In air, self-focusing of femtosecond laser beam occurs when its initial power is larger than $P_{cr} = 3.72\lambda_0^2/8\pi n_0 n_2$, where $\lambda_0$ is the incident laser wavelength, $n_0$ the linear refraction index and $n_2$ the coefficient of nonlinear Kerr index [1]. Through multiphoton ionization or tunneling ionization, a high density plasma ($1 \times 10^{16} \text{ cm}^{-3}$) is initiated [2]. Instantaneously the plasma involves a local reduction of refraction index and led the plasma defocusing effect. The self-focusing and plasma-defocusing effects repeat themselves resulting in a plasma channel we call filament, with its initiated position, length etc. being free to control. In the past few years, femtosecond filamentation has been proposed as an alternative technique for weather modification in the future [3]. However, before that, underlying physical mechanisms are still waiting to be discovered.

In the present experiment, a 1 Hz 660 mJ/27 fs 800 nm laser system was employed. It initiated a bunch of multiple filaments in about 10 cm long in the middle of an open cloud chamber ($50 \times 50 \times 20 \text{ cm}^3$). Temperature ($T$) and relative humidity (RH) around the filaments were measured of $T = -5 \pm 1^\circ \text{C}$ and $RH = 85 \pm 5\%$. A high speed camera (PCO Dimax HS4-32) recorded time evolution of water condensation around the filament zone from the side direction. Shock wave-assisted water condensation was observed for the first time, which appeared as a “cloud” line along the filament zone after laser was gone in 500 $\mu$s. The observed maximum size of the “cloud” droplets was around 300–400 $\mu$m, which was much larger than that of background particles (10–30 $\mu$m). It’s noteworthy that those “cloud” droplets stayed steadily along the filament zone, without being influenced by the natural convection at all. However, soon afterwards they evaporated completely in 4.0 ms.

Thermodynamic airflow motion stirred up around the filament zone was also studied. Experimental results shown it was accelerated sharply during the first 120 ms after the incident laser was gone (shock wave-assisted water condensation had already evaporated away). Its maximum velocity reached about 45 cm/s in 120 ms. After that, it started to slow down and in 500 ms, the thermodynamic airflow motion died out totally, leaving condensed droplets behind following the natural convection motion.

Measurement from a particle sizer (SMPS 3836, TSI) shown, in dry air ($T = 20^\circ \text{C}, RH = 40\%$), in 5 min, up to $2.25 \times 10^6 \text{ cm}^{-3}$ aerosols were produced by the 1 Hz filaments. It corresponded to about $7.5 \times 10^5 \text{ cm}^{-3}$ aerosols produced by each laser shot. Size of the laser-induced aerosols was among 40–100 nm. The maximum number density was of $9 \times 10^5 \text{ cm}^{-3}$ at 55 nm. Serious studies had shown they were mainly hygroscopic salts of $\text{NH}_4\text{NO}_3$ or $\text{H}_2\text{O-HNO}_3$, which seeded the “clouds” efficiently as long as the relative humidity was larger than 70% [4].

It’s known well that ever since the plasma being generated in fs time scale, ionized molecules collided with each other inside the filament which produced the hygroscopic aerosols in ps/ns time scale [5]. Then plasmas recombined back and deposited energy into fluorescence irradiation, molecular fragments, photo-oxidation reactions, thermal release etc. [4]. In the present experiment, laser-induced aerosols together with background aerosols were ready to be activated as cloud condensation nuclei (CCN) in ps time scale. The thermal release caused sudden thermal expansion inside the filament zone, during which shock waves were generated in $\mu$s time scale [6]. The shock wave propagated outwards under giga-pascals pressure [6]. In hence the surrounding humid air was squeezed into a high super-saturated zone. Water condensation was quickly triggered in both homogeneous and heterogeneous way in $\mu$s time scale. This agreed with the
observed “cloud” line in 500 µs after the incident laser was gone in the present experiment. Along with the thermal expansion inside the filament zone, a low density area was left behind. As reported, this thermal resulted low density area lasted up to 90 ms [7]. It sucked in air around and accelerated it quickly under the sharp density difference. In the present experiment, the lifetime of the low density area was supposed to be around 120 ms, which was a bit larger than the measurement given before. This might be due to the relative high laser pulse energy used here, so that more energy was deposited into thermal release. As the low density area decayed back to normal, thermodynamic airflow motion around died out.

In summary, we studied laser-induced water condensation in a time resolved way. It shown the water condensation began with the high density laser-induced aerosol formation (up to $2.25 \times 10^6 \text{ cm}^{-3}$ in 5 min). They were activated by shock wave-assisted super-saturated zone around the filaments in µs time scale. Size of the condensed droplets reached 300–400 µm, which was large enough to trigger precipitation through angulation in the practical cloud seeding. However, they evaporated away in 4.0 ms. This result indicated that in the real application, to create continuous shock-wave assisted large size droplets, femtosecond laser with pulse repetition rate of $> 1 \text{ kHz}$ is required. Thermodynamic airflow motion lived much longer time. It was accelerated in the first 120 ms to about 45 cm/s. It might also contribute to push the condensed particle growing further into precipitation, by enhancing collision between hygroscopic aerosols and water vapor and speeding up mixing of humid air with different temperature [5]. Our results are essential for both physical understanding and practical application of femtosecond laser technique for weather modification in the future.

References