

From random to controlled small-scale filamentation in water

H. Schroeder¹, J. Liu^{1,3}, and S. L. Chin²

¹Max-Planck-Institut für Quantenoptik, Laserchemie, Hans-Kopfermann-Str.1, 85748 Garching, Germany

²Center for Optics, Photonics and Laser (COPL), Department of Physics, Engineering Physics and Optics, Laval University, Quebec City, Canada G1K 7P4

³Laboratory for High Intensity Optics, Shanghai Institute of Optics and Fine Mechanics, Shanghai 201800, P. R. of China

hms@mpq.mpg.de

Abstract: Simple apertures such as slits and meshes inserted in the beam path of a powerful Ti:Sapphire laser pulse are suitable to produce stable 1-D and 2-D arrays of filaments in liquids. The thus imposed intensity gradients and diffraction patterns can overcome the inherent beam irregularities which naturally give rise to random small-scale multiple filamentations. This method is visualized by means of two photon fluorescence imaging.

© 2004 Optical Society of America

OCIS codes: (190.5530) Nonlinear optics, pulse propagation and solitons; (190.7110) Ultrafast nonlinear optics.

References and links

1. S. L. Chin, A. Brodeur, S. Petit, O. G. Kosareva, and V. P. Kandidov, "Filamentation and Supercontinuum Generation during the Propagation of Powerful Ultrashort Laser Pulses in Optical Media (White Light Laser)," *J. Nonlinear Opt. Phys. Mat.* **8**, 121-146 (1999).
2. V.P. Kandidov, O. G. Kosareva, I. S. Golubtsov, W. Liu, A. Becker, N. Akozbek, C. M. Bowden, and S. L. Chin, "Self-transformation of a powerful femtosecond laser pulse into a white-light laser pulse in bulk optical media (or supercontinuum generation)," *Appl Phys B* **77**, 149-165 (2003).
3. J. Kasparian, M. Rodriguez, G. Mejean, J. Yu, E. Salmon, H. Wille, R. Bourayou, S. Frey, Y.-B. Andre, A. Mysyrowicz, R. Sauerbrey, J. -P. Wolf, and L. Wöste, "White-light filaments for atmospheric analysis," *Science* **301**, 61-64 (2003).
4. P. Rairoux, H. Schillinger, S. Niedermeier, M. Rodriguez, F. Ronneberger, R. Sauerbrey, B. Stein, D. Waite, C. Wedekind, H. Wille, L. Wöste, and C. Ziener, "Remote sensing of the atmosphere using ultrashort laser pulses," *Appl. Phys. B* **71**, 573-580 (2000).
5. Q. Luo, W. Liu, and S. L. Chin, "Lasing action in air induced by ultra-fast laser filamentation," *Appl. Phys. B* **76**, 337-340 (2003).
6. H. Schroeder and S. L. Chin, "Visualization of the evolution of multiple filaments in methanol," *Opt. Commun.* **234**, 399-406 (2004).
7. S. A. Hosseini, Q. Luo, B. Ferland, W. Liu, S. L. Chin, O. G. Kosareva, N. A. Panov, N. Akozbek, and V. P. Kandidov, "Competition of multiple filaments during the propagation of intense femtosecond laser pulses," *Phys. Rev. A* (to be published).
8. W. Liu, S. A. Hosseini, Q. Luo, B. Ferland, S. L. Chin, O. G. Kosareva, N. A. Panov, and V. P. Kandidov, "Experimental observation and simulations of the self-action of white light laser pulse propagating in air," *New Journal of Physics* **6**, P. 6 (2004).
9. G. Mechain, A. Couairon, Y. -B. Andre, C. D. Amico, M. Franco, B. Prade, S. Tzortzakis, A. Mysyrowicz, and R. Sauerbrey, "Long-range self-channeling of infrared laser pulses in air: a new propagation regime without ionization," *Appl. Phys. B* **79**, 379-382 (2004).
10. S. Minardi, S. Sapone, W. Chinaglia, P. Di Trapani, and A. Berzanskis, "Pixellike parametric generator based on controlled spatial-soliton formation," *Opt. Lett.* **25**, 326-328 (2000).
11. A. Dubietis, G. Tamosauskas, G. Fibich, and B. Ilan, "Multiple filamentation induced by input-beam ellipticity," *Opt. Lett.* **29**, 1126-1128 (2004).
12. G. Méchain, A. Couairon, M. Franco, B. Prade, and A. Mysyrowicz, "Organizing Multiple Femtosecond Filaments in Air," *Phys. Rev. Lett.* **93**, 035003-1, 1-4 (2004).
13. V. P. Kandidov, M. Scalora, O. G. Kosareva, A. V. Nyakk, Q. Luo, S. A. Hosseini, S. L. Chin, "Towards a control of multiple filamentation by spatial regulation of a high-power femtosecond laser pulse," *Appl. Phys. B* submitted, 2004.

14. H. Schroeder, invited talk "2-D regular supercontinuum sources from a mesh," presented at the 2nd International Symposium of Ultrafast Intense Laser Science, Quebec City, Canada, Sept. 25-29, 2003.
 15. W. Koechner, *Solid-State Laser Engineering* (Springer-Verlag Heidelberg New York, 1988).
 16. V. A. Soifer, *Methods for Computer Design of Diffractive Optical Elements* (John Wiley & Sons, Inc., New York, 2002).
 17. S. Minardi, A. Varanavicus, A. Piskarskas, and P. Di Trapani, "A compact multi-pixel parametric light source," *Opt. Commun.* **224**, 301-307 (2003).
-

1. Introduction

An important and timely application of the filamentation of powerful femtosecond laser pulses in air is the remote detection of chemical and biological agents using a single laser. Such powerful femtosecond laser pulses can self-transform into white light laser pulses during filamentation in an optical medium [1,2]. The Teramobile team made use of the back scattered absorption of the white light laser by pollutant molecules in air and demonstrated the potential feasibility of such a remote sensing technique [3,4]. A complementary technique is the remote sensing of the nonlinear gain fluorescence of molecules inside the filament proposed by one of us (SLC) [5]. Other long range atmospheric applications could also be envisioned such as target recognition and lightning control [3], etc.

These potentially important applications rely upon the efficient generation and control of filaments. Note that a small perturbation in the laser pulse front can be amplified efficiently by self-focusing and filamentation [6]. Since pulse front inhomogeneity occurs in almost all laser pulses and since during propagation, the natural inhomogeneity in the optical medium would lead to further perturbations on the pulse front, multiple filamentation starting from such perturbations would naturally occur. That is to say, multiple filamentation is unavoidable normally when the peak power of the input pulse is high. Moreover, such multiple filaments will undergo competition for energy randomly [6-9]. Most of the time, the competing filaments die off before becoming mature [6,7]. A mature filament is one that has undergone the full processes of self-focusing and filamentation during propagation and will end with self-steepening resulting in the strong spectral broadening (white light laser or supercontinuum) [6]. This also means that the yield of strong field ionization, fragmentation and subsequent fluorescence of molecules in the path of the filaments in air would be very weak if the filament is not mature [7].

A practical challenge thus occurs. It calls for a systematic control of multiple filamentation. This means we have to find a way to overcome the natural fluctuations. This is indeed possible by introducing stronger artificial perturbation into the beam path. In Ref. [10] a lithium triborate parametric generator based on controlled spatial-soliton formation was realized by means of a gridlike photo slide placed in the pump-beam path. Noise-induced multiple filamentation in water could be forced into predictable and highly reproducible filamentation patterns by small input beam ellipticity [11]. The possibility to organize regular filamentation patterns in air by imposing either strong field gradients or phase distortions in the input-beam profile has been shown in Ref. [12]. The principle of the control process is being studied numerically for the propagation of a Ti:Sapphire laser pulse in methanol subjected to random light field fluctuations [13].

Here, we demonstrate the persistence of small-scale multiple filamentation when typical Ti: Sapphire laser pulses (2mJ, 50fs, 800nm) propagate in liquids. Then we illustrate the creation of 1-D and 2-D arrays of filaments by launching the beam through appropriate apertures (slits and meshes). In the ideal case multiple filamentation is completely controlled by the superimposed regular diffraction patterns. Furthermore, all filaments evolve nearly identically during the full propagation process as if there were no competition. Similar to Ref. [13,14], we choose a dye solution as the propagation medium to demonstrate the principle since the critical power for self-focusing is much lower (a few MW) than that of air and since we can directly visualize the filamentation process [6,14].

2. Experimental procedure

Figure 1 illustrates the “natural” situation when a 41fs unfocused Ti:Sapphire laser pulse with a peak power of 6.6×10^{10} W/cm² propagates through water (Rhodamine B in water). The bottom self-evident drawing shows the experimental set-up. The overall decrease of the fluorescence intensity along the beam path (top) is mainly caused by the dispersion of the group velocity and the near-infrared absorption of water ($\alpha=0.02\text{cm}^{-1}$). Images of the entrance beam (left pattern) and of the exit beam (right pattern) are also displayed. They represent two photon images from a 1mm thin dye cell. The reddish appearance is caused by the relatively high dye concentration. However, it is still sufficiently low to prevent cross-over effects due to excited dipole-dipole interactions. The indicated diameters refer to the 1/e levels of the two photon images. An obvious advantage of this profile taking technique is due to the fact that the powerful beam does not have to be attenuated (the image signal and the fundamental beam are separated afterwards).

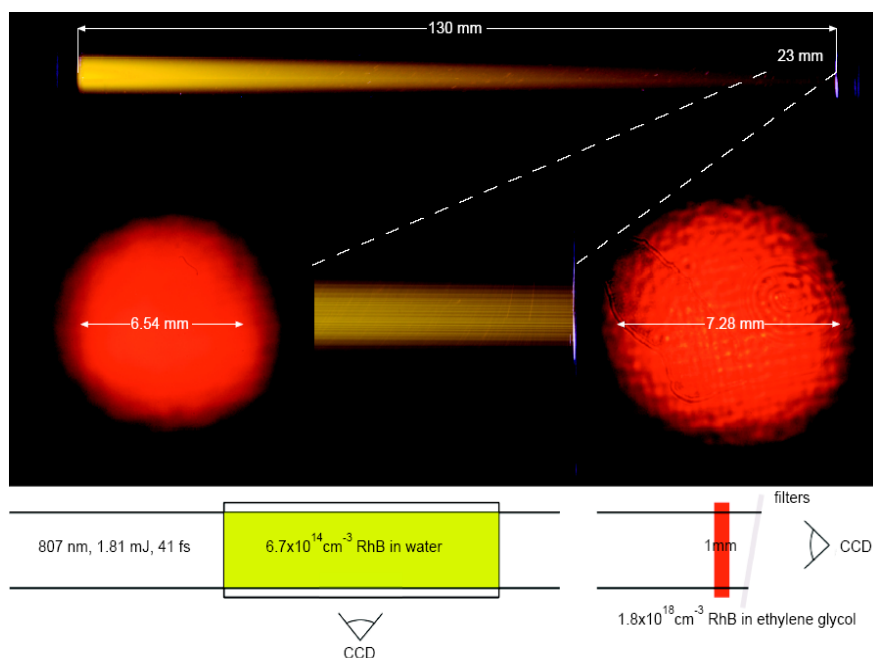


Fig. 1. Two photon fluorescence imaging illustrates small-scale multiple filamentation in water from nearly discernible ripples in the entrance beam (left image). The hot spots in the exit beam (right image) and the enlarged section of the side view demonstrate the appearance of randomly distributed filaments. The individual diameters refer to 1/e levels.

Although the impinging beam seems to be quite smooth and homogeneous, it is not. A closer inspection would reveal small intensity ripples on the percent level (mainly caused by diffraction, and interference from optical imperfections). These ripples appear randomly and uncontrolled in space but temporally stable after each fine adjustment of the laser system. Upon propagation, this initial “noise” is considerably amplified after a propagation length of about 10cm in this case. At this distance the whole beam was transformed into a multitude of filaments. This is shown by the enlargement of the end section of the beam (middle panel) and by the corresponding set of hot spots in the exit beam image. This typical scenario has previously been documented for high power ps laser pulses [15], it is called small-scale self-focusing. The usually expected whole-beam self-focusing (according to the popular B-integral) is not observed.

Figure 2 demonstrates the formation of a 1-D array of essentially parallel non overlapping filaments which are suited for a detailed analysis of the filamentation dynamics. To this end the impinging laser beam was simply clipped by a slit aperture built from razorblades. In order not to obscure the exit image by overlapping white light cones (giving rise to a strong one photon contribution) and to clearly display the diameter and the location of the developing filaments, we displaced the slit position a little apart from the beam center and added some positive chirp. In this way the input power was sufficiently reduced to prevent white light formation along the distance of interest (about 30mm). In the resulting propagation image we can recognize 37 isolated filaments. The initial vertical noise structure is most probably related to mechanical imperfections of the razorblades because we get a comparable entrance beam profile when launching a green homogeneous He-Ne laser beam (Rhodamine B is thus excited by one photon absorption) through the slit. This means in practical terms (inspecting the rather even distribution of the filament spots) that it should be possible to overcome the uncontrolled “natural” multiple filamentations by appropriate stronger disturbers with a predetermined local structure.

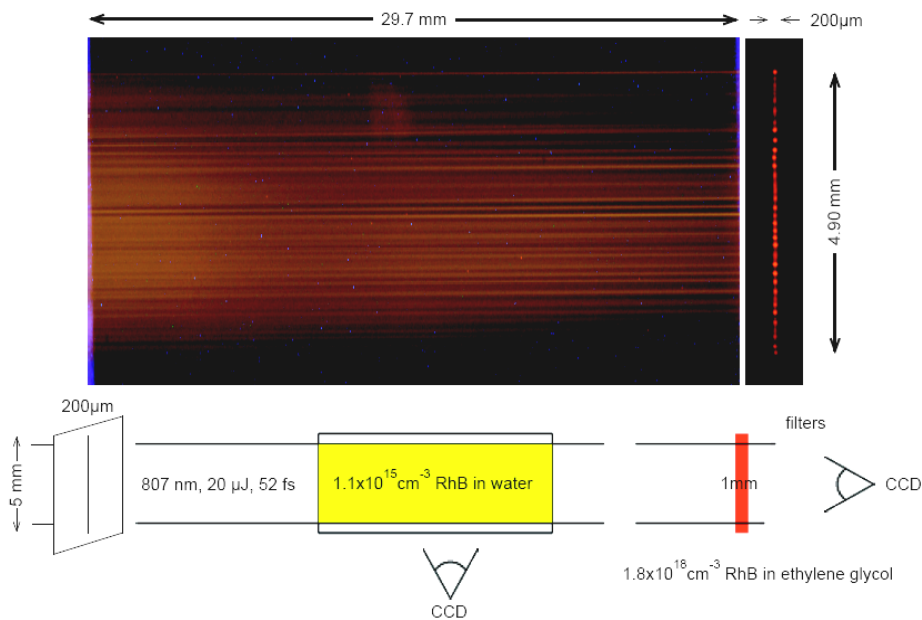


Fig. 2. Creation of a space-fixed 1-D array of filaments by launching the beam through a 200 μm wide slit built from razorblades with small mechanical imperfections. The available input intensity was somewhat reduced to avoid white light production along the propagation length. The image shows a side view of developing filaments and the corresponding spot structure of the exit beam.

A simple realization of this concept is depicted in Fig. 3. We introduce a metallic wire mesh into the beam path (11 \times 11 meshes, unit cell 497 μm \times 497 μm , wire width 54 μm). The wire mesh creates a symmetric diffraction pattern which changes from position to position along the optical axis. This is exemplified in Fig. 3(a) for six representative distances (13mm to 390mm). The images are false color two photon fluorescence images taken close to the optical axis. The size of each section is 1mm \times 1mm, so that we view an entire single mesh and half of the neighboring meshes. Although the generated diffraction patterns appear to be very rich and complicated they can easily be retrieved numerically in terms of Fresnel integrals (this is a practical advantage of rectangular apertures). At a distance of 390mm four bright diffraction

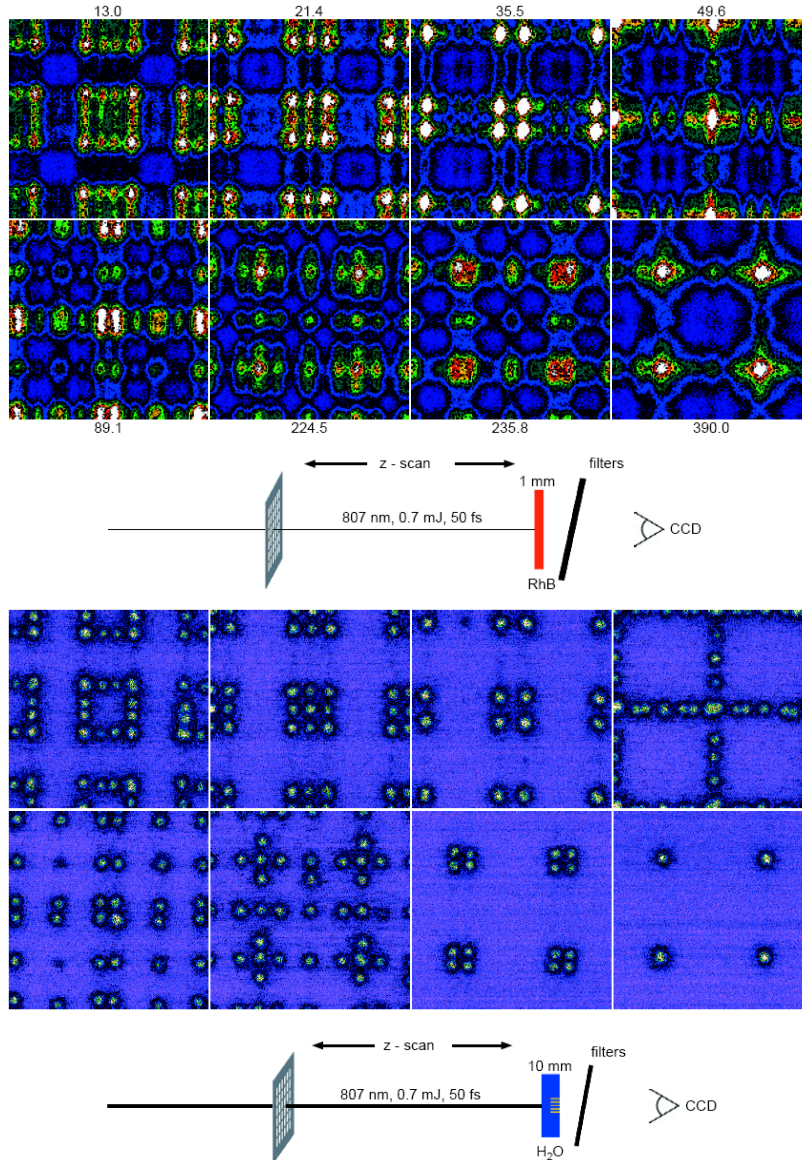


Fig. 3. Creation of space-controlled 2-D arrays of filaments by launching the beam through a wire mesh (11×11 meshes, mesh $497 \mu\text{m}$, wire diameter $54 \mu\text{m}$). (a) Characteristic diffraction patterns for various distances. The shown image sections ($1 \text{mm} \times 1 \text{mm}$) contain the center mesh and half of its neighbors. (b) Corresponding white light patterns when the dye cell from Fig. 3(a) is replaced by a 10mm water cell.

spots are located exactly in the shadow region of the wire crosses, in reminiscence of the famous Spot of Arago or Poisson Spot, respectively. The calculated local intensity enhancement with respect to unity entrance intensity is about 4.5, the maximum factor is close to 7. One can choose any pattern and allow it to propagate in the water cell. Those with sufficiently strong intensity modulations would be amplified into white light sources during the nonlinear propagation. Figure 3(b) shows the propagation effect of the corresponding patterns of Fig. 3(a) through the water cell. The output white light patterns show a one to one correspondence to the input patterns. No random hot spots can be detected; i.e., the control of filamentation is complete at least to the extent of the sensitivity of our experiment. In this

respect the mesh can be regarded as a simple diffractive optical element (DOE) which serves to concentrate the laser beam into quite peculiar contours [16]. Of course, it might also be possible to apply an array of micro lenses for filamentation control [17]. However, in this case the fundamental beam is finally divergent. Whereas, in our case, it remains essentially parallel. This aspect considerably simplifies the analysis of the filamentation process.

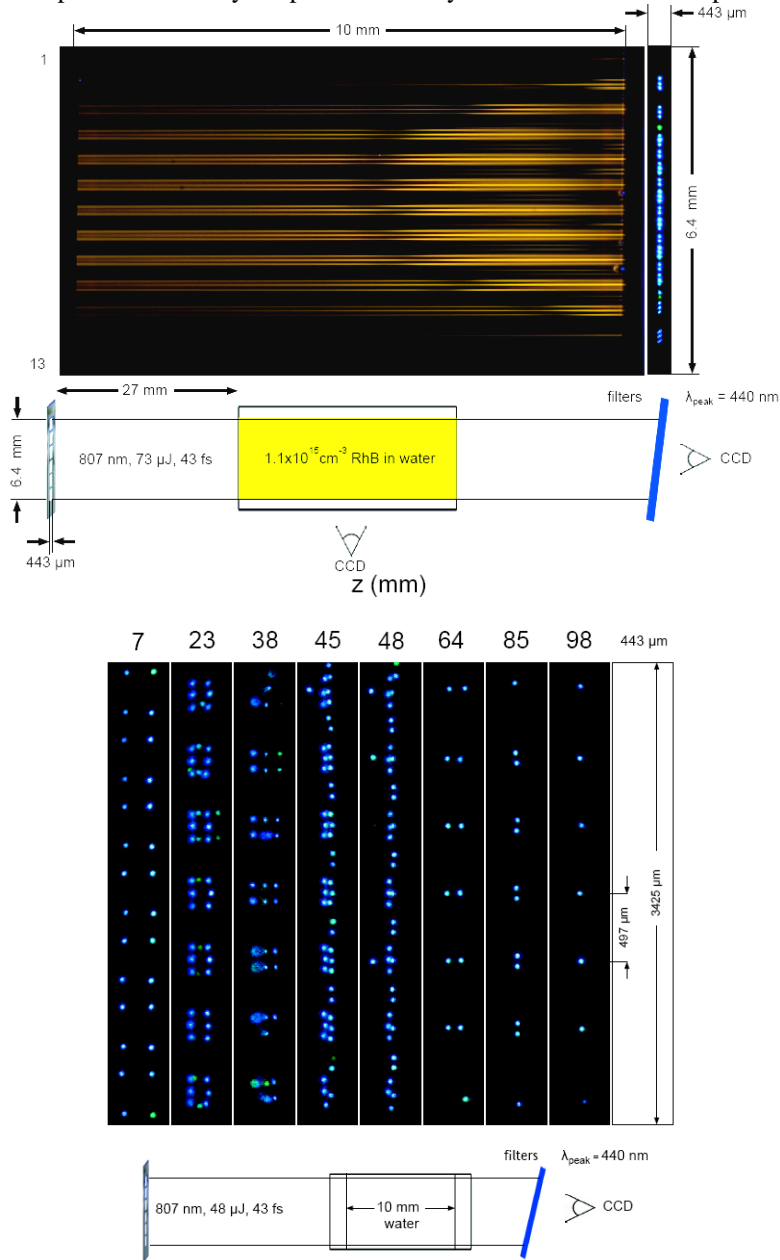


Fig. 4. Creation of space controlled 1-D arrays of filaments by launching the beam through a slit mesh (11 meshes, the same as in Fig. 3). (a) Side view of filamentation and generation of white light (bright zone on the right) and the corresponding exit image for a mesh to cell distance of 27 mm. (b) Characteristic white light patterns from a 7-mesh slit for various mesh positions. The slit size is given in the right panel for direct comparison.

Figure 4 exemplifies a similar control using a 1-D mesh. Figure 4(a) shows the side-view and the end-view of the periodic and regular filamentation for a mesh to cell distance of 27 mm. Figure 4(b) shows that we can choose the propagation to start from a number of different local symmetries by simply changing the mesh position. The right panel shows the overall mesh size (7 meshes) on the same scale. Again, no random hot spots or uncontrolled filaments can be detected within the sensitivity of our experiment. Because of their simple and variable symmetries, these patterns should also be suitable to investigate white light interferences.

Incidentally, such regular distinctive patterns (1-D and 2-D) after nonlinear propagation could be viewed as a systematic way of writing data storage points inside an appropriate data storage medium. Or, more generally, nonlinear propagation might offer a new technique to enhance small contrasts.

3. Conclusions

In conclusion, we have shown that it is possible to totally overcome multiple filamentation due to natural perturbation of the pulse front of a powerful femtosecond laser pulse during nonlinear propagation. The technique we use is to superimpose a regular diffraction structure that could dominate over the random fluctuation. This is possible if the power of the laser inside each element of the regular structure is much higher than the random fluctuations of the field.