White-light generation control with crossing beams of femtosecond laser pulses

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Abstract: We investigated the variations in generated white-light when crossing two femtosecond laser beams in a Kerr medium. By changing the relative delay of two interacting intense femtosecond laser pulses, we show that white-light generation can be enhanced or suppressed. With a decrease of the relative delay an enhancement of the white-light output was observed, which at even smaller delays was reverted to a suppression of white-light generation. Under chosen conditions, the level of suppression resulted in a white-light output lower than the initial level corresponding to large delays, when the pulses do not overlap in time. The enhancement of the white-light generation takes place in the pulse that is lagging. We found that the effect of the interaction of the beams depends on their relative orientation of polarization and increases when the polarizations are changed from perpendicular to parallel. The observed effects are explained by noting that at intermediate delays, the perturbations introduced in the path of the lagging beam lead to a shortening of the length of filament formation and enhancement of the white-light generation, whereas at small delays the stronger interaction and mutual rescattering reduces the intensity in the central part of the beams, suppressing filamentation and white-light generation.

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References and Links

1. Introduction

The controlling of light by light is a fascinating possibility. Several physical mechanisms were previously considered, including nonlinear optical interactions [1,2], laser-induced Kerr effect [3–7] and birefringence [8]. Such non-destructive and reversible mechanisms of the interaction of laser pulses form the basis for development of ultrafast optical gating techniques [9–11], including shutters and pulse pickers. The advent of femtosecond laser science has brought such advantages as high intensities and extremely fast optically-controlled switching times, which are orders of magnitude faster compared to their electronic counterparts. An intense pulse propagating in a Kerr medium can experience self-focusing [12–14] with modification of its spectrum due to nonlinear optical processes [15–18]. The increasing utilization of femtosecond laser pulses renewed interest in self-focusing phenomena [19], development of beam instabilities [20] and small-scale self-focusing [21], related to filamentation [22–26], white-light generation [22,27–33] and conical emission [34]. White-light generation is usually accounted for by self-phase modulation [17,35], however stimulated Raman scattering [36,37], parametric four-wave mixing [38–41], and cascading of these effects as the beam propagates can also contribute to the generation of a supercontinuum. The question we are interested in is how the interaction of two crossing beams affects the white-light generation process.

It was shown experimentally that two beams interacting in a gas [42,43] or in a liquid [44] can exchange their energy and mutually transform their spectra. In isotropic solid materials, such as glass, the nonlinear refractive coefficient is typically much larger than in a gas, and therefore the above mentioned optical phenomena, provided that similar laser parameters are maintained, develop on shorter distances. The interaction of two intense beams affects not only the initial beams themselves, but also the resulting spectral transformation. However, the effects of two-beam interaction in white-light generation were not investigated.

In this paper we show that depending on the relative delay between two interacting intense femtosecond laser pulses, white-light generation can be enhanced or suppressed. The output of white-light generation closely correlates with filament formation, which in turn depends on energetic characteristics of the pump beams.
2. Experimental setup

In the beam crossing setup shown in Fig. 1, we used amplified femtosecond laser pulses (Spitfire, Spectra Physics) with a pulse duration of 57 fs and maximum pulse energy of 1 mJ at a 1 kHz repetition rate. An input aperture, placed prior to the depicted setup, was used for adjusting the power of the beams; a typical aperture diameter of 2.7 mm was used for beams producing filamentation. A curved mirror with a focal length of 2.5 m was utilized to focus the radiation into the interaction region. A 50/50 beam splitter served to split the radiation into two arms. Each beam propagated with approximately equal optical path length. The optical path for one of the arms could be varied relative to the other arm by a computer-controlled motorized translation stage (GTS150, ESP301, Newport), providing a temporal resolution of 0.66 fs. The sample was made of flint glass (refractive index \( n = 1.81 \) and density 3.49 g/cm\(^3\)) and had a parallelepipedal shape with dimensions 32.27 mm \( \times \) 15.84 mm \( \times \) 12.60 mm. Immediately before the sample, the power of each of the beams was measured to have a typical average power of about 100 mW. These beams were focused and propagated in air at a relative angle of \( \theta_{\text{air}} = 2.7 \) deg, which corresponded to \( \theta = 2 \arcsin[\sin(\theta_{\text{air}} / 2) / n] = 1.48 \) deg, the crossing angle in the sample (further in the paper we always refer to the latter). In a series of angular-dependent experiments, the crossing angle in the sample was varied from 0.7 to 3.6 degrees. The beams crossed close to the entrance face of the sample to avoid significant dispersion of the pulses. The IR radiation around 800 nm and the white-light were separated by a dielectric mirror, effectively transmitting light from 400 to 700 nm and reflecting light at around 800 nm (see the graph in Fig. 1). The power of the IR (800 nm) radiation and that of white-light were measured (PD300-IR, Orphir) after the glass sample for radiation in the movable and fixed arms as a function of the relative delay. Alternatively, IR radiation and white light were imaged by projecting them onto a CCD camera. The spectra were recorded by an Ocean Optics (USB-2000) spectrometer using an integrating sphere. To assess the maximal effect of the dispersion, we measured (GRENOUILLE, Mesa Photonics) the pulse duration after passing the sample to be 84 fs.

3. Results

The power of the generated white-light closely correlated with the number of filaments; therefore, a series of pictures were taken of the cross-section of the beams with the formed filaments at equal time intervals of 6.6 fs in delay between the two arms (Fig. 2). In each
image, the upper light spot shows filaments in the movable arm and the lower spot corresponds to the fixed arm. The right panel shows a magnified view of the selected area, so that separate filaments can be distinguished and counted. From the images, we observed that the number of filaments varies in both arms, when the relative delay was changed. The number of filaments in the movable arm increased at negative delays and then dipped near zero delays. We define the delay time as the arrival time of the pulse in the fixed arm minus the arrival time of the pulse in the movable arm.

The number of filaments in the fixed arm qualitatively showed the reverse behavior with respect to the delay time when compared to the movable arm. The power output of white-light was measured in each of the arms by alternatively blocking half of the output plane; the total amount of white-light output produced in both arms was also measured. When the delay was gradually changed, the amount of generated white-light experienced variations in both arms (Fig. 3).
The power measured in the movable arm peaked at negative delays then dipped near zero delays level and then returned to the initial level at positive delays. The white-light output in the fixed arm qualitatively showed a reverse behavior with respect to the delay time as compared to the movable arm. Consequently, the amount of white light production increased for the pulse that was lagging, and the total output of white-light of both arms exhibited two well-pronounced peaks with a deep valley in between them. Around zero delay, the white-light level decreased to a level that was below the level found for large delays, when pulses do not overlap.

A more detailed view of the white-light generation as a function of delay was obtained by measuring the spectrum with the Ocean Optics spectrometer. To do this, the output beams were sent into an integrating cavity through a small opening, and the tip of an optical fiber was inserted into the cavity wall to collect light that was then analyzed. The measured spectrum (see Fig. 4) in the spectral interval 400-700 nm, which included the major part of the white-light spectrum and was limited not to include the portion with saturation due to radiation of pump beams, and the result of the integration in this spectral interval (presented in the lower graph) show a well pronounced suppression of the white light spectrum in the vicinity of the zero delay.

To better understand the correlation of the interaction of the beams with the generated white-light, we imaged beam cross-sections and measured the power of the transmitted IR radiation through the sample at different time delays, which revealed a
Fig. 5. Variations of the IR beams during the interaction: left top panel shows the dependence of the beam power vs. delay in fixed (red), movable (green) and in a larger area around movable arm (blue); the curves shifted up for better viewing; the power was measured in the respective circled areas (right top panel) showing the cross sections of beams on the CCD camera in the movable arm (top) and in the fixed arm (bottom) at zero delay, where arc-shaped wings of the intensity distribution are clearly visible. The bottom panel shows images of the beam cross sections taken at: large delays (−300fs) and 300fs (1 and 5), delays corresponding to maximal white-light output (−100fs) and +100fs (2, 4) and zero delay (3). The starting points of arrows indicate respective delays on the time scale.

Fig. 6. The dependence of the IR power on delay and images of the IR beams with the central parts blocked taken at: large delays (1, 5), at delays corresponding to maximal white-light output (2, 4) and zero delay (3). The starting points of arrows indicate respective delays on the time scale.

redistribution of the intensity in the cross-sections of the beams responsible for the reduction in power of the central parts of the beams near zero delays (Fig. 5). When only the power of the central parts (area within circles) of the two beams were measured by inserting an aperture, the power of both beams exhibited a dip near zero delays (green and red traces in Fig. 5). When the power in a larger area (shown for the movable arm by a blue rectangle in
top right panel of Fig. 5), covering the whole output of one beam was measured, the output was almost constant (blue trace in Fig. 5). This means that some of the energy in the beams was deflected to the peripheral areas. In fact, a well pronounced arc-shape extending from the central part of the beam was observed. Additional measurements of the total IR output of the two beams with the central spots of the beams blocked (Fig. 6) indeed showed a maximum in the power distributed in the peripheral area at close to zero delays. The interaction of the beams in the medium was found to depend on the mutual orientation of their polarizations. A decrease of the power in the central regions of the beams is the largest for parallel polarizations and reduces when the angle between polarizations increases to 90 deg (Fig. 7).

![Fig. 7. Variations of the IR radiation in the central spot of the fixed arm for different angles between polarizations of the beams in the two arms. The plots are shifted vertically for better viewing.](image)

![Fig. 8. Results on white-light suppression at different angles: (a). The total amount of white light of both beams measured vs. delay at the crossing angle of 1.1 deg is shown. The quantities used in Eq. (1) are also shown. (b). The dependence of the relative suppression, \( \xi \), of the white-light generation on the angle. The solid line connecting data points is a guide for the eye.](image)

To further understand the nonlinear interaction of the beams in the medium, we investigated the dependence of the observed variations in the white-light output as a function of crossing angle. To perform these measurements, the setup was somewhat modified; the
The main difference was the inclusion of a lens with a focal length of about 23 cm placed downstream of mirrors M3 and M6. For these measurements, the typical aperture size was about 1.7 mm, and the typical power per beam was about 8 mW. This series of measurements was intended to quantify the angular dependence of the suppression of the white light generation at delays close to zero. The total amount of white-light from both beams was measured. The effect of suppression was quantified by the relative suppression of the white-light output power,

$$\xi = \left( \frac{P_0 - P_{\text{min}}}{P_0} \right),$$

where \(P_0\) is the power of the white-light measured at large delays, and \(P_{\text{min}}\) is the power of the white-light at its minimum, observed at close to zero delays as is illustrated in Fig. 8(a). The dependence of the relative change of the white-light output power on the crossing angle is presented in Fig. 8(b).

Figure 8(b) shows significant oscillations of the white-light suppression effect with the crossing angle. Below we provide some arguments that these oscillations can be related to the interference pattern that is changing with the crossing angle. The interference of the beams in the interaction region was numerically simulated in the Fresnel approximation for zero delay between the pulses, which corresponds to the observation of the strongest suppression effect of white-light generation. Using the fact that the crossing angle is small and integrating over the bandwidth of the laser pulse we obtained the following expression for the intensity distribution of the two focused and crossing beams:

$$\frac{I(x, y)}{I_0} = |c\tau(2\pi^{1/2}z)^{-1/2} \int dk (k - k_0)^2 c^2 \tau^2 / 4 | \int dt' \int dx' \exp[i(k((x-x')^2 + (y-y')^2)/(2z))] \times$$

$$\times \exp[-(x'^2 + (r - y')^2)(1/a^2 - ik/\sqrt{2f})] + ik((r - y') \sin(\theta/2) + z \cos(\theta/2)) +$$

$$\exp[\frac{(r - y') \sin(\theta/2) + z \cos(\theta/2)}{1/a^2 - ik/\sqrt{2f}}] + ik((r - y') \sin(\theta/2) + z \cos(\theta/2))] |^2,$$

where \(I_0\) is the initial intensity of the beams (far from the focal region), \(\tau\) and \(a\) are the pulse duration and the beam radius in the initial spatio-temporal distribution of the intensity, which is assumed to be Gaussian, \(c\) is the speed of light, \(f\) is the focal length, \(\theta\) is the crossing angle, \(k_0 = 2\pi / \lambda\) and \(\lambda\) is the light carrier wavelength. It is assumed that the beams are initially separated by the distance \(2r = 2f \sin(\theta/2)\) and propagate from left to right symmetrically along \(z\)-axis in the \(yz\)-plane. Figure 9 shows in false colors the results of the calculation of the intensity distribution with the parameters close to experimental values: \(\tau = 57\ \text{fs}, \ a = 0.85\ \text{mm}, \ f = 23\ \text{cm}, \ \lambda = 800\ \text{nm}\). In the interaction region strong intensity variations can be seen that should also affect nonlinear processes, such as self-focusing and filamentation. With the increase of the crossing angle the interaction region along \(z\)-axis shrinks, and the spatial scale of the intensity variations reduces, while the number of

Fig. 9. The calculated intensity distribution of two crossing beams in the \(yz\)-plane for crossing angles \(\theta = 0.66^\circ\) (a), \(\theta = 0.99^\circ\) (b) and \(\theta = 1.27^\circ\) (c).
oscillations increases. The simulation in Fig. 9 for three angles corresponding to the initial portion of the dependence of Fig. 8(b) with the strong variations of the white-light suppression show significant changes in the intensity distribution with the crossing angle. These changes should have stronger influence for just few interference maxima and minima appearing for these small angles. Thus, the changes in the interference pattern can be expected to modulate the nonlinear interaction of the beams and consequently also affect the white-light suppression effect.

4. Discussion

We observed two main effects in the nonlinear coupling between the two beams: (1) white-light generation can be increased or decreased by varying the relative delay of two interacting pulses and depends on the crossing angle; for parallel polarizations of the beams and crossing angles $\leq 3.6$ deg the contrast of this process is significant enough that it can be described as “switching” the white-light on and off; (2) changes in the intensity distributions of the beams associated with the appearance of arc-shaped lateral extensions, especially pronounced at small delays, were observed.

We will begin our discussion with the second effect, to which mainly two processes contribute. Firstly, it is known that in the diffraction of a beam on a transparent dielectric cylinder, a series of arcs centered around the cylinder appears in the forward direction due to reflection from the cylinder’s surface as well as refraction and transmission [45–47]. In the latter case, the cylinder works as a one-dimensional lens. For a cylinder of small size, diffraction from the cylinder plays a noticeable role. In our experiments (Fig. 5 and Fig. 6) we see arc-shaped formations extending from one beam to the other. This suggests that the interaction of the beams creates a spatially dependent variation of the refractive index in the beam crossing area. Each beam interacts with this structure to some extent similarly to the interaction with a dielectric cylinder. This nonlinear cross-beam refraction results in the formation of the observed arc-shaped lateral extensions as in Fig. 5 and Fig. 6.

Secondly, we point out that another possible mechanism of rescattering of light into observed arc-shaped lateral extensions of the cross-sectional distributions of the laser intensity of the pulses after their interaction is a four-wave mixing process [48,49]. In an isotropic medium and for the pulse delays close to zero, pairs of waves $k_1, k_2$ with wavevectors obtained by a rotation relative to the symmetry axis (see Fig. 10) satisfy phase matching conditions, and thus can experience parametric amplification in the four-wave mixing process driven by the pump beams $k_1, k_2$. The clarification of the exact nature of the formation of the lateral extensions of the beams requires further investigation. Now discussing the first effect, observed with the experimental arrangement of Fig. 1, namely the variations of white-light generation with changing delay, we note that at the conditions used in our experiment, the Rayleigh range is much longer than the length of the sample, and therefore the diffraction effects are relatively weak. The beam peak power $P = 1.9$ GW exceeds many times the self-focusing threshold, $P_c = 3.77 \frac{k_0^2}{8\pi n_0 n_2} = 0.53 MW$ for an 800nm pulse in flint glass ($n_0 = 1.81, n_2 = 10^{-15}$ cm$^2$/W).

The self-focusing length is determined by Marburger’s expression [50]

$$L_{sf} = 0.367 k a_0^2 \sqrt{\left(\sqrt{P/P_c} - 0.852\right)^2 - 0.0219}^{7/2}.$$ 

At the entrance face of the sample, the central bright spot of the beam diffraction on the entrance aperture has a radius of the 0.9 mm measured by the radius of the first dark circle in the diffraction pattern, and taking the radius at the $e^{-1}$ level (the formula for $L_{sf}$ is derived for a Gaussian beam, and we assume a
Fig. 10. Wave-vector diagram showing four-wave interaction: \( k_1,2 \)-pump pulses, \( k'_1,2 \)-amplified waves; the circle is perpendicular to the symmetry axis \( OO' \), the ends of all vectors of equal length satisfying phase matching conditions for four-wave interaction lie on a circle.

Gaussian distribution as approximation) the beam radius is determined to be \( a_0 \sim 0.42 \text{ mm} \), and the length of the self-focusing of the beam is \( L_s = 1.7 \text{ cm} \). Thus, the self-focusing length is comparable to the length of the sample. The presence of small-scale perturbations in the beams leads to the development of filaments on shorter lengths [51,52]. Since the filament formation length is comparable to the sample length, even relatively small variations of the laser intensity result in changes in the number of filaments, and consequently the amount of generated white light. In our experiments, the increase in the refractive index in the high-intensity central interaction region produces an additional lensing effect, providing the necessary initial increase of the intensity for development of filaments. However, if the mutual beam refraction-diffraction process becomes so strong that it significantly reduces (as is seen in Fig. 5 and Fig. 6) their peak intensity, which takes place at near zero delays, then the filamentation and white-light generation processes are suppressed (see Figs. 2–4). As the results in Fig. 5 show, the total power of the IR beams did not noticeably change with the delay variation. This demonstrates that in our case the main effect is due to the re-distribution of the laser intensity in the cross sections of the beams, rather than due to energy exchange between the IR beams [42–44,53–55], which can increase for strongly chirped pulses.

An interesting aspect of the beam interaction is the strong dependence on the mutual orientation of polarizations (Fig. 7). This points to the important role of interference in this interaction (see also Ref [44]). For beams with perpendicular polarizations, the increase of the intensity in the region of the intersection corresponds to their summation. The laser beams with the same orientation of polarizations can effectively interfere, reaching quadrupled values of a single beam intensity at the peaks. With an increase of the crossing angle the mutual coherence of the two pulses reduces leading to a reduction of the interaction length and can account for the reduction of the observed relative variation of the generated white light resulting with an increasing crossing angle, as is seen in Fig. 8(b).

The interference of the pulses leads to strong variations in the intensity (Fig. 9), which modulates the nonlinear contribution to the refractive index due to the optical Kerr effect and, as a result, changes the filamentation conditions for the beams propagating through their interaction region. The possibility to control the output of white light was limited to crossing angles \( \leq 3.6 \text{ deg} \).

5. Conclusion

We observed variation of white light generation in two interacting beams within a nonlinear Kerr medium. With a decrease in the relative temporal delays between the laser pulses, an enhancement of the white-light output was observed. This white-light enhancement was suppressed at delays near zero. The level of the suppressed output was less than the output level found at large delays, when pulses do not overlap in time. The enhancement of the white light generation occurred in the beam that was lagging. Thus, by changing the relative delay between the two interacting pulses, the white light generation can be controlled, namely
enhanced or suppressed. Arc-shaped lateral extension patterns of the IR light distributed around the main beams were observed and found to be more pronounced for smaller delays, where the powers of the central parts of IR beams exhibited dips. When the beams start interfering, following a decrease in the delay, this initially leads to an increase of the intensity in the interaction region, inducing respective changes in the refractive index due to the Kerr effect. As a result, after the beams pass each other they experience a cylindrical lensing effect, leading to a local increase of intensity and enhanced filament formation. However, with a further decrease of the delay, the redistribution of the laser intensity due to formation of the lateral extensions becomes significant and results in the decrease of both the peak intensity in the central parts of the beams and the number of created filaments and, consequently, in a lower level of the generated white-light.

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