RAPID COMMUNICATION

Enhancement of third harmonic generation in air filamentation using obstacles*

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The intensity of third harmonic emission in air filamentation disturbed by copper fibers and alcohol droplets has been investigated experimentally. Enhancement of the third harmonic emission up to more than one order of magnitude has been observed. The physical mechanism of third harmonic enhancement is attributed to suppression of the destructive interference by comparison of the experimental results and it is closely related to the type, size, and relative position of the obstacles.

Keywords: third harmonic emission, filamentation, femtosecond laser pulse

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1. Introduction

In the last decade, propagation of intense femtosecond laser pulse in air has been studied extensively due to the generation of filamentation and other interesting phenomenon such as supercontinuum and high order harmonic generation.[1–4] One of the promising applications of the filamentation is to obtain coherent ultraviolet laser pulses with durations of a few femtoseconds.[5–7] Limitation of achieving such high efficient harmonic generation using laser pulse is the interaction length as well as laser intensity in the interaction zone. While in laser filamentation, the formation of a long plasma channel and the intensity clamping provide both conditions at the same time. Therefore, third-harmonic (TH) emission, which is the lowest order harmonic generation in two color filamentation, has been studied intensively in the past few years. Much effort has been paid on increasing the TH conversion efficiency in air filamentation. TH conversion efficiency up to 0.2% has been reported till now by controlling different initial conditions.[8–11] However it is still too weak for practical applications.

Recently, enhancement of TH emission up to two orders of magnitude in air filamentation has been reported in the numerical simulations and the experiments respectively by inducing different disturbances. The physics of the enhancement effect is not quite clear, leading to different interpretations of the phenomenon. For example, it has been demonstrated that TH emission generated in filamentation has axial and off-axis components. The enhancement of TH emission is attributed to the reconstruction of filamentation and diffraction of axial TH to the background by the insertion of obstacles.[12–15] Some interpret the enhancement effect in other aspects which are based on the Gouy phase shift effect[16] of focused Gaussian beam. TH emission generated before and after the geometrical focus in filamentation would interfere destructively. Hence, introduction of the obstacles dramatically suppresses the destructive interference of TH emission with the result of a significant enhancement of TH emission.[17–19]

In this paper, we experimentally investigate the TH emission in air filamentation disturbed by copper fibers and alcohol droplets respectively. Enhancement of the TH emission up to one order of magnitude has been observed. Comparison of the experimental results shows that the enhancement of TH emission is mainly attributed to the suppression of the destructive interference in filamentation. Apart from the kind of obstacle, the size and relative position of the introduced obstacles also play an important role in enhancement of TH emission.

2. Experiment setup

The experiment is conducted using a Ti: sapphire chirp pulse amplification (CPA) laser system,[20] which has a central wavelength of 800 nm and a 10 Hz repetition rate. Laser pulse duration measured by an auto-correlator is about 60 fs. Figure 1(a) shows the schematic experimental setup. A femtosecond laser pulse is focused by a lens with a focal length of 2 m to generate filamentation near the focus. TH emission is generated in the filamentation and it co-propagates with

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the fundamental pulse. After the filamentation, the transmitted beam is collected by a fused silicon lens to the slit of the spectrometer (Model: PI Acton INS-150-252F). The lens is large enough for collecting TH emission whether there is obstacle or not. The fundamental pulse is blocked by a high reflected mirror around 800 nm and appropriate neutral density filters are put in front of the spectrometer to avoid saturation. In the experiment, naked copper fibers and alcohol droplets are used as obstacles. The droplets are generated by a droplets generator (MicroDrop technologies), which consists of a dispenser head (Model: MD-K-130) and a control electronics (Model: MD-E-201-H). The size of droplets is about 70 μm, which is the same as that of the copper fibers, by choosing the dispenser head. Both the fibers and droplets are placed in the center of the filamentation core by a three-dimensional motorized stage (SGSP series, Sigma Koki) with a moving step of 10 μm. The repetition rate of dropping droplets and the temporal delay between the trigger signal and the dropping droplets are adjusted by the control electronics to make sure that they can interact with every single laser pulse temporally.

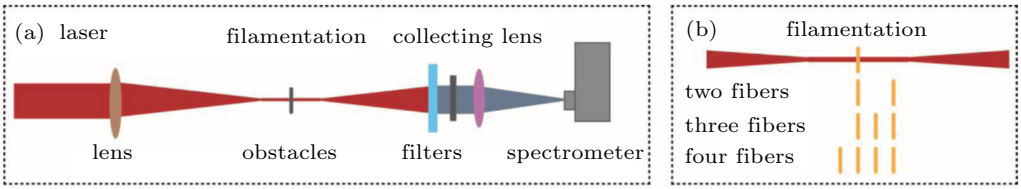


Fig. 1. (color online) (a) Schematic experimental setup; (b) placement of the multi-fibers.

3. Results and discussion

Laser filamentation is formed by focusing a femtosecond laser pulse in air with an initial energy of 10 mJ. The filamentation extends about 15 cm and the strongest fluorescence emission appears at a distance 207 cm away from the focal lens. The spectral distribution of the initial laser pulse is shown in Fig. 2(a). The central wavelength of the fundamental pulse is measured to be 798 nm, with a spectral width (FWHM) of about 19 nm. So the central wavelength of the generated third harmonic emission is about 266 nm, as shown in Fig. 2(b).

Figure 2(b) presents the intensity of the TH emission with thin copper fibers. The TH intensity of the undisturbed case is also presented (solid line) for comparison. Firstly, only one copper fiber is used as the disturbance and it is placed at 207 cm where the relative intensity of fluorescence is highest. Intensity of the TH emission is observed to be 5 times in magnitude compared with that of the undisturbed case as expected. The results are the same as those reported in Ref. [21].

Then multi-fibers are used to try to increase the intensity of TH emission. The detailed arrangement of multi-fibers is shown in Fig. 1(b). When two fibers are used, they are placed at 207 cm and 211 cm respectively; in the case of three and four fibers, a fiber is added at 209 cm and then 205 cm respectively. As shown in Fig. 2(b), enhancement of TH intensity can be observed with multi-fibers (two and three fibers) over that without obstacles. But the enhancement effect weakens when more fibers are used. If the enhancement is mainly attributed to reconstruction of filamentation or diffraction of the axial TH to background, more fibers introduce multiple circles of reconstruction of filamentation and eventually more TH emission is diffracted. So the intensity of TH emission in the far field should be higher. However, it is not shown in the experimental results. In our opinion, TH emission reaches its maximum in filamentation when it is disturbed by the first fiber in the near focus and its

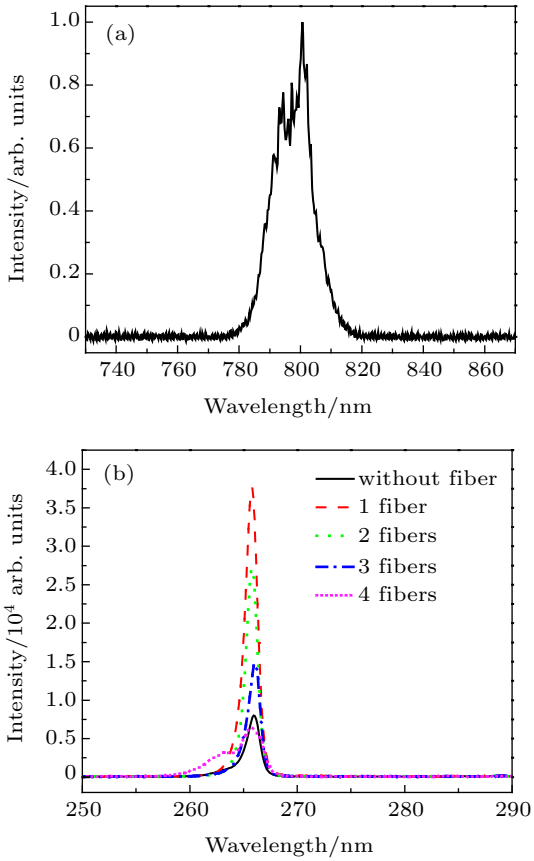


Fig. 2. (color online) (a) Spectral distribution of the fundamental pulse. (b) Intensity of TH emission disturbed by thin copper fibers: no disturbances (solid line); single fiber (dash line); two fibers (dot line); three fibers (dash-dotted line); four fibers (short dot line).

further propagation is affected by the following fibers. Especially in the last case with four fibers, when there is a fiber before the position of brightest fluorescence, the intensity of TH emission is even lower than that of the undisturbed case because TH emission does not reach its maximum in the first process. Therefore, suppression of the destructive interference plays the dominant role in enhancement of TH emission in the experiment.

Although enhancement of TH emission can be obtained using copper fibers, this approach has the limitation that the copper fiber is not only a disturbance but also a block to the propagation of light. In order to further increase the conversion efficiency of the TH emission in air filamentation, transparent media like alcohol droplet is also used as disturbance in the experiment. Figure 3 shows enhancement of the TH emission disturbed by alcohol droplets. Intensity of TH emission is increased about 30 times in magnitude compared with the undisturbed case, and it is more efficient than that using a single copper fiber. The reasons are as follows. On the one hand, the interface[22] between air and alcohol droplet would suppress the destructive interference effect leading to the sudden increase of TH emission. On the other hand, alcohol droplet is transparent media that fundamental pulse and generated TH emission can go through. Nonlinear third-order susceptibility in alcohol droplets is a thousand times higher than that of air. So the increase of nonlinearity would also play an important role in the enhancement of TH emission.[23]

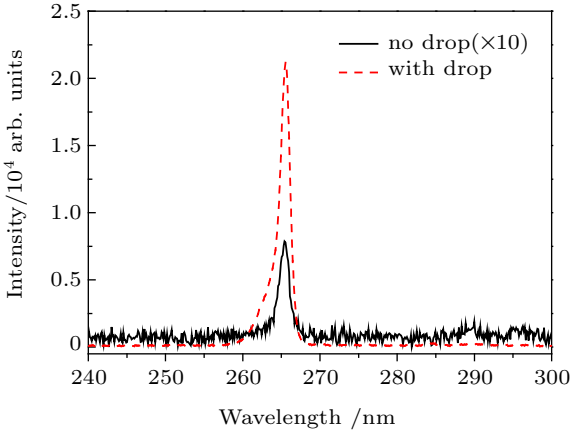


Fig. 3. (color online) Intensity of TH emission disturbed by alcohol droplets (dash line) compared to the undisturbed case (solid line).

Finally, we also try to characterize the role of position and size of the obstacles in enhancement of TH emission, apart from different sorts of obstacles. Figure 4(a) shows the intensity of generated TH emission as a function of longitudinal position of alcohol droplets. In the experiment, the position of the droplet can change in the propagation direction of the filamentation. TH intensity is very sensitive to the position of droplets. When the interaction position is in 207 cm where the filamentation is strongest, enhancement of the TH emission is the most effective. Moving the droplet to both directions along the filamentation, enhancement of the TH emission weakens. So the enhancement mechanism of suppression of the destructive interference is further demonstrated. Enhancement of TH emission with different copper fiber size is also shown in Fig. 4(b). The TH intensity is a little bit higher when a 100-μm fiber is used than that of the 70-μm one, which has the same trend with the simulation results in the reference.[19] Enhancement of TH intensity differs from the size of obstacles, which indicates that optimal size of obstacle may relate to the size of plasma channel. It should be demonstrated systematically in our future works.

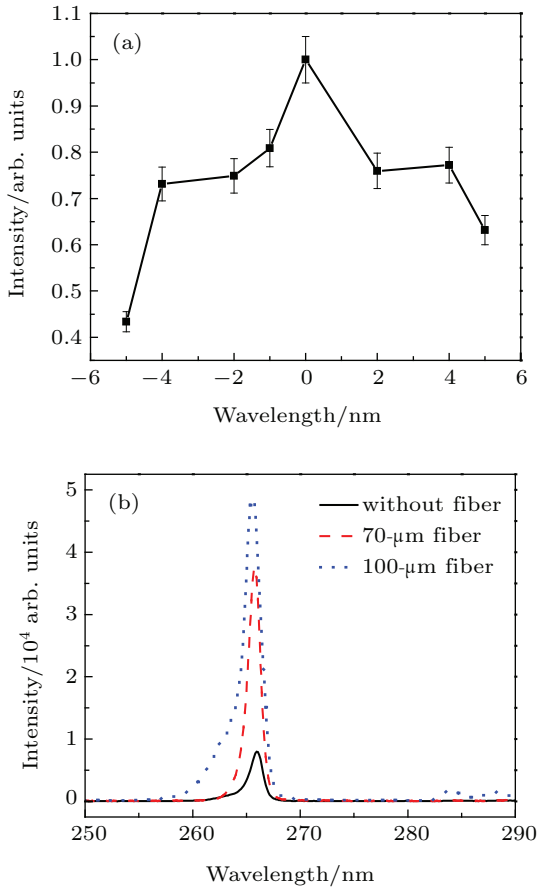


Fig. 4. (color online) (a) The TH intensity as a function of droplet position; (b) the dependencies of TH intensity on the size of obstacles: 70-μm fiber (dash line) and 100-μm fiber (dot line).

4. Conclusion

In conclusion, enhancement of the TH emission in air filamentation has been observed by disturbing filamentation with obstacles experimentally. Enhancement of TH emission up to 5 times in magnitude has been achieved with disturbances of copper fibers while it is about 30 times with alcohol droplets. A comparison of the results demonstrates that the suppression of the destructive interference in filamentation plays a dominant role in enhancement of TH emission. Besides, both the

interface effect and the increase of nonlinearity contribute to the TH enhancement in the case of alcohol droplets, which results in a higher conversion efficiency. Dependence of TH emission on the size and relative position of obstacles is also investigated in the experiment. Further enhancement of TH emission in filamentation can be achieved by optimizing the nonlinear properties of obstacles as well as the size and relative position, which we should study in depth in our future work.

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