Experimental observation and simulations of the self-action of white light laser pulse propagating in air

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Abstract. We present here a recent experiment on long-distance free propagation of powerful ultrafast laser pulses. A large divergence of the beam pattern at the anti-Stokes side was experimentally observed, which contrasts the tiny spots at the Stokes side at long distances, while the pattern at the central laser wavelength was practically unchanged (self-guiding). White light laser self-interference patterns were also recorded and discussed.

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1. Introduction

Filamentation is a universally observed non-linear phenomenon when a powerful femtosecond laser pulse propagates inside optical media. Filamentation in air has stimulated a lot of research interest because of its potential application in atmospheric sensing, such as pollutant detection [1]–[5] and lightning control [6]–[8].

Filamentation is popularly explained by the moving focus model [9, 10]. In this model, the laser pulse is treated as a series of successive thin slices. Different slices will reach their self-foci at different propagation distances according to their powers [11, 12]. This gives the perception of one self-focused beam moving along the propagation axis. On the other hand, as each slice self-focuses, the intensity becomes very high, resulting in the production of plasma at the self-focus. After the laser pulse has disappeared, the series of self-foci leave behind a plasma column. This plasma column is more often referred to as a filament. In air, the filament diameter was measured to be about 100 µm [13].

An important consequence of plasma generation is its defocusing effect. It balances the self-focusing effect and leads to a limited beam diameter as well as limited peak intensity. This is known as intensity clamping [14]–[16]. Due to intensity clamping, the energy concentrated into a small filament region is also limited. Experiments [13, 17, 18] have shown that only a small percentage of the total energy is located inside the filament region; more than 90% of the total energy is distributed in the wide background, whose size is of the order of the input beam diameter. We call this background the ‘reservoir’. Simulations have shown that the laser pulse experiences a significant dynamic energy exchange between the filament region and the (background) reservoir during propagation [19, 20].

Plasma generation, together with the self-steepening of the trailing part of the pulse during the propagation [21], [23]–[25], induces a very large asymmetric spectral broadening towards the blue side. The whole spectrum of the laser pulse could cover a range from the near-ultraviolet to the infrared (IR). As discussed in [23, 26], the broad spectrum of the pulse is popularly called the supercontinuum [27] and the laser pulse turns into a chirped white light laser [23, 26]. Self-generated plasma also partially diffracts the laser pulse. The diffraction pattern is in the form of a conical emission of the visible wavelength of the laser spectra [13, 28, 29]. The shorter wavelengths diffract with larger angles.

It is also well known that when the laser power is much higher than the critical power for self-focusing, multiple filaments will be generated by the intensity perturbation across the wavefront. In atmospheric sensing that usually uses multi-terawatt (TW) laser pulses, the imperfect initial beam quality [30] and refractive index fluctuation caused by turbulence [31] will be the sources of perturbation and will easily induce the formation of multiple filaments. In this case, the propagation of laser pulse may not be simply the generation of a superposition of every individual filament. It has been observed recently that, during propagation, filaments dynamically interfere with each other, thus providing favourable or unfavourable conditions for the development of a long and homogeneous plasma column [32].

In this paper, we report an experiment on long-distance propagation (∼100 m) of 1.3 TW laser pulses from a Ti–sapphire laser in air. In this experiment, multiple filaments were produced. By recording the laser beam patterns at several propagation distances and at different wavelengths, the evolution of the laser beam profile during the filamentation process was studied. We observed that the part of the pulse in the visible (anti-Stokes) side of the spectrum had a much larger divergence than that in the IR (or Stokes) side. At the same time,
we recorded complicated and spectacular interference patterns at the visible wavelengths. We interpret it as a manifestation of self-interference of the white light laser. During the experiment, we also noticed the beam self-guiding at the original wavelength region. We conclude that self-guiding is due to interactive energy exchange between small filament regions and the (background) reservoir during the propagation [18, 19]. As far as we know, this is the first systematic experimental study on the beam pattern evolution of a TW laser pulse over a long propagation distance.

2. Experimental set-up

The experimental set-up is shown in figure 1. In our laser system (built by Spectra Physics), the oscillator (Tsunami, pumped by Millennia) delivers 25 fs pulses into a typical CPA amplifier system (Spitfire). The pulse is stretched and then amplified by a 1 kHz regenerative amplifier. After that, a pulse slicer picks up one pulse from the output of the regenerative amplifier at a repetition rate of 10 Hz. This 10 Hz seed beam is again sent to a 4-pass amplifier, which is pumped by the second harmonics output of two Nd:YAG lasers (GCR-350). Finally, the amplified pulses are compressed to 45 fs (at FWHM, measured by Positive Light single shot autocorrelator). After the compressor, the laser spectrum is centred at 800 nm with a bandwidth of about 30 nm (FWHM). The beam diameter was 2.5 cm at the 1/e² level. To perform the long-distance propagation experiment, the laser beam was made to pass through a 180 m long corridor through a 10 m long pipe. The CaF₂ exit window of the pipe was 1.5 cm thick. To avoid self-focusing inside the pipe before it arrived at the corridor, the compressor and the pipe were pumped down to a pressure of 10⁻³ Torr. The laser energy just after the exit window was fixed at 60 mJ pulse⁻¹, which corresponds to a peak power of 1.3 TW and is almost 500 times the critical power for self-focusing in air (∼3 GW) [25]. At this energy level, the beam profile after the exit window is shown in figure 2(b). Figure 2(a) gives the beam profile immediately after the compressor (i.e. before entering the pipe) for comparison. It was noticed that, after the window, the inhomogeneity of the intensity distribution is...
enhanced inevitably. It was associated with the self-focusing inside the window, since the peak power of our laser pulse was much higher than the critical power for self-focusing in CaF$_2$ ($\sim$7 MW) [18].

The laser pulse was sent from the exit window to the end of the corridor. The propagation distance inside the corridor was about 100 m. During the experiment, all the lights in the corridor were turned off to keep a completely dark experimental environment. To study the laser beam pattern, we prepared an imaging set-up on a small carrier. A white screen (laser-printer-quality paper) was vertically fixed at the edge of the carrier. During the experiment, the carrier was moved along the laser propagation axis with the laser beam located at the centre of the screen. As shown in figure 1, at the two sides of the propagation axis, two cameras were installed in front of the screen at small angles ($\sim$20$^\circ$), with respect to the propagation axis. One of the cameras was a commercial digital camera (Canon A40) and its exposure time was set to be 100 ms. The other was a CCD camera (Cohu 4810), which was triggered by the laser pulse. Therefore both cameras were able to take single shot images of the laser patterns on the screen. While the digital camera was used to take true colour pictures, the CCD camera recorded the laser beam profiles at different wavelengths by putting different band pass filters in front of it. The bandwidth of each filter was 10 nm. In addition, various neutral density filters were used to attenuate the light intensity entering the CCD camera. The two cameras were not synchronized with each other. We carefully checked the pictures obtained by the cameras, and were assured that the aberration effect in our results caused by the small angle between the cameras and screen was negligible.

3. Experimental results

The first position of the white screen was 10 m away from the pipe’s exit window ($d = 10$ m). Figure 3(a) is a typical true colour picture taken by the digital camera. Figure 3(b) demonstrates
Figure 3. Picture taken by digital camera and CCD camera at $d = 10 \text{ m}$: (a) true colour, (b) 800 nm, (c) 700 nm and (d) 950 nm; (a) and (b) indicate that the laser beam had already collapsed into several unstable hot spots, which induced multiple filaments during propagation.

the pictures at 800 nm taken by the CCD camera. The total optical density (OD) of the neutral density filters used before the CCD camera is shown in the picture. As shown in figures 3(a) and (b), at this position we clearly see that the laser beam collapses into several hot spots. It manifests the starting of the formation of multiple filaments. It is due to the effect of the inhomogeneity in the enhanced beam profile after the propagation through the window. The air turbulence in the corridor might also contribute to the wavefront inhomogeneity. The hot spots were not stable. Their intensities fluctuated from shot to shot as well as their spatial distribution. Sometimes, we observed that one or two hot spots were more intense than the others. The spectral broadening was not sufficient in this case. Our CCD camera could detect signals from 700 to 950 nm only. The patterns at 700 and 950 nm are presented in figures 3(c) and (d) respectively. The images taken at different wavelengths—apart from 800 nm—are confined to a few small regions where the stronger hot spots were located. In tune with the fluctuation of the hot spots, the signals in figures 3(c) and (d) also fluctuated.

For wavelengths below 700 nm, we could not record anything with the CCD camera. This suggests that, in our experiment, fluorescence from the paper was not strong enough to be recorded, since printer paper fluorescence was mostly centred at 420 nm with a long tail extending to 600 nm [33]. We also noticed that a few holes (with diameter $<1 \text{ mm}$) could be formed on the paper after a certain time of exposure to the high-intensity laser. Hence, the paper was changed often during the experiment to keep a good screen-surface quality.
Figure 4. Picture taken by digital camera at $d = 20$ m. Laser pulse turned into white light laser (supercontinuum).

When the white screen was moved to a distance $d = 20$ m, white (laser) light (i.e. supercontinuum) appeared. It was recorded by the digital camera (all colours) as shown in figure 4. Similar to the situation at 10 m distance, the intensity distribution among the hot spots was not uniform. At this distance, a few hot spots with higher intensities gave rise to the generation of the white light laser pulse. The white pattern in figure 4 is not due to the saturation of the camera by the scattered 800 nm light because, if this were so, the hot spots in the patterns in figure 3(a) would have given us white patterns already. The images for different wavelengths, which could be recorded by the CCD camera extend to 550 nm in the short wavelength side, as shown in figure 5. For wavelengths longer than 950 nm, we did not have the proper filters and hence we did not study further the longer wavelengths. Those pictures were also not stable, owing to the fluctuation of hot spots seen at the distance of 10 m. In fact, this fluctuation behaviour of hot spots, which also induces a fluctuation in the white light laser, occurs all through the experiment.

An interesting feature observed at $d = 20$ m was that we were able to record the radial fringe pattern surrounding the central spots at 650 nm as shown in figure 5(c). To record these fringe patterns, we had to reduce the number of neutral density filters. Consequently, stronger spots were saturated and appear as white spots in the CCD camera pictures. The unsaturated picture taken at the same wavelength is shown in figure 5(b). For wavelengths below 650 nm, we did not see a similar fringe pattern even when the pictures were overexposed (e.g. figure 5(a) at 550 nm). But at 700 nm (figure 5(d)), we observed more complicated patterns including ring structures and radial fringes. The pattern seen at 700 nm was larger than that at 650 nm. Furthermore, we did not see similar rings and fringes at the Stokes side (wavelength longer than the principal laser wavelength) as shown in figures 5(f) and (g).

Figure 6 presents the true colour pictures taken by the Canon digital camera at $d = 30$ m. Besides the central strong white spot, different coloured rings were formed with different radii. Blue coloured rings were located outside, whereas red coloured rings were located inside. This is the well-known conical emission. Also, in figure 6, we could identify the complex pattern which is a combination of the ring structures and fringes as shown in figure 5(d). Figure 7 gives
detailed spatial information at several wavelengths recorded by the CCD camera at this distance. As seen in figure 7(a), the CCD camera was able to detect the images at wavelengths as short as 400 nm. The complex patterns were observed mostly from 450 to 700 nm (figures 7(b)–(d)). On the other hand, from the CCD pictures in figure 7, we notice that the lights, which are in the visible region from 400 to 700 nm, were no longer restricted to small regions. They had expanded into larger patterns. In contrast, the signals of IR light were still contained in small regions as shown in figures 7(f) and (g). No clear dimensional change was observed at 800 nm, but the pattern is fragmented into many more pieces as depicted in figure 7(e). We could also distinguish some weak structures at the edge of the laser profile in figure 7(e).

At \( d = 40 \text{ m} \), the total beam size became bigger and the complex pattern was more pronounced (figure 8). We observed the complex pattern at all the anti-Stokes wavelengths.

**Figure 5.** Picture taken by CCD camera at different wavelengths \((d = 20 \text{ m})\). Radial fringes and ring structure are observed in (c) and (d).
recorded by the CCD camera (figures 9(a)–(d)). In figure 9, the spectra from 400 to 750 nm diverge much more compared to figure 7, and that shorter wavelengths have larger divergence (see figures 9(a)–(d)). Starting from 600 nm, the light patterns were larger than the CCD camera’s imaging area; thus we could not observe the full size of the beam at those wavelengths (figures 9(a)–(c)). But on the Stokes side (figures 9(f) and (g)), the IR patterns look more like a collection of hot spots and do not diverge much. The central wavelength pattern still remains almost of the same size (figure 9(e)). Similar wavelength-dependent divergence phenomena were observed at longer distances of 60 and 80 m.

True colour pictures taken at $d = 60$ m are shown in figure 10. Figure 11 shows the wavelength dependence of the patterns. Due to the large divergence of the total beam size, the CCD camera could not retrieve enough spatial information at wavelengths as short as 550 nm (figure 11(a)). The rest of the pictures in figure 11 lead to the same conclusion as discussed above.

At the last position $d = 80$ m. The total beam size became too large and it was difficult to record full pictures using both the digital and CCD cameras. For this case, a few select recorded pictures are shown in figures 12 and 13. The complex patterns are more spectacular and complicated. The visible light beam size became larger with increasing distance. Again, we did not observe any significant change in beam size for 800 nm IR light (Stokes side).

4. White light generated by multiple filaments

4.1. Numerical simulations

The phenomenon observed in our experiment was the generation of white light by a pulse with strongly non-unimodal beam profile formed in the course of multiple filamentation. To describe this effect, one has to consider the non-stationary transformation of the radiation in $(x, y, z, t)$ co-ordinates. Assuming that pulse propagation occurs along the $z$-axis with group velocity $v_g$,
Figure 7. Pictures taken by CCD camera at different wavelengths ($d = 30\,\text{m}$). Observable wavelength had extended to 400 nm. Besides the interference patterns in (b)–(d), Stokes side spectra ((f) and (g)) show a much smaller divergence than those on the anti-Stokes side.

The equation for the slowly varying amplitude of the electric field $E(x, y, z, t)$ is given by

$$2ik \left( \frac{\partial E}{\partial z} + \frac{1}{v_g} \frac{\partial E}{\partial t} \right) = \frac{\delta^2}{\delta x^2} E + \frac{\delta^2}{\delta y^2} E + \frac{2k^2}{n_0} (\Delta n_k + \Delta n_p) - i\kappa \alpha E,$$

where the first two terms on the right-hand side of equation (1) describe diffraction. In the third term, we take into account the non-linearity of the medium. Following [30], we represent the Kerr contribution $\Delta n_k$ in the form $\Delta n_k(t) = n_{2\text{eff}}(t)|E(t)|^2$, where

$$n_{2\text{eff}}(t) = \frac{1}{2} n_2 \left\{ 1 + |E(t)|^{-2} \int_{-\infty}^{t} H(t - t')|E(t')|^2 \, dt' \right\}. \quad (2)$$
Figure 8. Picture taken by digital camera at \( d = 40 \) m. Interference patterns, more pronounced compared with figure 4, are observed.

The response function \( H(t) \) was defined in [34]. For the Gaussian pulse with 45 fs duration (FWHM), we used \( n_{2\text{eff}}(0) = 0.57n_2 \) and the effective critical power for self-focusing \( P_{\text{cr eff}} = 11.8 \) GW.

The plasma contribution to the refractive index \( \Delta n_p \) is given by

\[
\Delta n_p = -\frac{\omega_p^2}{2\omega_0^2},
\]

where \( \omega_p = \sqrt{4\pi e^2 N_e/m_e} \) is the plasma frequency and \( \omega_0 \) the laser central frequency corresponding to \( \lambda = 800 \) nm.

The free electron density \( N_e(x, y, z, t) \) depends on the spatial co-ordinates and time according to the kinetic equation

\[
\frac{\partial N_e}{\partial t} = R(|E|^2)(N_0 - N_e),
\]

where \( N_0 \) is the density of neutals consisting of oxygen and nitrogen in air. To calculate the ionization rate \( R(|E|^2) \) in equation (4), we used the model [35] for the ionization of a hydrogen-like atom in the linearly polarized electric field \( E \). Effective charges of molecular oxygen and nitrogen ions are taken from [36] in order to fit the experimental data on ion yields.

The last term on the right-hand side of (1) describes the energy loss due to ionization, where the absorption coefficient \( \alpha = I^{-1}m\hbar\omega(\partial N_e/\partial t) \) \( m \) the order of the multiphoton process, \( I = c|E|^2/8\pi \) the laser pulse intensity and \( k = 2\pi/\lambda \) the wavenumber.

In equation (1), we have neglected the terms associated with self-steepening and material dispersion in air. Indeed, these phenomena mainly affect the temporal shape of the pulse as well as the white light conversion efficiency and, to a lesser extent, they affect the transverse distribution of different wavelengths [25], which we are currently interested in.

Non-stationary simulations of multifilamentation with the actual beam distribution obtained at the laser system output (see figure 2) and quantitative comparison of experimental and numerical results require powerful computational resources. To make a qualitative comparison...
A strong wavelength-dependent divergence is seen. A shorter wavelength has a larger divergence angle. IR light did not show a clear divergence.

of the experimental results using a computer of moderate size, we have modelled the inhomogeneities on the beam profile at the laser system output as the sum of two Gaussian functions. The complex amplitude of the electric field at the beginning of the propagation was given by

\[
E(x, y, z = 0, \tau) = e^{-\tau^2/2\tau^0} E(x, y),
\]

where

\[
E(x, y) = E_1 \exp \left[ - \frac{(x-x_1)^2 + (y-y_0)^2}{2a_1^2} \right] + E_2 \exp \left[ - \frac{(x-x_2)^2 + (y-y_0)^2}{2a_2^2} \right],
\]

Figure 9. Pictures taken by CCD camera at different wavelengths \((d = 40 \text{ m})\). A strong wavelength-dependent divergence is seen. A shorter wavelength has a larger divergence angle. IR light did not show a clear divergence.
Figure 10. Picture taken by digital camera at $d = 60$ m. It clearly shows the interference pattern constructed by the radial fringes and rings.

Figure 11. Pictures taken by CCD camera at different wavelengths ($d = 60$ m). CCD camera could not record patterns at short wavelengths.

\[ \tau = t - z/v_g \] is the retarded time, $E_{1,2}$, $(x_{1,2}, y_0)$ and $a_{1,2}$ are, respectively, the amplitude, position and radius of the first (the second) perturbation. The value of $\tau_0 = 27$ fs corresponds to the pulse duration used in the experiment and is equal to 45 fs FWHM. The peak power of the pulse (5) was calculated as

\[ P_{\text{peak}} = \frac{c}{8\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} |E(x, y)|^2 \, dx \, dy. \] (6)
Figure 12. Picture taken by digital camera at $d = 80$ m. The whole laser beam was larger than the digital camera’s field of view.

Figure 13. A few pictures taken by CCD camera at different wavelengths ($d = 80$ m).

In the simulations, the ratio of the peak power (6) to the critical power for self-focusing $P_{cr eff}$ was set at 11.

In the initial pulse distribution (5), the radius and intensity of the larger perturbation were $a_1 = a_0 = 0.18$ mm and $I_1 = c|E_1|^2/8\pi \approx 10^{13}$ W cm$^{-2}$, and the same values for the smaller perturbation were $a_2 = 0.045$ mm and $I_2 = c|E_2|^2/8\pi \approx 10^{13}$ W cm$^{-2}$. The relative location of the two perturbations was $x_1 = 3.25a_0$, $x_2 = 4.75a_0$ and $y_0 = 4a_0$. The intensity $I(x, y)$ corresponding to the electric field given by equation (5) is shown in figure 14. The propagation distance $z$ is scaled by the diffraction length of the first perturbation $z_d = ka_0^2$. 

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Discussion of the multiple filament development from a pulse with two initial perturbations is presented elsewhere [32]. In this paper, we concentrate on the spatial distribution of the supercontinuum produced by multiple filaments. The spectral intensity $S(x, y, z, \lambda)$ at the selected wavelength $\lambda$ is given by the equation

$$S(x, y, z, \lambda) = \left| \int_{-\infty}^{\infty} E(x, y, z, \tau) e^{-i\Delta_{\text{tot}} \tau} d\tau \right|^2. \quad (7)$$

Figures 15–17 show the evolution of the spectral intensity $S(x, y)$ with the propagation distance and wavelength. In each figure, there are four plots corresponding to the blue side of the supercontinuum, namely for $\lambda = 600, 650, 700$ and 750 nm, one plot for the laser wavelength of $\lambda = 800$ nm and two plots for the IR part of the spectrum, $\lambda = 850$ and 900 nm. Similar intensity scales are observed in all the plots on the blue side of the spectrum (for $\lambda = 600–750$ nm, maximum intensity $= 1.75 \times 10^{-2} S_{\text{max}}$) and on the red side of the spectrum (for $\lambda = 850$ and 900 nm, maximum intensity $= 1.75 \times 10^{-1} S_{\text{max}}$). Here $S_{\text{max}}$ is the maximum intensity at the laser central wavelength.

The first series of spatial distributions $S(x, y, z, \lambda)$ is shown at $z = 0.10 z_d$ (figure 15) soon after the filament formation. In agreement with the experimental results registered at $d = 20$ m, the simulated supercontinuum signal is rather weak at 600 and 650 nm, but increases and reveals rings at 700 and 750 nm. Initially, rings appear around the filament formed earlier in the propagation from the smaller perturbation at $x_2 = 4.75 a_0$. The long-wavelength components appear and remain at the locations of filaments, without showing any divergence.

With increase in propagation distance, more filaments are formed (indicated by the intensity at $\lambda = 800$ nm in figure 16) and their ring structures interfere with each other ($\lambda = 650–750$ nm at $z = 0.11 z_d$ in figure 16). Comparison of the spectral intensity at $\lambda = 700$ nm observed in the experiment at $z = 0.048 z_d = 30$ m (figure 7(d)) and obtained in the simulations at $z = 0.11 z_d$ (figure 16) reveals a more pronounced undisturbed ring structure on one side and interference.
Figure 15. Simulated spatial distribution of the supercontinuum at different wavelengths. Propagation distance \( z = 0.10 z_d = 0.10 k a_0^2 \), where \( a_0 = 0.18 \) mm. The spectral intensity is normalized to the maximum intensity \( S_{\text{max}} \) obtained at the laser central wavelength of 800 nm. There are three different spectral intensity scales on the plot: the first one for \( \lambda = 600–750 \) nm, the second one for \( \lambda = 800 \) nm and the third one for \( \lambda = 850 \) and 900 nm.

strips on the other side. In the experiment, less disturbed rings were on the left-hand side and the simulations were on the right-hand side. On the IR side, new maxima at 850 nm indicate arising of new filaments (in figure 16, compare the plots for \( \lambda = 800 \) and 850 nm).

Further development of multifilamentation (\( \lambda = 800 \) nm in figure 17) was accompanied by a diverging interference pattern (see intensity distributions at \( \lambda = 650–750 \) nm, \( z = 0.15 z_d \) in figure 17), which simultaneously became more complicated due to many sources of rings arising at different positions in the propagation. For \( \lambda = 750 \) nm in figure 17 we have saturated several
maxima in order to preserve the overall intensity scale for short-wavelength components and to show the expansion of the interference pattern. On the IR side of the spectrum, the divergence was not observed. The increasing number of spectral intensity peaks at both 850 and 900 nm in figure 17 demonstrates the appearance of new filaments.

General features observed both in the experiment and simulations are the increase in conversion efficiency with propagation distance extending to the supercontinuum, divergence of ring structures produced by each filament with distance, development of the complicated interference pattern in the visible range with increasing number of filaments, and confined propagation of long-wavelength components.

Figure 16. Same as in figure 12 for \( z = 0.11z_d = 0.11ka_0^2 \), where \( a_0 = 0.18 \) mm. The spectral intensity scales are similar to those plotted in figure 12 to show the increase in conversion efficiency with propagation distance up to the supercontinuum.
Figure 17. The same as in figures 12 and 13 for $z = 0.15zd = 0.15ka_0^2$, where $a_0 = 0.18$ mm. The spectral intensity scales are similar to those plotted in figures 12 and 13 which show the increase of the conversion efficiency with propagation distance up to the supercontinuum.

4.2. Divergence of anti-Stokes spectral components versus confined propagation of the radiation on the Stokes side of the spectrum

The simulated map of supercontinuum generation in a filament [37] shows that the sources of anti-Stokes spectral components are located both on the filament axis and in the surrounding rings, while the maximum of the Stokes wavelengths is on the beam axis. The spatial distribution of anti-Stokes and Stokes wavelengths for each particular filament out of many is in agreement with the results for a single filament. The blue spectral components are generated both on the filament axis and in the rings (see figures 5, 7, 9, 11, 13 and 15–17). Due to the divergence of
short-wavelength components, the radius of a spatial ring at a certain wavelength increases with propagation distance. Sources of the long-wavelength spectral components are located in front of the pulse on the axis of each of the filaments and do not diverge with propagation.

Qualitative interpretation of the spatial divergence in the anti-Stokes and Stokes parts of the spectrum can be made on the basis of the non-linear phase growth $\phi_{nl}$ in the course of propagation (see e.g. [38]). The phase growth $\phi_{nl}$ along the direction $z$, where the intensity and electron density are assumed to be independent of $z$, is given by

$$\phi_{nl}(x, y, z, \tau) = -kz(\Delta n_k + \Delta n_p) = -kz\left(n_{2eff}|E(x, y, \tau)|^2 - \frac{2\pi e^2 N_e(x, y, \tau)}{m\omega^2}\right).$$  (8)

In the leading part of the pulse or the subpulses, into which the initial pulse is split, the major contribution to the non-linear phase comes from the Kerr non-linearity, since the plasma needs time to be created. The frequency shift $\Delta \omega = \frac{\partial \phi_{nl}}{\partial \tau} = -kzn_{2eff}\frac{\partial |E|^2}{\partial \tau} \sim -\frac{\partial I}{\partial \tau} < 0$
in the growing front of the pulse is negative, while the radial derivative

$$\frac{\partial \phi_{nl}}{\partial r} = -kzn_{2eff}\frac{\partial |E|^2}{\partial r} > 0$$
is positive due to undisturbed spatial profile with the on-axis intensity maximum. (Here $r = \sqrt{x^2 + y^2}$ and $r = 0$ correspond to the centre of the filament.) Therefore, the radiation in the front of the pulse is frequency-shifted towards the IR and converges towards the filament axis. The result of this is confined propagation of the Stokes components of the supercontinuum.

Later in the pulse, the second term on the right-hand side of equation (8) starts to contribute to the non-linear phase growth. The corresponding frequency shift

$$\Delta \omega = \frac{\partial \phi_{nl}}{\partial \tau} = \frac{kz(2\pi e^2/m\omega^2)\partial N_e}{\partial \tau} > 0$$
is positive, while the radial phase derivative

$$\frac{\partial \phi_{nl}}{\partial r} = \frac{kz(2\pi e^2/m\omega^2)\partial N_e}{\partial r} < 0$$
is negative due to the rapid decrease of electron density in the off-axis direction. As a result, the anti-Stokes components of the supercontinuum experience a non-linear (plasma-induced) divergence.

4.3. Self-interference of white light laser

Another interesting feature observed in our experiments and simulations, is the beautiful white light laser patterns consisting of radial fringes and ring structures in the visible region. White light laser interference is the source of this spectacular phenomenon. We had mentioned in section 2 that our laser pulse broke up into several hot spots after passing through the window at the end of
the vacuum pipe. Each of the hot spots became an individual white light laser source and multiple filaments were induced. These white light sources (filaments) were coherent. Thus, interference among these filaments was inevitable. Since this kind of interference happens inside the laser pulse itself, we call it self-interference.

Inter-filament interference was first mentioned in [28]. Our previous experiment had demonstrated an interference pattern for two filaments at 800 nm [30]. In [30], the mechanism is proposed as an interference between the background laser light and the diffracted laser light from the small self-focusing region. As mentioned in section 1, most of the laser energy was outside the small self-focusing region [23]. This was also confirmed by our results (see figures 5(e), 7(e), 9(e), 11(c), 13(b) and 15–17). This part of the energy forms the background laser light (reservoir). The background laser light propagates more like a plane wave. The laser light diffracted by the plasma from the self-focal region, has a positive wavefront curvature. When the plane wavefront overlaps with the wavefront having a certain curvature, interference gives rise to the ring structure.

The rings surrounding the filament contain not only the radiation at the central laser wavelength but also the anti-Stokes components of the pulse frequency spectrum. This can be directly seen from the spatio-temporal maps of supercontinuum generation [38]. Thus, each filament has its own system of conical emission rings. When the separation distance between the filaments is large enough, the system of rings at the blue-shifted wavelength can be easily seen in the simulations. In figures 15 and 16, for the wavelengths $\lambda = 650, 700$ and 750 nm, the filament formed from the perturbation at $x_2 = 4.75a_0$ exhibits a system of nearly undisturbed rings at these wavelengths. Other filaments were formed later in the propagation and their system of rings were immediately disturbed by the first filament’s system and cannot be distinguished so clearly, on the scale used in the present study. For the same reason, independent system of anti-Stokes rings cannot be easily obtained in the experiment. Due to the large input peak power and hot spots arising in the glass window, many filaments were developed simultaneously in the propagation and overlap with each other’s rings. (Some indication of partially undisturbed ring structures can be seen on the upper-left side of the experimental results in figures 7(b) and (d).) Due to the coherence of the white light coming from multiple filaments, the overlap of the conical emission rings produced by each filament presents the interference of these rings.

4.4. Self-guiding of the laser pulse at the central wavelength

Besides the spectacular self-interference patterns of the white light laser, the self-guiding behaviour of the initial laser wavelength (i.e. 800 nm) was considerable. It means that at the initial laser wavelength, the laser beam diameter varies only inside a limited region during the propagation. We plot the beam diameter as a function of the propagation distance in figure 19. Since the beam profile after $d = 10$ m was no longer uniform, we defined the diameter as the position where the laser beam could be distinguished from the background. It is illustrated in figure 18, where the transverse beam profile at 80 m is shown. Strong modulation is evident. The inset of figure 18 gives the beam profile at $d = 10$ m as another example. This definition of the diameter is reasonable because our corridor was in complete darkness; hence the background superimposed on the laser light should not change with distance. The results in figure 19 demonstrate a small beam-size variation at different positions. It is different from the monotonic increase in beam diameter in the linear propagation case. To compare with the propagation of the long-pulse laser beam, the uncompressed laser pulse (200 ps) was sent through the corridor. An
Figure 18. Definition of beam diameter at 800 nm using beam profile at \( d = 80 \) m as an example. Due to the strong irregularity of the beam profile, the diameter is defined at the position where it is distinguished from background. Inset: beam profile at \( d = 10 \) m.

Figure 19. Beam diameter at 800 nm as a function of propagation distance. Overall divergence is much smaller than the linear propagation (open triangle).

obvious beam divergence at the end of the corridor \( (d = 100 \) m) was observed, shown by open triangles in figure 19. The beam size was almost three times larger than the initial beam size. It is in strong contrast with the non-linear propagation case.

This beam self-guiding behaviour during filamentation could be explained by the energy exchange between the filament regions and the surrounding background (reservoir) [19, 20].
The energy exchange process is first initiated by self-focusing. Self-focusing concentrates energy into a small hot spot. Due to ionization, the energy quickly stops flowing into the hot spot from the surrounding region. The collapse of the beam stops, i.e. the plasma defocuses the laser pulse and the laser energy is pushed out from the hot spot (diverging). The same process occurs for multiple hot spots (filaments). This dynamical process of the energy interchange between the hot spots and the background light (reservoir) repeats during the laser pulse propagation until there is enough power for self-focusing. Consequently, laser energy is devoid of dispersion spreading out radially through natural linear diffraction. Rather, the exchange of energy between the self-foci and the surrounding tends to keep the beam diameter within a limited value, i.e. self-guiding.

5. Conclusions

In summary, white laser light was generated and multiple filaments were developed with 45 fs, 60 mJ laser pulses over 100 m. At different positions, laser beam profiles were recorded by a commercial digital camera and a CCD camera. Wavelength-dependent divergence was observed experimentally and obtained in numerical simulations. In the visible range, each filament produced its own system of conical emission rings, the radii of which increase with propagation distance. The rings, produced by different filaments at the same wavelength, interfere with each other. The interference patterns consist of ring structures and radial fringes. We refer to this unique interference as ‘self-interference’ of the laser pulse.

The IR components of the pulse spectrum were generated due to Kerr non-linearity of air and converge towards the filament axis. The number of maxima found in both the experiment and simulations on the transverse distribution of the red-shifted spectral components corresponds to the number of filaments at the same propagation distance. Besides, it was noticed that the initial 800 nm laser beam propagates with an almost constant diameter, i.e. self-guiding. This is due to the dynamical interaction of self-focusing and plasma defocusing effects that keep the pulse energy within a limited diameter.

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