LIBS-LIDAR USING FEMTOSECOND TERAWATT LASER FOR MEASUREMENT OF CONSTITUENT OF MICROPARTICLES IN AIR

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ABSTRACT

We experimentally demonstrated the remote sensing of constituents of microparticles in air by laser-induced-breakdown-spectroscopy (LIBS) LIDAR using femtosecond terawatt laser pulses. Laser pulses of 70 fs duration and 130 mJ energy generated filaments when focused at a focal length of 20 m, and were irradiated onto artificial salt water aerosols in air at a 10 Hz pulse repetition rate. Na fluorescence was observed remotely at a distance of 16 m using a 318-mm-diameter Newtonian telescope, a spectrometer, and an ICCD camera. These results show the possibility of the remote measurement of the constituents of atmospheric particles, such as aerosols and clouds, by LIBS-LIDAR using femtosecond terawatt laser pulses.

1. INTRODUCTION

Remote identification of the chemical composition of atmospheric aerosols and clouds is attractive for the study of acid rain and for understanding cloud-aerosol interactions, which are important in developing global climate models [1]. Moreover, remote detection and identification of microparticles in air is desired to monitor toxic materials distributed in the atmosphere.

LIDAR is a powerful tool for remote measurement of aerosols and clouds, in terms of parameters such as extinction coefficient, particle size distribution and shape [2]. However, measurement of the constituents of aerosols and clouds has not been achieved by ordinary nanosecond LIDAR. Recently, ultrashort-pulse high-intensity Ti:sapphire lasers have been developed, and several applications of these lasers to LIDAR have been reported [2-7]. Multiphoton absorption occurs due to the high peak power when ultrashort, high-intensity laser pulses are irradiated onto aerosols. Using this effect, remote detection and identification of bio-aerosols by laser-induced fluorescence (LIF) LIDAR have been reported [6].

Laser-induced breakdown spectroscopy (LIBS) is attractive for analyzing the composition of all types of materials without wavelength tuning. In LIBS, the laser beam has to be focused on the targets to obtain high energy density enough to ablate them. However, in the case of remote sensing for a long distance, the focusing spot becomes large. In addition, since the focusing range is limited by the Rayleigh length, the plasma can only be created over short ranges, which include only a small portion of targets such as microparticles in air.

The propagation of an ultrashort high-intensity laser pulse in the atmosphere produces a bundle of filaments owing to the equilibrium between Kerr-lens focusing and plasma defocusing [8-12]. Filaments can be generated for a long distance as far as 2 km [13] and a single filament propagates for several tens of meters with a high intensity of the order of 10^{13} W/cm^2 [8]. Since this laser intensity is larger than the threshold for producing plasmas, filaments can generate plasma emission on the surface of a target far from the laser. Therefore, filaments are very useful for remote LIBS. Remote sensing of the constituents of metals using filaments has been demonstrated [14]. In the case of the measurement of constituents of microparticles in air, the use of filaments has a greater advantage because a bundle of filaments can ionize a large number of microparticles in air, which can realize LIDAR measurements.

Here we report the first demonstration of remote sensing of constituents contained in microparticles in air by LIBS-LIDAR using filaments generated by femtosecond terawatt laser pulses.

2. EXPERIMENTAL SETUP

The experimental setup is shown in Fig. 1. We used artificial salt water aerosols, which were generated from salt water (300g/L) using an ultrasonic humidifier, as a target for measurement simulating sea-salt particles. The diameter of aerosol particles was measured to be below 10 μm using a particle analyzer (Oxford lasers;

VisiSizer). Generated salt water aerosols were introduced in a tube with an inner diameter of 20 cm and a length of 5 m.

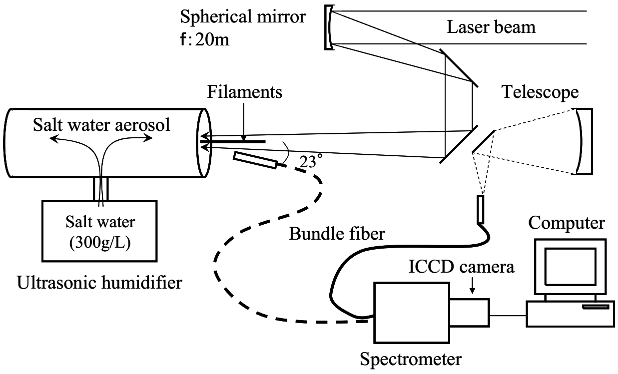

Fig. 1. Experimental setup for LIBS-LIDAR measurement of Na fluorescence from salt water aerosols irradiated with filaments generated by femtosecond laser pulses.

The measurements of Na fluorescence from aerosols were performed using a Ti:Al_2O_3 laser (Thales laser, Alpha 10/US-20TW), which is based on the chirp pulse amplification (CPA) technique. Laser pulses of 130 mJ energy, 70 fs pulse width, 2 TW peak power, and 10 Hz repetition rate were focused using a spherical mirror of 20 m focal length, and irradiated onto the salt water aerosols. The laser beam was focused at the end of the tube. However, salt water aerosols were so dense that laser beams almost disappeared after propagating for about 2 m in the tube.

LIDAR measurements of the fluorescence from the salt water aerosols were performed using a Newtonian telescope located at the back of the final laser-beam-sending mirror located 16 m apart from the entrance of the tube. The collected light by the telescope was fed into the spectrometer (Jobin Ybon, HR460) through the bundle fiber, and detected by the ICCD camera (Andor, DH734-18F-03). The specifications of the LIDAR receiver were shown in Table 1.

3. EXPERIMENTAL RESULTS

3.1 Filament generation

Figure 2 shows the laser beam propagating around the entrance of the tube. Although the photo was taken with the accumulation of several shots, the propagation of each filament was clearly observed. A laser beam profile at the entrance of the tube is inserted in Fig. 2. Although the laser beam was not focused at this point, multiple filaments were observed as several bright spots in the beam profile. These results show that femtosecond terawatt laser pulses produced a number of filaments in a large beam profile for a long distance, which is a great advantage for the measurement of atmospheric microparticles by LIBS-LIDAR.

Table 1. Specifications of LIDAR receiver.

Telescope	
Type	Newtonian
Primary mirror diameter	318 mm
Focusing length	1.5 m
Bundle fiber	
Input size	φ3.37 mm
Output size	400 μm×20 mm
Each fiber core diameter	210 μm
Numerical aperture	0.37
Length	3 m
Spectrometer	
Focusing length	460 mm
Grating size	76x76 mm
Aperture ratio	f/5.3
ICCD camera	
Quantum efficiency at 589nm	10 %
Dark current	2.4 e-/p/s
CCD format	1024x1024
Pixel size	13 μm

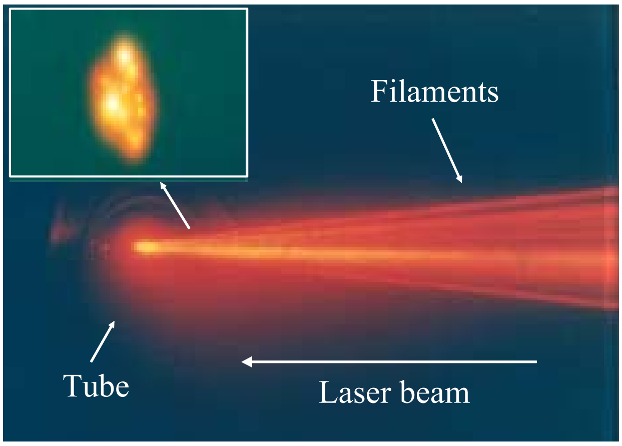

Fig. 2. Laser beam propagating around the entrance of the tube. The insert is a laser beam profile showing multiple filaments at the entrance of the tube.

3.2 Fluorescence measurement of salt water aerosols

For the preliminary experiments of the LIDAR measurements, the fluorescence from the salt water aerosols was directly measured at the entrance of the tube. At this point, the laser beam was not focused and a lot of filaments were generated as shown in Fig. 2. The fluorescence was collected by a bundle fiber set at an angle of 23 degrees from the backward direction of the laser, as shown with a dotted line in Fig. 1. The collected light was fed into a spectrometer and detected by an ICCD camera.

Figure 3 shows the spectroscopic measurement results of Na fluorescence from salt water aerosols irradiated with filaments generated by femtosecond laser pulses along with the emission of a spirit lamp containing salt obtained for a reference. Since the laser shot timing had a jitter of 10 ns, the gate width of the ICCD camera was set at 20 ns. The delay time of the ICCD camera gate timing from the laser shot was determined by the following manner. First, the center wavelength of the spectrometer was set at 800 nm. Then the gate timing of the ICCD camera, at which the fundamental wavelength of the Ti:Al_2O_3 laser and the white light emission generated by self-phase modulation of the laser pulses was observed most strongly, was defined to be delay zero. At zero delay, the white light emission was so strong that Na fluorescence could not be observed. The spectra obtained with delay times of 20 ns, 40 ns, and 120 ns are shown in Fig. 3 (a). The fluorescence signals for 1,000 laser pulses were accumulated for each measurement. The Na fluorescence is clearly observed at the 20 ns delay time due to the reduction of the white light emission, and D_1 and D_2 lines of Na were observed as well as from the spirit lamp as shown in Fig. 3 (b). These results show that Na fluorescence from plasma produced by irradiating laser pulses onto salt water aerosols was observed. Na fluorescence becomes weaker at the 40 ns delay time, and almost no fluorescence is observed at the 120 ns delay time. The signal intensity at longer wavelength increases due to white light generated by self-phase modulation of laser pulses at the 20 ns delay time, and decreases at longer delay times. These results show that it is possible to identify the salt water aerosols by LIBS using laser-produced filaments.

3.3 Lidar measurement

LIDAR measurements of constituents of salt water aerosols using filaments were performed. Figure 4 shows the spectroscopic measurement results of Na fluorescence from salt water aerosols subtracting background obtained without the aerosols and laser pulses. For each measurement of LIDAR and background, signals for 10,000 laser pulses were accumulated. D_1 and D_2 lines of Na were observed for the LIDAR measurements at the 20 ns delay time of the ICCD camera gate timing as well as from the spirit lamp shown in Fig. 3 (b).

As shown above, we succeeded in remote sensing of constituents of microparticles in air by LIBS-LIDAR using filaments generated by femtosecond terawatt laser pulses. The methods mentioned above can be used for the remote measurement of the constituents of atmospheric aerosols and clouds. In the case of aerosol measurements by LIDAR, white light emission generated from laser pulses after passing through the

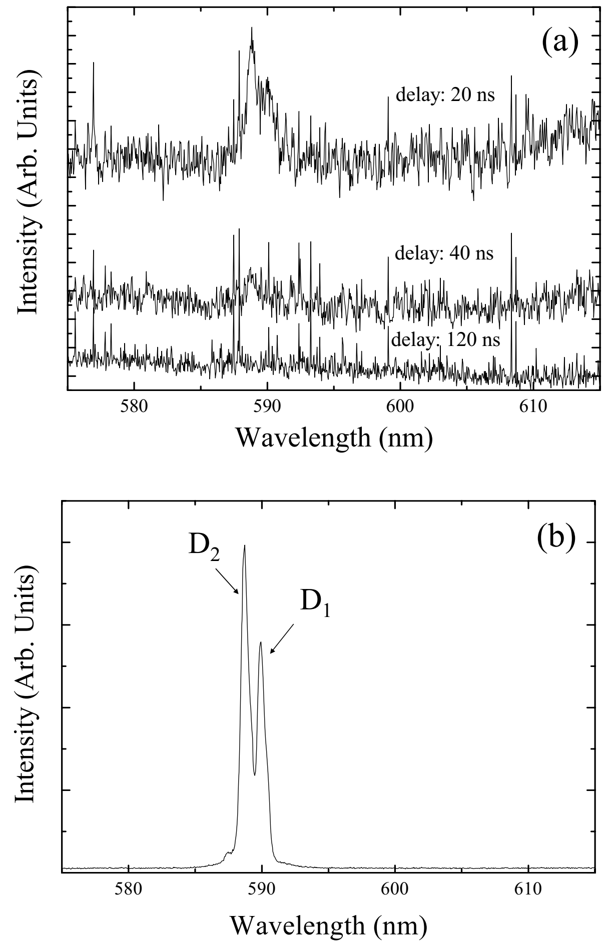

Fig. 3. Spectroscopic measurement results of Na fluorescence from (a) salt water aerosols irradiated with filaments generated by femtosecond laser pulses with delay time of 20, 40, and 120 ns from laser irradiation, and (b) spirit lamp containing salt.

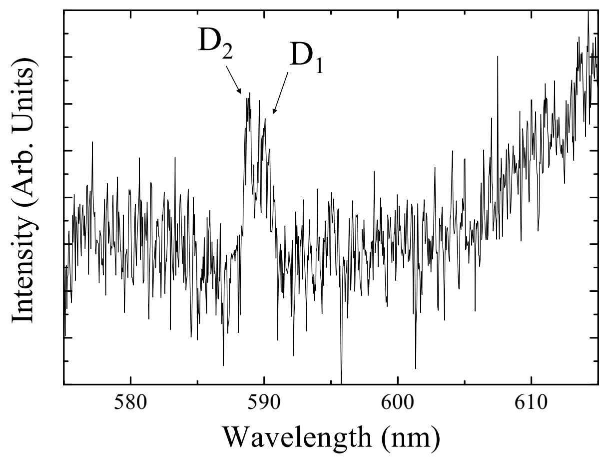

Fig. 4. Na fluorescence from salt water aerosols irradiated with filaments generated by femtosecond laser pulses measured by LIBS-LIDAR.

measurement point may cause considerable noise for the measurements if a coaxial configuration of the laser beam and telescope is used as described in this paper. However, for example, a biaxial or bistatic configuration can solve the problem.

4. CONCLUSIONS

In conclusion, we experimentally demonstrated the remote sensing of constituents of microparticles in air by LIBS-LIDAR using femtosecond terawatt laser pulses. Laser pulses of 70 fs duration and 130 mJ energy generated filaments when focused at a focal length of 20 m, and were irradiated onto artificial salt water aerosols in air at a 10 Hz pulse repetition rate. Na fluorescence was observed remotely at a distance of 16 m using a 318-mm-diameter Newtonian telescope, a spectrometer, and an ICCD camera. The signal intensity of white light emission generated by self-phase modulation of laser pulses was reduced by adjusting the delay time of the ICCD camera gate timing from the laser shot. These results show the possibility of the remote measurement of the constituents of atmospheric microparticles, such as aerosols, clouds, and toxic materials, by LIBS-LIDAR using femtosecond terawatt laser pulses. Moreover, remote detection and identification of bioweapons as a defense measure against terrorism, for which recently there has been a strong need, should be an attractive candidate for the application of this technique.

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