1-J white-light continuum from 100-TW laser pulses

Yannick Petit,[1] Stefano Henin,[1] Walter M. Nakaema,[2] Pierre Béjot,[1] Axel Jochmann,[3] Stephan D. Kraft,[3] Stefan Bock,[3] Ulrich Schramm,[3] Kamil Stelmaszczyk,[2] Philipp Rohwetter,[2] Jérôme Kasparian,[1,*] Roland Sauerbrey,[3] Ludger Wöste,[2] and Jean-Pierre Wolf[1]

[1]*Teramobile, GAP, Université de Genève, 20 rue de l'Ecole de Médecine, CH-1211 Genève 4, Switzerland*
[2]*Teramobile, Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany*
[3]*Forschungszentrum Dresden-Rossendorf, Bautzener Landstraße 400, D-01328, Germany*

(Received 10 November 2010; published 7 January 2011)

We experimentally measured the supercontinuum generation using 3-J, 30-fs laser pulses and measured white-light generation at the level of 1 J. Such high energy is allowed by a strong contribution to the continuum by the photon bath, as compared to the self-guided filaments. This contribution due to the recently observed congestion of the filament number density in the beam profile at very high intensity also results in a wider broadening for positively chirped pulses rather than for negatively chirped ones, similar to broadening in hollow-core fibers.

DOI: 10.1103/PhysRevA.83.013805 PACS number(s): 42.65.Jx, 42.65.Ky

I. INTRODUCTION

The propagation of ultrashort laser pulses in air or other transparent media is characterized by filamentation [1–5], a propagation regime where a dynamical balance is established between Kerr self-focusing and defocusing by further nonlinear processes. The plasma generated at the nonlinear focus is generally considered as the main process in that regard, although we recently suggested that the higher-order Kerr effect could play this role [6], in particular for longer wavelengths [7] and/or shorter pulses [8]. Filamentation is now well characterized from the mJ, GW level to the sub-J, TW levels, with potential major atmospheric applications such as lightning triggering and control or laser-induced condensation [9,10].

Recently, we demonstrated that filamentation still occurs at multi-J levels [11], with similar physics as at lower powers. However, at intensities above $1-2 \times 10^{11}$ W/cm^2, we observed a change in the spatial dynamics of multifilamentation [12]. Below this threshold, the number of filaments is proportional to the incident power P, one filament being generated per $5-7P_{\mathrm{cr}}$ available in the incident pulse [13]. The critical power for filamentation P_{cr}, which is close to 3 GW in air at 800 nm, is the minimum incident power required for the self-focusing to balance diffraction. In this regime, filaments are typically 100–1000 times more intense than the surrounding photon bath (5×10^{13} W/cm^2 [14,15] vs some 10^{10} W/cm^2) and roughly concentrate 10% of the total beam fluence although covering only a few 10^{-4} of the total beam surface [2,3]. As a consequence of such difference in intensity levels, the contribution of filaments to the nonlinear behavior of the beam is so dominant that the contribution of the photon bath can be fully neglected.

In contrast, above $1-2 \times 10^{11}$ W/cm^2, the filament number is governed by the surface of the beam profile. As a consequence, filaments cover a constant fraction of the beam surface. Since their intensity is still clamped, the fraction of the beam fluence carried by the filaments decreases, the photon bath intensity increases. It can even exceed the TW/cm^2 level, reducing the intensity ratio between the filaments and the bath to one order of magnitude [12]. Furthermore, filaments have been observed to be connected by regions of higher intensity within the photon bath. In this regime, a substantial contribution of the photon bath to the nonlinearity can therefore be expected.

In this article, we investigate the spectral broadening along the horizontal propagation of a 3-J, 100-TW laser pulse over 42 m in air. We measure a conversion efficiency of several tens of percentage points, resulting in up to 1 J of white-light continuum. Furthermore, in this high-intensity regime, the effect of the chirp is opposite to the standard behavior in the usual low-intensity filamentation regime, but rather comparable to that observed in hollow-core fibers, where a positive chirp yields more spectral broadening than a negative one [16,17]. Finally, we observe a line at 771 nm, which we interpret as a Rabi-shifted oxygen atomic multiplet OI line.

II. EXPERIMENTAL SETUP

Experiments were performed using the Ti:Sa chirped-pulse amplification (CPA) chain of the Forschungszentrum Dresden-Rossendorf providing up to 3-J, 100-TW pulses of 30 fs duration, at the repetition rate of 10 Hz and central wavelength of 800 nm. The initial spectrum was 60 nm broad (full width at half maximum). The pulse energy was adjusted by using a half-wave plate and a polarizer before the grating compressor. The pulses were transported in a vacuum tube to a 42-m-long experimental hall, where they were launched into air, collimated (i.e., as a parallel beam) with a diameter of ~10 cm, through a 6-mm-thick fused silica window. Detuning the compressor allowed us to impose a chirp onto the pulses. In the following, the chirp values always refer to the exit of the output window.

The pulses underwent multiple filamentation in the high-intensity regime, as described in detail elsewhere [12], that is, with a relatively low filament number as compared with the available power, and a high photon bath intensity. The beam-averaged spectrum was collected on a diffusive screen using a 10-μm core fiber of 25° aperture (full angle), fixed 50 cm away from the screen. It was analyzed by an OceanOptics USB4000 spectrometer providing 0.27-nm resolution over the

*jerome.kasparian@unige.ch

200-to-1100-nm range. Spectral calibration of the spectrometer was checked to be better than 1 nm for two specific wavelengths of HeNe laser at 632.8 mn and frequency-doubled Nd:YAG laser at 532 nm. The spectra were numerically averaged over two to three single-shot acquisitions and corrected by the spectral response of the spectrometer.

We quantitatively characterized the spectral broadening using three independent approaches: (i) the full width at $1/e^2$; (ii) the second moment of the intensity distribution; and (iii) the efficiency of the supercontinuum generation, that is, the ratio of the energy in the continuum ($\lambda < 765$ nm and $\lambda > 845$ nm) to the total incident pulse energy. These three approaches yielded similar results and dependencies, so that in the following we mainly focus on the generation efficiency.

The analysis of the experimental data was supported by numerical simulations of the output spectrum after the pulse propagation. In order to compare the respective contributions of the photon bath and the filaments to the spectral broadening, we independently investigated two regimes. In the first one, we considered a photon bath without filamentation, by performing $1D + 1$ (propagation distance and time) simulations based on the unidirectional pulse propagation equation described in Ref. [18]. In the second regime, we simulated a filamenting beam in a $2D + 1$ (propagation distance, radial distance, and time) propagation model based on the nonlinear Schrödinger equation as detailed in [7].

III. RESULTS AND DISCUSSION

The afore-mentioned limited number of filaments, relative to the incident power of the laser pulses, could have been expected to reduce the white-light generation efficiency. However, we do not observe such reduction. Rather, the white-light generation appears quite as efficient as in previous experiments in the low-intensity regime (300 mJ, 100 fs) using the Teramobile laser [19] (Fig. 1).

In particular, in the present experiment, the exponential decrease on the wings of the spectrum is comparable with that of the spectrum in experiments at lower intensity [19] (130 nm/decade vs 140 nm/decade on the blue side of the spectrum). Still, considering the incident energy in the present work, the comparable conversion efficiency of several tens of percentage points (see Fig. 3) results in an unprecedented supercontinuum energy of up to the joule level.

We also observed that the filament number appears to have very little influence on the white-light conversion efficiency and that positive chirps are more favorable to spectral broadening than negative ones. As shown in Figs. 2 and 3, at a fixed energy or power, the more positive chirps (+100 fs) yield up to twice as much spectral broadening as its negative counterpart. This finding is unexpected since (i) the supercontinuum is generally considered to originate from the filaments, and (ii) negative chirps lead to self-compression due to group-velocity dispersion and are expected to yield a higher peak power and thus to generate more white-light by increasing the temporal variations of the intensity, as well as by favoring the formation of filaments. For example, 100-fs negatively chirped pulses result in almost Fourier-limited pulses after 42 m of propagation. In contrast, positively chirped pulses spread temporally all along their propagation.

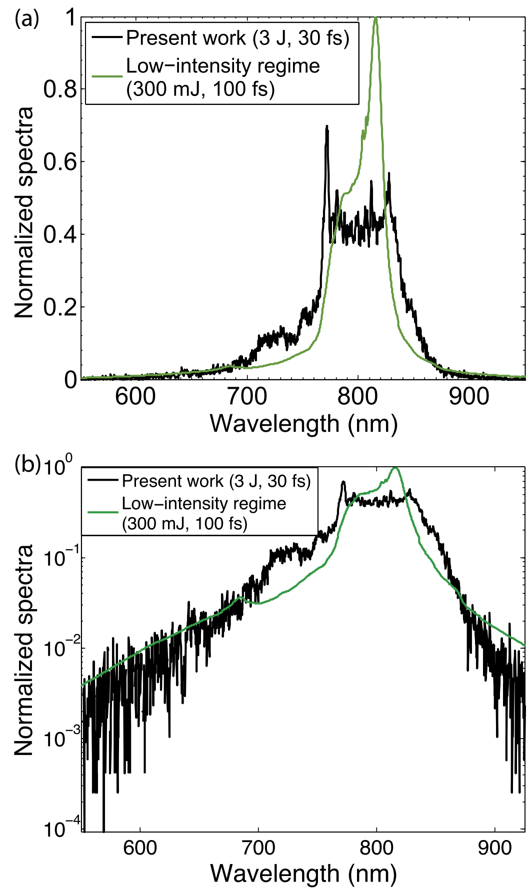

FIG. 1. (Color online) Efficiency of the white-light generation by a 2.53-J, 30-fs, Fourier-limited laser pulse in 10-cm beam diameter after 42 m of propagation, and by the Teramobile laser (300 mJ, 100 fs in 5-cm beam diameter) after the filamenting region ($z \sim 40$ m) [19]. Both spectra are normalized to their total area: (a) linear scale; (b) logarithmic scale.

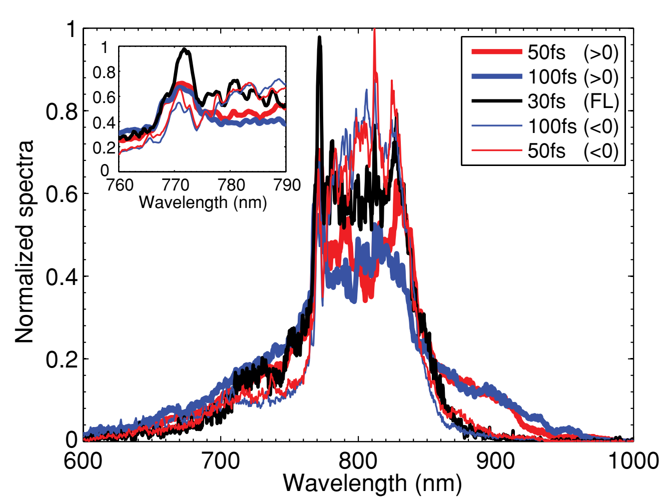

FIG. 2. (Color online) Spectra generated after 42-m propagation of a 2.53-J pulse of 30-fs Fourier-limited (FL) duration and the same pulse both positively and negatively chirped (50- and 100-fs pulse durations). All spectra have the same normalization factor, so that their amplitudes can be compared. (Inset) Blowup of the 760-to-790-nm spectral region, exhibiting a peak at 771.5 nm for Fourier-limited and positively chirped pulses.

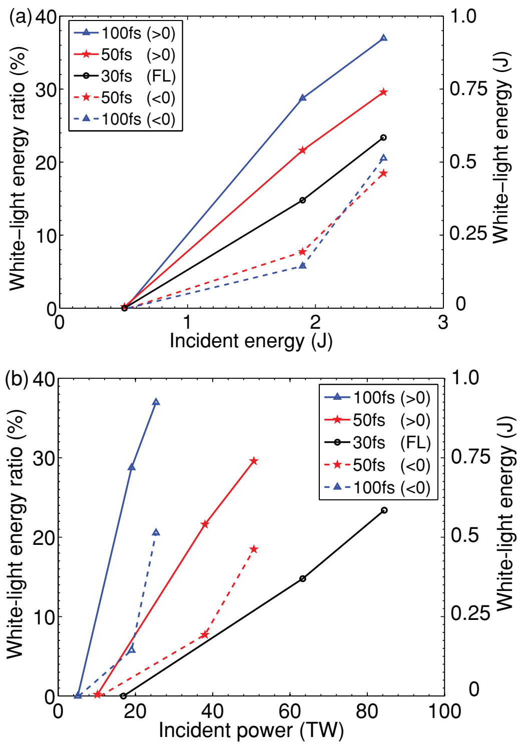

FIG. 3. (Color online) Conversion efficiency and corresponding energy of the white-light continuum after 42-m propagation of a 2.53-J pulse, as a function of incident pulse energy (a) and power (b). The continuum is defined as the spectral range out of the 765-to-845-nm spectral range.

These observations can, however, be understood by considering the role of the photon bath in the white-light generation. A rough evaluation of this contribution can be obtained by considering the classical expression of the spectral broadening by self-phase modulation,

$$\frac{\Delta\omega}{\omega_0} = n_2 \frac{dI}{dt}\frac{z}{c}, \tag{1}$$

where $\Delta\omega$ is the frequency shift, ω_0 is the incident frequency, $n_2 = 1.1 \times 10^{-19}$ cm^2/W [20] is the nonlinear refractive index of air, I is the incident intensity, z is the propagation distance, and c is the speed of light. After $z = 42$ m, the intensity of the photon bath ($I \sim 1$ TW/cm^2 [12]) yields $\Delta\omega/\omega_0 \sim 0.5$, showing that the self-phase modulation of the photon bath yields sufficient spectral broadening to account for the spectra observed in our experiment. Note that the use of a larger value of n_2 (e.g., 3.2×10^{-19} cm^2/W [2]) would result in an ever-wider broadening. This contribution can indeed be observed in real-color photographs of the beam on a screen (Fig. 4). It will even be enhanced on the high-intensity regions connecting the filaments.

Since the detection system collects light from the whole beam profile, the recorded spectrum can be considered, to the first order, as the weighted sum of the contributions from the photon bath and from the filaments. The weight of any portion of the beam profile S is the integral of the fluence over its area. In other words, the respective contributions of the filaments and the photon bath to the observed spectrum are roughly proportional to the total energy they respectively carry.

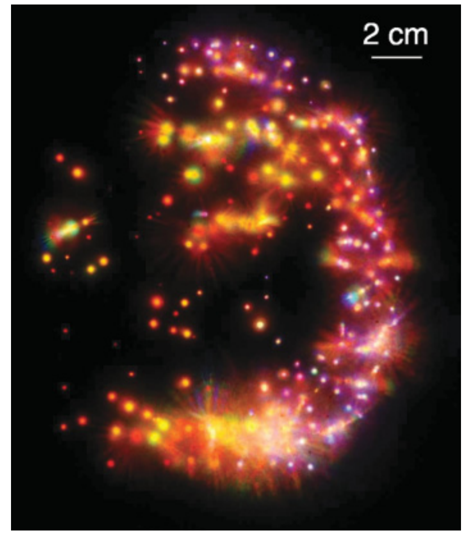

FIG. 4. (Color online) Beam profile on a screen of a 30-fs, 1.65-J pulse after 15-m propagation, that is, at the beginning of the filamenting region. The white contribution from the photon bath is clearly visible.

In our high-intensity regime, comparing the contributions to spectral broadening of 500 filaments of 100 μm diameter and 5×10^{13} W/cm^2 with that of 80 cm^2 of photon bath conveying 1 TW/cm^2 yields an energy ratio of 98%/2% in favor of the photon bath. Furthermore, as discussed earlier, the latter is widely broadened. Therefore, in spite of their locally more efficient broadening, the filaments will provide a minor source of spectral broadening in this configuration.

In contrast, at lower intensity, for example, under the “low-intensity” conditions displayed in Fig. 1, the contribution of 50 filaments has to be compared with a bath typically conveying a few TW. In this case, the filaments bear ~10% of the total beam power and will therefore contribute substantially to the overall spectrum. Furthermore, in the latter case the photon bath intensity of ~0.1 TW/cm^2 only allows a marginal relative spectral broadening within it. As a result, the photon bath contribution to the spectral broadening is marginal, as commonly observed in experiments at lower intensity [19].

Considering that the photon bath is the main source of spectral broadening provides a clear interpretation of our observation that positively chirped pulses efficiently generate white light. In the photon bath, especially over long propagation distances, the main nonlinearity at play is self-phase modulation (SPM), which shifts the leading edge of the pulse to longer wavelengths and the trailing edge to shorter wavelengths. In positively chirped pulses, the leading edge already bears the longer wavelengths, and the shorter wavelengths are on the trailing edge. This time-dependent frequency offset within the pulse is therefore reinforced by SPM. On the other hand, if the pulse is negatively chirped, SPM redshifts its blueshifted leading edge and blueshifts its redshifted trail. A positive chirp is therefore more favorable to the spectral broadening than a negative chirp. Our results show that, under our conditions, this effect overrides the pulse temporal recompression or spreading due to group-velocity dispersion.

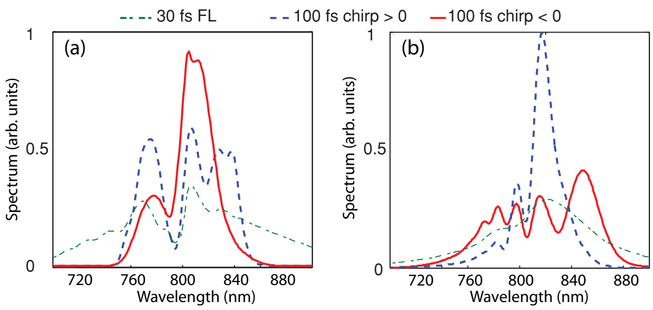

FIG. 5. (Color online) Simulated transversely integrated spectrum for pulses of 30 fs, Fourier-limited, and 100 fs, positively and negatively chirped. (a) $1D+1$, 30 mJ/cm^2 (1 TW/cm^2 for 30-fs pulses); (b) $2D+1$, 0.81 mJ (9 P_{cr} for 30-fs pulses).

This mechanism leading to wider spectral broadening for positive chirps has already been observed in hollow-core fibers [16,17]. Indeed, supercontinuum generation by the photon bath can be expected to be an essentially one-dimensional problem, quite similar to propagation in a fiber. Such regime is also similar to the one observed in the case of two beams crossing each other in glass, where substantial SPM within the photon bath depleted the power available for filament generation and hence reduced the filament number [21].

Chirp therefore provides a convenient way of controlling the width of the supercontinuum generated by high-intensity laser pulses. Such control can be used to maximize the generated bandwidth, as is required, for example, for multispectral Lidar applications [1,4], but also to concentrate the white light over a restricted spectral region around the fundamental wavelength for specific applications [22].

The contrast between the high- and low-intensity regimes is illustrated in Fig. 5, which displays the simulated spectra generated by the photon bath and by a filamenting beam. In the $1D+1$ code, representative of the photon bath after 42 m propagation, with 30 mJ/cm^2, a positive chirp yields more broadening in the 750-to-850-nm spectral region in qualitative agreement with the experimental observations. However, the broader spectrum obtained for FT-limited pulses provides a conversion efficiency (as defined in Fig. 3) of 50%, vs 10% for 100-fs, positively chirped pulses and 5% for 100-fs, negatively chirped pulses, which does not quantitatively reproduce our experimental results. This deviation could be due to the fact that our model treats the photon bath as homogeneous, disregarding the interactions between the filaments, especially the higher-intensity strings along which they are organized [12]. However, adequately considering these structures would require three-dimensional modeling of the full beam, which is well beyond our current computing capabilities.

In contrast, in the $2D+1$ simulation of a filamenting beam on a reduced scale compatible with computing capabilities (0.81 mJ, i.e., 9 P_{cr} at 30 fs, propagation distance 1.5 m), the main effect of the chirp is to affect the well-known shift the central beam wavelength [23]. Furthermore, a negatively chirped pulse broadens the spectrum more efficiently than a positively chirped one. The chirp dependence of our experimental data regarding the spectral broadening therefore supports the preceding conclusion that the photon bath rather than the filaments provides the main contribution to the observed spectral broadening at the TW/cm^2 intensity level investigated in the present work.

As shown in the inset of Fig. 2, our experimental data exhibit a sharp line at 771.5 nm. This line is clearly visible for Fourier-limited and positively chirped pulses, with up to a few percentage points of the total spectrum energy. It is much weaker for negative chirps. This line may be the oxygen atomic multiplet OI($3p^5P$–$3s^5S$) centered at 777.4 nm in the plasma generated in air by the filaments [24,25]. This line would subsequently be Rabi-shifted due to the high intensity, as recently observed by Compton *et al.* [26]. The influence of the chirp could be understood by considering that the proposed process requires two steps: (i) the formation of excited atomic oxygen and (ii) the stimulation of the emission by the same incident pulse. Since the excitation mainly occurs at the peak of the pulse where the intensity is highest, stimulation would mainly occur on the trail of the pulse. It would therefore be more efficient if the blue side of the spectrum is still significant on this trail, that is, for positively chirped pulses. If this interpretation can be confirmed, the observation of a Rabi shift may suggest that further unexpected effect occurs in the high-intensity multifilamentation regime, although such effects are beyond the scope of the present work.

IV. CONCLUSION

As a conclusion, by investigating the supercontinuum generation from 3-J, 30-fs laser pulses, we have observed the occurrence of a high-intensity filamentation regime where the photon bath, rather than the filaments as in the case of lower intensity, provides most of the spectral broadening. As a consequence, similar to broadening in hollow-core fibers, the widest spectral broadening is observed for positively chirped pulses rather than for negatively chirped ones. The substantial contribution of the photon bath also allows the generation of an unprecedented level of 1 J of supercontinuum.

[1] J. Kasparian *et al.*, Science **301**, 61 (2003).
[2] A. Couairon and A. Mysyrowicz, Phys. Rep. **441**, 47 (2007).
[3] L. Bergé, S. Skupin, R. Nuter, J. Kasparian, and J.-P. Wolf, Rep. Prog. Phys. **70**, 1633 (2007).
[4] J. Kasparian and J.-P. Wolf, Opt. Express **16**, 466 (2008).
[5] S. L. Chin, S. A. Hosseini, W. Liu, Q. Luo, F. Théberge, N. Aközbek, A. Becker, V. P. Kandidov, O. G. Kosareva, and H. Schroeder, Can. J. Phys. **83**, 863 (2005).
[6] P. Béjot, J. Kasparian, S. Henin, V. Loriot, T. Vieillard, E. Hertz, O. Faucher, B. Lavorel, and J.-P. Wolf, Phys. Rev. Lett. **104**, 103903 (2010).

[7] W. Ettoumi, P. Béjot, Y. Petit, V. Loriot, E. Hertz, O. Faucher, B. Lavorel, J. Kasparian, and J.-P. Wolf, Phys. Rev. A **82**, 033826 (2010).

[8] V. Loriot, P. Béjot, W. Ettoumi, Y. Petit, J. Kasparian, S. Henin, E. Hertz, B. Lavorel, O. Faucher and J.-P. Wolf (to be published in Laser Physics.), e-print arXiv:1011.0841v1.

[9] J. Kasparian *et al.*, Opt. Express **16**, 5757 (2008).

[10] P. Rohwetter *et al.*, Nat. Photon. **4**, 451 (2010).

[11] P. Béjot *et al.*, Appl. Phys. Lett. **90**, 151106 (2007).

[12] S. Henin *et al.*, Appl. Phys. B **100**, 77 (2010).

[13] G. Méjean *et al.*, Phys. Rev. E **72**, 026611 (2005).

[14] J. Kasparian, R. Sauerbrey, and S. L. Chin, Appl. Phys. B: Lasers Opt. **71**, 877 (2000).

[15] A. Becker, N. Aközbek, K. Vijayalakshmi, E. Oral, C. M. Bowden, and S. L. Chin, Appl. Phys. B **73**, 287 (2001).

[16] Z. Zhu and T. G. Brown, Opt. Express **12**, 689 (2004).

[17] J. M. Dudley, G. Genty, and S. Coen, Rev. Mod. Phys. **78**, 1135 (2006).

[18] P. Béjot, B. E. Schmidt, J. Kasparian, J.-P. Wolf, and F. Legaré, Phys. Rev. A **81**, 063828 (2010).

[19] P. Maioli, R. Salamé, N. Lascoux, E. Salmon, P. Béjot, J. Kasparian, and J.-P. Wolf, Opt. Express **17**, 4726 (2009).

[20] V. Loriot, E. Hertz, O. Faucher, and B. Lavorel, Opt. Express **17**, 13429 (2009); **18**, 3011(E) (2010).

[21] K. Stelmaszczyk, P. Rohwetter, Y. Petit, M. Fechner, J. Kasparian, J.-P. Wolf, and L. Wöste, Phys. Rev. A **79**, 053856 (2009).

[22] V. P. Kandidov, O. G. Kosareva, I. S. Golubtsov, W. Liu, A. Becker, N. Akozbek, C. M. Bowden, and S. L. Chin, Appl. Phys. B **77**, 149 (2003).

[23] A. M. Zheltikov, Phys. Rev. A **79**, 023823 (2009).

[24] P. Rambo, J. Schwarz, and J.-C. Diels, J. Opt. A **3**, 146 (2001).

[25] Y. E. Geints, A. M. Kabanov, G. G. Matvienko, V. K. Oshlakov, A. A. Zemlyanov, S. S. Golik, and O. A. Bukin, Opt. Lett. **35**, 2717 (2010).

[26] R. Compton, A. Filin, D. A. Romanov, and R. J. Levis, Phys. Rev. Lett. **103**, 205001 (2009).