subsaturated and more ‘natural’ conditions have not been explored. In this work, the main objective is to investigate the laser-induced effects under an inverted temperature gradient (cold at the top, warm at the bottom—‘imitating’ the natural environment in a mini-laboratory scale). In this case, the cold plate was placed at the top of the chamber, and in doing so it was possible to observe nucleation by firing the laser through a subsaturated zone (∼60% RH). It is to be noted that, in this new configuration, it is impossible to quantitatively compare these results with previous experiments—in particular, the previous experiment in SIOM described in [19–21]. Many parameters, such as the cold plate’s minimal operating temperature (∼18°C in our case versus ∼46°C in the SIOM case [19]), the laser pulse energy (1.95 mJ versus 9 mJ), or even the thermodynamic conditions inside the chamber are different. Most importantly, they fired the laser in a supersaturated zone (∼127%RH at −29°C) while it is a subsaturated zone in our case.

In this work, the inverted temperature gradient was approached by two sets of experiments: in the first, only the mass of NO$_3^−$ ions coming from condensation was measured through ion chromatography for ∼1.5 h of laser irradiation at different distances from the cold plate. The water mass could not be measured in this first experiment due to rapid evaporation of the droplets falling into the receptacle. The ion chromatography results suggested that condensation was induced by the laser filaments. In the second experiment, a cold finger was added to the bottom of the receptacle to obtain an estimate of the total growth in water droplets inside the receptacle for ∼3 h of laser irradiation at a distance of ∼1 cm from the cold plate. The cold finger was used to help preserve the droplets inside the receptacle. In the second case we detected an increase in water droplets when the laser was used.

2. Experimental setup

The experiments were conducted using an 800 nm Ti–sapphire laser (Spectra Physics, Spitfire) emitting pulses of 1.95 mJ/40 fs at a repetition rate of 1 kHz (figure 1). The laser pulses were focused into the chamber using a plano-convex lens of 50 cm focal length. A filament of ∼2 cm length was created inside the cloud chamber. A probe CW laser beam at 532 nm wavelength of 8 mm diameter and 4 W output power (Coherent, Verdi) was also used to illuminate the interaction zone inside the chamber. Scattering of the probe beam in the interaction zone was recorded using a digital camera. A computer-controlled translation stage was used to control the beam height. A plastic receptacle ∼10 cm in diameter and ∼15 mm in height was placed ∼1 mm below the filament in the middle of the chamber to collect any condensation residue. The height of this receptacle was adjusted manually when the filament height was changed such that the distance between the filament zone and the receptacle was unchanged. Two chamber designs were used for the experiments: a glass container (insulated using foam) measuring 59 × 29 × 52 cm$^3$ and a plastic (Plexiglas) container measuring 26 × 18 × 30 cm$^3$. The top side of these chambers was covered with a 3 mm thick aluminum plate (insulated) in which a thermoelectric cold plate (model CP-200HT-TT, TE technologies) of surface area 21.6 × 16.1 cm$^2$ was inserted. The advantage of this type of cold plate was that it could be placed in any direction or position and its performance would remain the same. The cold plate was fully computer controlled and the lowest temperature attainable on the cold plate during the experiments was −18°C. The chambers were designed to let in air from the outside through gaps at the bottom to reduce the overall water concentration inside and thus obtain a subsaturated atmosphere. Also, water was constantly heated slightly and kept at a temperature of ∼35°C in a beaker at the bottom to keep the relative humidity stable. Inside the chamber, a thermometer was used to measure the temperature and another sensor was used to scan the relative humidity and temperature profiles in the chamber.

3. Methods and analysis

Temperature and humidity distributions were measured in the glass chamber prior to the experiments and can be seen in figures 2(a) and (b). Figure 2(a) shows the case where the air flow was sealed at the bottom of the chamber. The position of the humidity sensor was changed every half-hour to a new position. The first 10 min helped stabilize the sensor and, from that point on, the temperature and humidity were measured 10 min later and 20 min later, completing the half-hour period. The errors of these measurements were determined by looking at their fluctuation over 20 min. The humidity sensor, due to its physical size, had to be placed at a minimal distance of 5 mm from the cold plate. We can notice that the error bars are much higher when farther away from the plate. This indicates that the ‘natural’ turbulence in the chamber causes the value of the relative humidity to fluctuate as we move away from the cold plate. For this reason the laser irradiation was carried out at a distance smaller than or equal to ∼1.5 cm. The relative humidity in the lab was measured to be ∼24%. Nevertheless, it is interesting to see the relative humidity drop rapidly and
stabilize at a mean value of 40% inside the chamber as the sensor was moved far away from the cold plate.

The receptacles were cleaned in advance using ultrapure water to remove any dust particles or charged molecules. For the first set of experiments inside the insulated glass chamber, the nitrate mass measurements were performed in two steps; the condensate in the receptacle was first collected without any irradiation following a period of 90 min, then accumulated in another identical receptacle after irradiation for the same amount of time. Since the cold plate was removable, the snow and ice on the cold plate was melted and collected separately. These four samples were then taken to a chemistry lab and prepared for the ion chromatography tests. The irradiations were done at distances of 0.5, 1.0 cm (0.9 cm for the closed chamber), and 1.5 cm from the cold plate for both closed and open conditions of the cloud chamber. The receptacle was manually positioned accordingly each time.

A digital camera was used to record the Mie side scattering of the probe beam in the chamber during each experiment. We could visually observe turbulence created by the filament similar to that observed in the SIOM experiment [19]. Figure 2(c) shows the condition inside the closed chamber without irradiation and figure 2(d) the effects of irradiation. The pictures were taken from a video that was...
recorded at 14 frames s$^{-1}$. We can see in figure 2(c) traces made by droplets generated in the cool saturated zone and traveling in random directions with random velocities. On the other hand, a filament induces small mist packets that travel towards the direction from which the laser is incoming. In the case of figure 2(d), the mist packets travel towards the left. Many of these droplets fell into the receptacle but many flew away. During the experiment, the scattering due to the natural and filament-induced turbulence in the chamber have been recorded and have also been seen by the naked eye. The trail after the mist packet is due to the very low frame rate and exposure of the camera used for the experiment.

On the receptacle, during and after irradiation, very small droplets could be clearly identified by eye, most of which tend to evaporate as soon as they appear, since the temperature at the receptacle height is not low enough to conserve them. Some droplets could be seen at the bottom of the receptacle in figure 2(d). The droplets must have come from the supersaturated region near the cold plate after the filament-induced HNO$_3$ molecules were generated and dispersed throughout the inverted chamber [11, 19]. In spite of the fact that very few measurable water droplets were left in the receptacle once it was taken out of the cloud chamber, we thought that if the filament-induced water droplets had really fallen into the receptacle and then evaporated, the evaporation should have left behind some HNO$_3$. The latter is the seed of nucleation and condensation of the water droplets, according to recent work from the Teramobile group [11–17] and the work in SIOM [19]. Thus, the ‘empty’ receptacle was rinsed using ultrapure water (Nanowater) and the whole was transferred to a cleaned plastic container and diluted to a volume of 6 ml. Then three replicas were made from this solution and used for the ion chromatography measurements to provide absolute concentrations (mg l$^{-1}$) of NO$_3^-$ in each sample, from which the absolute mass of HNO$_3$ was obtained from the knowledge of each sample’s volume. Measurements were done on these samples using an ion chromatograph (Dionex Model ICS 1000 equipped with a Model AS40 automated sampler, Model IonPacAS14A 4 mm × 250 mm anion-exchange column and Model DS6 Heated conductivity cell detector) to determine the mass of NO$_3^-$ present inside the receptacle and on the cold plate. The ion chromatograph was calibrated using aqueous potassium nitrate (KNO$_3$) standards. The results are shown in figures 2(e) and (f). The error bars (±1σ) were obtained using the uncertainty on the slope and intercept of the calibration curve calculated by linear regression. The reactions in the oxidative chemistry of nitrogen triggered by filamentation during the generation of HNO$_3$ have been previously proposed and confirmed by Kasprian et al [11–17] and Ju et al [19–21], respectively. The next paragraph gives a comparison of the production of HNO$_3$ in these two experiments [13, 19] together with the current one.

The number of molecules of HNO$_3$ created per second in the active filament volume was estimated from the data available in [13, 19] and figure 2(e). These values are presented in table 1. For the Teramobile experiment, one has to use the amount of molecules of NO$_2$ ($3 \times 10^{13}$ molecules/pulse) given in [13]. By assuming that all the NO$_2$ gets converted into HNO$_3$, it is possible to obtain an upper limit for the number of molecules of HNO$_3$ ($6 \times 10^{14}$ molecules/pulse) per laser pulse in the active filament volume; hence, $6 \times 10^{14}$ molecules s$^{-1}$ taking into account the 10 Hz repetition rate of their laser beam. For the case of SIOM and this experiment, it is only possible to get a lower limit on the production of HNO$_3$ molecules due to the fact that these two cases made use of a cloud chamber. In a cloud chamber, not all the HNO$_3$ could be measured since an unknown amount of this HNO$_3$ attached itself to various surfaces inside the chamber—and in the case of the open chamber it could even escape. For the SIOM case, the HNO$_3$ production is calculated using the values of the mass density of the snow pile that is amassed below the filament after the laser irradiation (13 mg over a surface of 1.5 cm × 2.0 cm) and the HNO$_3$ concentration (0.032 mol l$^{-1}$) [19]. The HNO$_3$ molecules accumulated on the surface of the cold plate outside the snow pile was significant, judging from the observation that the pH values in the snow pile and outside were similar (pH = 2 and 3 respectively). It is thus believed that the value of the rate quoted in table 1 is much smaller than the total rate. In the current experiment, only the HNO$_3$ measured on the cold plate and in the receptacle have been taken into account, and the values ~0.014 mg (cold plate) and ~0.004 mg (receptacle) from figure 2(e) are used for the estimation. Obviously, the production rate of HNO$_3$ was higher in the two cases using 1 kHz beams.

The data in figures 2(e) and (f) once again confirm the same observations as in [19]—i.e. filamentation did induce the formation of HNO$_3$—and it is possible to check this by noticing that in the absence of the filament, the amount of HNO$_3$ found in the receptacle and cold plate is nil (bottom lines, figures 2(e) and (f)). As the filament is moved away from the cold plate, the amount of NO$_3^-$ found on the cold plate decreases and inside the receptacle it remains almost constant for both cases of closed and open chambers. Our explanation as to what happens in figures 2(e) and (f) is based on the premise that the filament-induced turbulence remains more or less constant at all heights. When the filament is near the cold plate, the humidity is higher and a greater quantity of HNO$_3$ is produced. Also, the proximity to the cold plate

<table>
<thead>
<tr>
<th>Research group</th>
<th>Production of HNO$_3$ in filament volume (molecules s$^{-1}$)</th>
<th>Laser conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>COPL</td>
<td>$&gt;3.2 \times 10^{10}$, from cold plate and receptacle</td>
<td>1 kHz, 1.95 ml/pulse, 40 fs</td>
</tr>
<tr>
<td>SIOM [19]</td>
<td>$\gg1.4 \times 10^{14}$, from snow pile on cold plate</td>
<td>1 kHz, 9 ml/pulse, 50 fs</td>
</tr>
<tr>
<td>Teramobile [13]</td>
<td>$\sim6 \times 10^{14}$, direct measurement using analyzer</td>
<td>10 Hz, 12 ml/pulse, 80 fs</td>
</tr>
</tbody>
</table>


The following model is proposed in the current work to explain the increase in water condensation that has been observed inside the chamber. As the femtosecond laser pulse is fired into the chamber, a filament is created in the middle of the chamber. The filament, through various ionization processes such as multiphoton-absorption and photodissociation, splits the individual molecular components of air such as O₂, N₂, CO₂, and H₂O. Through the oxidative chemistry process proposed by the Teramobile group [13], binary clusters of HNO₃–H₂O are formed. These clusters grow into condensation nuclei in the proximity of the active filament volume and are then propelled throughout the chamber with the help of the heavy turbulence induced around the filament [11] due to the high repetition rate, as observed by the SIOM group [19–21]. Large quantities of these nuclei are sent upwards due to convection induced by the high local temperature around the filament. They then reach the saturation zone near the cold plate, where they are cooled and grow into larger droplets due to the high water vapor concentration. A large proportion of these droplets ends up condensing on the cold plate, while a smaller proportion ends up falling down—some onto the receptacle, as shown by the increase in water mass. Thus, nuclei created in a subsaturated zone can be sent up to the saturated and supersaturated zones and result in condensation through the process described above.

### 4. Conclusion

High repetition rate (1 kHz) femtosecond laser filaments were generated inside a subsaturated zone (60% relative humidity) in an inverted cloud chamber (cold at the top, warm at the bottom, mimicking the ‘natural’ condition in a mini-scale). Significant water condensation was observed. Ion chromatography of the condensate indicated that binary clusters of H₂O–HNO₃ were formed in the proximity of the filament. They were then propelled by the turbulence induced by the filaments into the upper saturated and supersaturated zones to form droplets of water.

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**Table 2.** Mass measurements of condensate found inside the receptacle in the presence and absence of filaments (second experiment).

<table>
<thead>
<tr>
<th>Water bath temperature (°C)</th>
<th>Mass of liquid without filament (g)</th>
<th>Mass of liquid with filament (g)</th>
<th>Mass difference (g)</th>
<th>Nitrate mass inside receptacle with filament (mg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>35</td>
<td>0.8536</td>
<td>1.7539</td>
<td>0.9003</td>
<td>0.0137</td>
</tr>
</tbody>
</table>
Technical support from Mr M Martin and Ms M Marois is also acknowledged. Zhizhan Xu and Ruxin Li acknowledge the support of the National Basic Research Program of China (Contract No: 2011CB808102) and the National Natural Science Foundation of China (Contract No: 60921004).

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