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Path length and spectrum of single-cycle mid-IR light bullets in transparent dielectrics

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Abstract. **Filamentation of femtosecond laser radiation with a wavelength of 800–3900 nm and a power slightly exceeding the critical self-focusing power is studied using the spectral method and the method of laser coloration in LiF crystal. It is found that the length of a filament formed in the single-pulse regime increases with increasing excitation wavelength from a few tens of micrometres at 80 nm to hundreds of micrometres at 3900 nm. In the spectral region of anomalous group velocity dispersion, starting from 2600 nm, the initially smooth luminescence profile of the long-lived induced colour centres acquires a periodic structure, demonstrating the formation of a light bullet with a duration of about one cycle of the light field oscillation and a diameter smaller than 10 μm. The path length of such bullets does not exceed 0.5 mm in the single-pulse regime and 2.7 mm in the waveguide regime. A consequence of periodic modulation of the bullet light field in the process of propagation, observed experimentally and confirmed by calculations, is the appearance of sidebands near the excitation wavelength, as well as the appearance of visible spectral components in the supercontinuum radiation, whose angular divergence increases with increasing wavelength.**

Keywords: *filamentation, femtosecond pulses, anomalous group velocity dispersion, plasma channels, light bullets.*

1. Introduction

One of the interesting and urgent problems of modern nonlinear optics is the study of the processes that lead to the formation of light bullets (LBs) [1], i.e., the space- and time-localised self-consistent nonlinear excitations formed in transparent dielectrics under the action of high-power ultrashort laser pulses. The implementation of such self-consistent structures capable of propagating through considerable distances in a medium without spreading and significant distortions is one of the most difficult experimental problems not yet solved. In homogeneous media with cubic nonlinearity, the three-dimensional solitons are unstable and can collapse [2], so that the issue of their experimental implementation remains open. On the other hand, the studies of ultrafast dynamics of compressed wave packets having a length of 1–2 optical oscillations, a transverse size of the wavelength order, and a peak intensity up to 10^{14} W cm^{-2}, produced during femtosecond laser pulse filamentation in bulk isotropic transparent dielectrics free of any guiding structures become rather urgent for extreme laser optics due to the appearance of subterawatt sources of ultrashort pulses in the mid-IR spectral range [3], promising for multiple applications.

From the point of view of formal description, the possibility of compressing a pulse in time is analogous to spatial self-focusing, with the role of diffraction played by the dispersion of the medium. The compression is possible for the negative group velocity dispersion (GVD) [4]. In contrast to the spatial compression of light beams in amplifying media at self-focusing of near-IR pulses that leads to the power growth restriction, which was studied as early as in the past century [4, 5], in the mid-IR region the group velocity dispersion becomes anomalous. This leads to the nonlinear compression of pulses simultaneously in time and space, i.e., the LB formation. The peak powers of femtosecond pulses generated by up-to-date lasers can achieve a few petawatt, while the peak intensities of unfocused radiation approach 10^{13}–10^{14} W cm^{-2}. These values can essentially exceed the threshold of LB formation, accompanied by considerable losses due to the change of not only the spatial but also the temporal and spectral parameters of the pulses in the process of propagation through optical elements of the setup. In the pulse spectrum the supercontinuum (SC) radiation is formed, whose band covers the spectral region from UV to IR.

The term 'light bullet' was introduced in Ref. [1], where the concept of LB formation under the nonlinear optical interaction of a wave packet in a dispersion medium with cubic nonlinearity was formulated based on the analysis of the quasi-optics equation in the aberration-free approximation [6]. Under conditions of anomalous group velocity dispersion (AGVD), the role of a compressor is played by the propagation medium itself, where the pulse is compressed in time due to the phase modulation caused by Kerr nonlinearity. The possibility of forming a quasi-periodic sequence of LBs in an isotropic nonlinear medium with AGVD was predicted in Ref. [7]. Because for many transparent dielectrics GVD is anomalous in the mid-IR range, the invention of high-power femtosecond radiation sources in this wavelength range [3] opened the way for the experimental observation of broadband SC generation in the process of filamentation under the conditions of zero and anomalous GVD by many research teams [8–12]. The numerical results on the LB sequence formation in the course of pulse filamentation under AGVD conditions [13] were confirmed experimentally by the autocorrelation measurements [14]. The generation of subse-

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quent bullets occurs due to the energy supply from the leading edge of the pulse caused by the light field phase modulation under AGVD conditions. The measured LB duration was about two periods of optical oscillation during filamentation in fused silica at a wavelength of 1800 nm; in this case, according to the estimates of the plasma channel length the bullet path length did not exceed a few millimetres [14]. The interpretation of such experiments in Ref. [15] gave the LB path length of 10 mm in sapphire, and in Ref. [16] the path length in fused silica was estimated as a few centimetres, which essentially exceed the values obtained in Refs [13, 14]. At present, the LB path length is not estimated unambiguously, and its determination is of great practical interest, in particular, for atmospheric optics, to which the results obtained in solid-state dielectrics can be scaled.

Light bullets can be detected by the glowing of plasma channels and the scattered SC radiation, formed by each bullet. In the strong field of a light bullet, the low-temperature laser plasma is generated. The defocusing of radiation in this plasma leads to a steep fall of intensity at the trailing edge and, as a consequence, to super-broadening of the SC spectrum towards the anti-Stokes region [17]. Under AGVD conditions, a narrow isolated wing is formed in this spectral region; it was studied in detail in Refs [18, 19]. In our experiments [20] performed with SiO_2, we found that in the course of filamentation of radiation with a power exceeding a critical power of self-focusing, a sequence of LBs is formed in which bullets release equal discrete portions of SC energy. The plasma channels together with the colour track of the scattered SC were photographed from the side, but the exposure of the obtained photos ($\sim$10000 pulses) was too long to determine their parameters unambiguously under natural fluctuations of pulse parameters. The simultaneous appearance of the plasma channel and an abrupt increase in the signal, recording the energy in the visible band of SC, indicated the formation of the next LB in the filament that had a threshold character. The experiment allowed one to relate the LB formation with the appearance of a plasma channel and the release of a discrete energy portion in the visible band of the SC spectrum in agreement with the results of numerical modelling [13] and the interference model of supercontinuum generation [21]. Although the SC energy was measured for each pulse, it was essentially necessary to accumulate the signal from at least a few hundred light pulses to get the information about the bullet path length from the glowing tracks of plasma channels and scattered SC, induced by its light field. A similar situation arises in the method of three-dimensional imaging used by a number of foreign groups [15]. The irreproducibility of the laser pulse parameters from pulse to pulse leads to essential distortion of the obtained information.

2. Methods

The laser coloration method [22–24] is free of the above drawbacks. In this method the dynamics of SC appearance and development in the process of filamentation of single femtosecond mid-IR pulses in LiF is detected by the change in the density of long-lived colour centres (CCs), arising in the light field due to multiphoton processes in the generated filament. The luminescence of these CCs is so intense that the long-lived structures comprised by them, created by only one laser pulse, may be easily detected and studied under the subsequent illumination by a cw laser at their absorption band near 450 nm. The application of the laser coloration method in the LiF crystal allows direct recording of the light field amplitude variation and the parameters of extremely compressed wave packet propagating in the filament [23, 24]. First, this method allows thorough investigation of the three-dimensional structure of the LB light field during the entire propagation through the material with a spatial resolution better than 1 μm (determined exclusively by the resolution of the used microscope) after the propagation of only one pulse through the material. Second, due to the multiphoton nature of the CC generation, this method provides the registration of parameters only in the near-axis part of the wave packet, i.e., the intensity distribution directly in the LB cross section without the surrounding background. Third, the laser coloration method is not an on-line method, so it does not record the radiation of SC, conical emission, and plasma channel, always present in the on-line methods and leading to erroneous estimation of the LB lifetime and path length in the material, when it is necessary to accumulate the signal from many hundreds and thousands of laser pulses. Here these values are determined unambiguously, since only one laser pulse is used in the method. The appearance of long-lived colour centres in LiF is accompanied by an increase in the refractive index at the filament axis and the formation of an optical waveguide in the dielectric volume [22]. The use of multipulse excitation allows the implementation of the waveguide regime of LB propagation, practically unexplored in homogeneous materials.

3. Measurement scheme and experimental results

The experimental studies were performed at the spectroscopic stand of the Shared Use Centre at the Institute of Spectroscopy of RAS. The experimental setup is thoroughly described in Refs [23, 24]. We used the pulses with a repetition rate from a few Hz to 1 kHz at wavelengths 800–3900 nm, belonging to the region of normal, zero, and anomalous GVD in LiF. The FWHM duration of the pulse amounted to 40 fs at the wavelength 800 nm, 70 fs at the wavelength 1240 nm, and 100 fs at the wavelength 2500–3900 nm; the pulse energy varied from 0.2 to 100 μJ. The laser pulses were focused onto the input face of the 40-mm-long LiF crystal using a thin CaF_2 lens with the focal length 200 mm at the wavelength 800 nm.

Figures 1–4 present the results of applying the laser coloration method to measure the CC structures, as well as the SC spectra obtained both in the single-pulse and multipulse filamentation regimes in LiF. In the latter case, the long-lived waveguides were formed by a different number of pulses at a low repetition rate (4 Hz) to exclude thermal effects, and then without changing the sample position the spectral and angular characteristics of SC or the LB tracks were measured for a different number of pulses. The measurement scheme is analogous to that in Ref. [24]. The density profiles of CCs induced in LiF under single-pulse filamentation of femtosecond pulses in the mid-IR and near-IR ranges at a wavelength varied from 800 to 3900 nm are presented in Fig. 1. It is seen that under the action of a single pulse at the wavelength corresponding to the normal GVD in LiF, the length of the region with high intensity of the light field (i.e., the filament length) increases with the wavelength from 70 μm at 800 nm to 120 μm at 1240 nm (the region of zero GVD) and then to 250 μm at 2100 nm (the region of weak AGVD). In this case, the filament profiles are rather smooth.

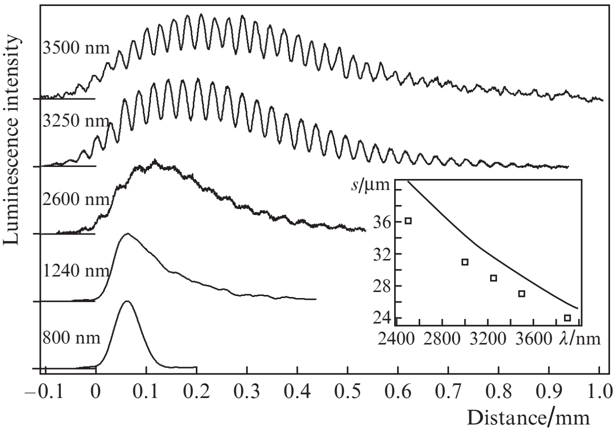

Figure 1. Luminescence profiles of colour centres induced in LiF under single-pulse filamentation of femtosecond pulses of the mid-IR and near-IR ranges at a wavelength varied from 800 nm to 3500 nm. The inset shows the dependence of the CC density modulation period on the wavelength (see also Fig. 2); the solid line is the result of calculation.

At sufficiently large AGVD (starting from 2600 nm), a regular structure consisting of colour centres appears in the profiles, which directly reproduces the influence of the light field absolute phase of the extremely compressed wave packet on the nonlinear optical interaction (Fig. 2). This influence recorded as a strictly periodic variation of the density of CCs with the distance leads to the light bullet 'breathing' during its passage through the filament [23, 24]. The period of LB 'breathing' decreases with increasing wavelength (see the inset in Fig. 1, and Fig. 2). It is well known that when a pulse having a duration of the order of one light oscillation cycle (the so-called single-cycle pulse) propagates through a dispersion medium, the cyclic periodic modulation of the light field maximal amplitude occurs during its propagation [25] due to the difference of the envelope and frequency carrier velocities of the pulse. This process is typical only for single-cycle pulses, and with the increased number of periods, the modulation effect practically vanishes. As to the light bullets, from the theoretical point of view if they arise in a medium with high enough anomalous dispersion, then they would compress to the duration equal to one field cycle, although experimentally it has not been observed yet. In our experiments [23, 24] the possibility of detecting such LBs using the laser coloration methods was implemented for the first time. Therefore, the appearance of the observed regular modulation of the CC density profile, induced at the filamentation in LiF (see Fig. 2), evidences in favour of the single-cycle LB formation.

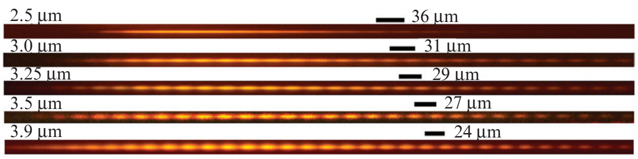

Figure 2. Luminescence tracks of CCs induced in LiF under single-pulse filamentation of femtosecond pulses at a wavelength varied from 2500 to 3900 nm.

4. LB path length

According to the experimental data, the path length of an LB somewhat grows with increasing wavelength, but, in contrast to the data of Refs [13, 14], does not exceed a few hundred micrometres, which corresponds to the lifetime of the order of a few picoseconds. The view of the LB track does not change with increasing laser pulse energy from the threshold of filament formation to the generation of the second LB, the structure of which exactly coincides with that of the first LB (Fig. 3), thus confirming the bullet robustness [20]. However, the spread of filament starting points in the formation of the second LB is significantly greater than the corresponding spread for the first bullet. Therefore, the length of the second filament recorded at a multipulse exposure (as well as the plasma channel length) essentially exceeds the length of the first one, to say nothing of the real length recorded in the single-pulse regime. This yields a wrong estimate of the LB lifetime and path length in the material when interpreting practically all the experiments performed earlier (see, e.g., [15, 16]), since they used a multipulse exposure.

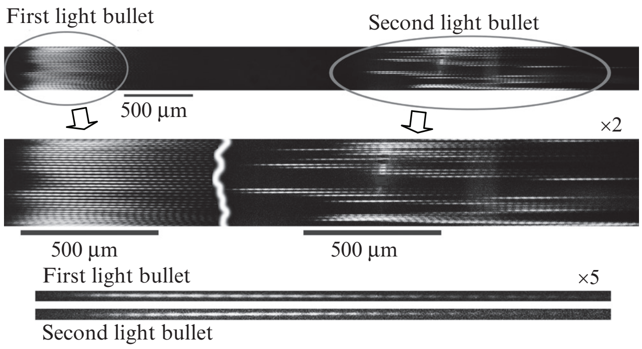

Figure 3. Luminescence tracks of CCs induced under single-pulse filamentation of femtosecond pulses at a wavelength of 3500 nm in LiF. The pulse energy corresponds to the appearance of two light bullets.

5. Spectral measurements and analytical calculation

When an LB was formed in the process of filamentation in LiF, we recorded the appearance of side bands, shifted to the long-wavelength and short-wavelength sides in the spectrum of the femtosecond pulse at the sample output (Fig. 4). We explain these bands by the contribution of the LB, in which the periodic modulation of the light field in the process of propagation leads to the appearance of side bands near the excitation wavelength, shifted by the modulation frequency; this effect being well-known in radiophysics. Similar structure of the light bullet spectrum was recently observed and reported in Ref. [26] (without explanation of reasons) under filamentation of 2250-nm femtosecond pulses in sapphire (see Fig. 8 in Ref. [26]). The modulation also leads to the appearance of visible-range spectral components in the SC radiation [27], whose angular divergence increases with increasing wavelength, i.e., in contrast to the common conical emission, the long-wavelength components propagate at greater angles than the short-wavelength ones.

The growth of angular divergence with increasing wavelength of the SC spectral components is explained within the

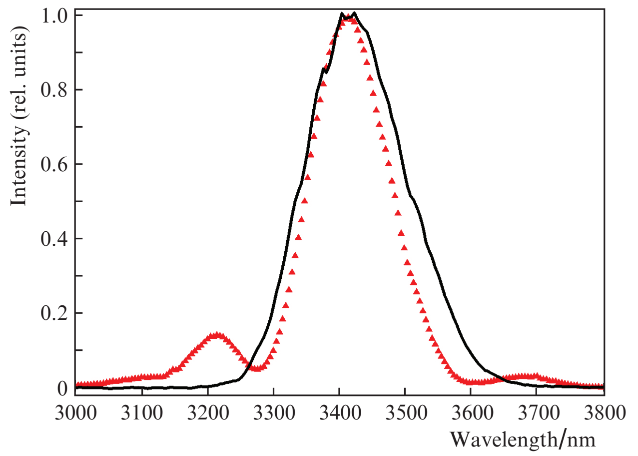

Figure 4. Spectra of femtosecond pulses at a wavelength of 3425 nm at the input (solid curve) and the output (points) of the sample in the process of single-pulse filamentation in LiF.

framework of the interference model [21], which considers the LB as a broadband source of SC moving in the medium with the velocity close to the group velocity of the pulse. Previously, using this model the physical interpretation of the anti-Stokes band appearing in the SC spectrum of the studied LB was given, as well as that of the shift and narrowing of this band with increasing incident radiation wavelength [28]. Using the interference model, the dispersion equation was derived that allows the SC anti-Stokes band maximum to be calculated and the results of the known experiments to be generalised [29].

Consider the frequency-angular spectrum of the conical emission from a single-cycle LB within the framework of the interference model. The LB 'breathing' is a periodic variation of the electric field strength amplitude E with the propagation coordinate z, which in the first approximation can be presented as

$$E(z) = E_0 - E_0 \frac{h}{2}\left(1 - \cos\frac{2\pi}{\Lambda} z\right), \quad (1)$$

where E_0 is the maximal value of the light field strength in the LB; h is the relative value of the strength oscillations; $\Lambda = \lambda_0 v_g/[2\pi n(\lambda_0)\Delta v]$ is the oscillation period; λ_0 is the centre wavelength; v_g is the LB group velocity; Δv is the difference between the phase and group velocity; and $n(\lambda)$ is the refractive index of the medium. From the interference model point of view, the periodic variations of the LB field amplitude lead to similar oscillations of the amplitude of the moving point source of the supercontinuum. In this case, the frequency-angular distribution of the intensity of SC spectral components, formed by the LB at its path length L in the far-field zone, is determined by the expression [21]:

$$I(\theta,\omega) = \text{const} \times \left| \int_0^L \left[1 - \frac{h}{2}\left(1 - \cos\frac{2\pi}{\Lambda} z\right)\right] \exp[\mathrm{i}\Delta\varphi(\theta, \omega, z)]\mathrm{d}z \right|^2, \quad (2)$$

where

$$\Delta\varphi(\theta, \omega, z) = z\left\{\frac{\omega_0 - \omega}{v_g} - [k_0 - k(\omega)\cos\theta]\right\}; \quad (3)$$

ω_0 is the centre frequency of the pulse; ω is the SC radiation frequency; $k(\omega)$ describes the medium material dispersion; and $k_0 = 2\pi/\lambda_0$. Performing the integration in Eqn (2), we obtain

$$I(\theta,\omega) = \text{const} \times \left| \left(1 - \frac{h}{2}\right)\frac{L}{2} \operatorname{sinc}\Psi_0 \exp(\mathrm{i}\Psi_0) + \frac{h}{2}\frac{L}{4}\operatorname{sinc}\Psi_1 \exp(\mathrm{i}\Psi_1) + \frac{h}{2}\frac{L}{4}\operatorname{sinc}\Psi_2 \exp(\mathrm{i}\Psi_2) \right|^2, \quad (4)$$

where

$$\Psi_0 = \frac{L}{2}\left\{\frac{\omega_0 - \omega}{v_g} - [k_0 - k(\omega)\cos\theta]\right\}; \quad (5)$$

$$\Psi_1 = \frac{L}{2}\left\{\frac{\omega_0 - \omega}{v_g} - [k_0 - k(\omega)\cos\theta] + \frac{2\pi}{\Lambda}\right\}; \quad (6)$$

$$\Psi_2 = \frac{L}{2}\left\{\frac{\omega_0 - \omega}{v_g} - [k_0 - k(\omega)\cos\theta] - \frac{2\pi}{\Lambda}\right\}. \quad (7)$$

The conditions $\Psi_0 = 0$, $\Psi_1 = 0$, and $\Psi_2 = 0$ determine the angular position of the conical emission interference maxima depending on the frequency ω of the SC spectral component.

Figure 5 shows the frequency-angular spectra of the visible part of the SC in LiF measured experimentally and calculated analytically using Eqns (4)–(7) under conditions of the experiment. In the calculated spectrum, as well as in the experimentally recorded one, one can see the appearance of the conical emission rings in the 'inverse' direction, for which the spectral component with the argument Ψ_2 is responsible. Obviously, with an increase in the path length L the brightness of the rings is expected to grow, which is observed in the experiment [27], when the number of pulses increases and the transition to the waveguide regime occurs. The components of the supercontinuum with the arguments Ψ_0 and Ψ_1 lie in

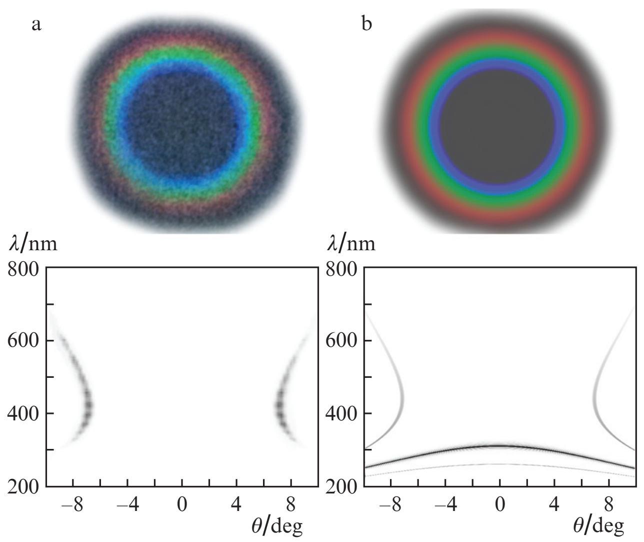

Figure 5. View (top) and spectral-angular distribution (bottom) of the SC anti-Stokes wing for the pulse at 3100 nm experimentally measured after the action of a sequence of 2000 pulses (a) and calculated analytically using the interference model (b).

the wavelength region from 200 to 300 nm and are absorbed by the induced colour centres in the case of multipulse filamentation, which explains their absence in the spectral-angular distribution, observed in the experiment (Fig. 5a).

Thus, the detected 'anomaly' in the frequency-angular spectrum of the LB conical emission in the induced CC waveguide is one more spectral manifestation of the LB 'breathing' in the process of its propagation along the filament.

6. LB path length in the induced waveguide

In the regime of multipulse filamentation in LiF, a waveguide is formed, in which the LB path length can significantly increase. In Fig. 6, one can see the appearance of a single-cycle LB at the output of the waveguide with a length up to 3 mm, induced by tens or even hundreds of pulses. However, with the growth of the number of pulses the probability of this event falls, and after 500 pulses the LB does not always appear (no statistical studies have been carried out). The most important conclusion is the fact that in all studied waveguide regimes the path length of the light bullet did not exceed 3 mm.

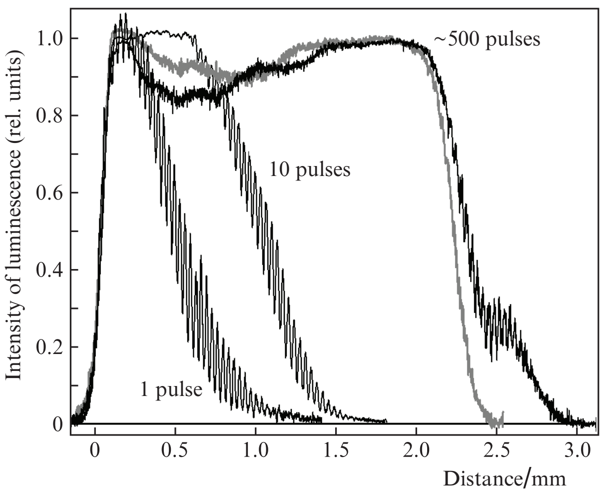

Figure 6. Luminescence profiles of CCs induced in LiF under filamentation of femtosecond pulses at a wavelength of 3100 nm after the passage of 1, 10, and 500 pulses.

7. Conclusions

The experiments using the laser coloration method allowed the measurement of the filament length in the transparent dielectric, arising under the effect of a single femtosecond pulse. It is shown that this length is essentially smaller than that observed in earlier papers, where the measurements were executed with the necessity of accumulating the data from many hundreds and thousands of pulses with fluctuating energy. It is found that the length of the filament formed in the single-pulse regime increases with increasing wavelength from a few tens of micrometres at 800 nm to hundreds of micrometres at 3900 nm. Starting from 2600 nm the luminescence profile of induced long-lived colour centres acquires a periodic structure. This structure is an evidence of the formation of a single-cycle light bullet at the wavelengths in the region of strong anomalous group velocity dispersion. Thus, for the first time we recorded an LB with a duration of about one cycle of the light field and a diameter less than 10 μm and measured the bullet path length, which did not exceed 0.5 mm for the single-pulse regime and 2.7 mm in a waveguide arising during multipulse filamentation. We observed the appearance of side maxima near the excitation wavelength, when an LB appeared in the femtosecond filament. For the first time we recorded the appearance of visible spectral components in the supercontinuum radiation, whose angular divergence increases with increasing wavelength. As follows from the interference model, the appearance of side bands and the inversion of the angular divergence of conical emission arise due to the periodic modulation of the light field in the process of single-cycle LB propagation, related to the difference between its phase and group velocity. The appearance of side maxima near the excitation wavelength at this modulation can be used to detect single-cycle LBs in different dielectrics at femtosecond filamentation under condition of anomalous group velocity dispersion.

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