Regular ‘breathing’ of a near-single-cycle light bullet in mid-IR filament

S V Chekalin\textsuperscript{1}, V O Kompanets\textsuperscript{1}, A V Kuznetsov\textsuperscript{2}, A E Dormidonov\textsuperscript{3} and V P Kandidov\textsuperscript{3}

\textsuperscript{1} Institute of Spectroscopy RAS, 108840, Moscow, Troitsk, Russia
\textsuperscript{2} Irkutsk Branch of Institute of Laser Physics SB RAS, 664033, Irkutsk, Russia
\textsuperscript{3} Moscow Lomonosov State University, Physics Department and International Laser Center, 119991, Moscow, Russia

E-mail: chekalin@isan.troitsk.ru and dormidonov@gmail.com

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Abstract
Experimental and numerical studies of a temporal evolution of a light bullet formed in isotropic LiF by mid-IR femtosecond pulse (2600–3350 nm) of power, slightly exceeding the critical power for self-focusing, are presented. For the first time regular oscillations of the light bullet field peak amplitude during its propagation in a filament were registered by investigation of induced color centers in LiF. It was revealed that color centers in a single laser pulse filament have the strictly periodic structure with a length of separate sections about 30 $\mu$m, which increases with a laser pulse wavelength decreasing. It was shown that the origin of a light bullet modulation is a periodical change of the light field amplitude of a near single-cycle wave packet in a filament, due to a difference of the wave packet group velocity and the carrier wave phase velocity.

Keywords: light bullet, filamentation, near single-cycle wave packet, color centers

1. Introduction
One of the most interesting of filamentation regimes is a process of a ‘light bullet’ (LB) formation, when simultaneous self-focusing of the beam and self-compression of a laser pulse occur. The conception of the LB formation due to the nonlinear optical self-action of a laser pulse in a dispersive medium with a cubic nonlinearity was firstly formulated by Silberberg [1] from an analysis of the quasi-optical equation in the aberrationless paraxial approximation. A promising approach for the pulse self-compression is to use the anomalous group velocity dispersion (GVD) in the presence of positive self-phase modulation (SPM) that could cause the formation of an extremely compressed wave packet. In the case of anomalous GVD the light field is contracted to the central temporal slices of the pulse from its edges [2]. Pulse compression in time and space due to the Kerr nonlinearity, self-phase modulation at anomalous GVD, self-focusing, self-steepening, and defocusing of the pulse tail in laser-induced plasma creates a LB in a filament [3–5]. In the case of 1800 nm pulse filamentation in fused silica the peak intensity in a LB achieves the value about $10^{14}$ W cm$^{-2}$, its duration becomes less than two oscillation cycles of the light field for the central wavelength, a temporal profile being considerably different from the Gaussian. The evolution of a 1800 nm LB temporal-spatial structure in sapphire was investigated in [6]. In [7] a long glowing channel of a 1900 nm pulse filament is considered as a long-lived quasi-soliton, which does not change its shape over the distance of several centimeters. The short-lived LBs sequence formation has been confirmed numerically and experimentally by autocorrelation measurements in [4, 5, 8, 9]. LBs are robust structures and the formation of each LB in a sequence is accompanied by the outburst of a certain portion of SC energy in the visible range [10, 11].

An important and still poorly investigated aspect of filamentation is related to the compressed wave packet carrier envelope phase (CEP) induced effects along the few-cycle pulses propagation in a transparent medium [12–14].
In this letter we present the results of experimental and numerical studies of a temporal and spatial structure of a single LB formed in isotropic material LiF by a mid-IR (2600–3350 nm) femtosecond pulse of power slightly exceeding the critical power for self-focusing. For the first time a periodical change of LB field peak amplitude due to CEP effect during LB propagation in isotropic crystal were recorded. We used the laser coloration method that gives a unique possibility to restore the filament spatial structure after exposure [15]. This experimental method, based on microscopic observation of laser-induced color centers (CC), allowed to register the strictly periodic structure with a length of separate sections about 30 μm in the ~1 mm length filament created by a single laser pulse. Numerical simulation showed that the origin of the recorded periodic structure is the light field amplitude modulation of a near-single-cycle LB in a mid-IR filament.

2. Experimental setup

The pulses at wavelengths of 2600–3350 nm, corresponding to the region of anomalous dispersion in lithium fluoride, were generated by a travelling-wave optical parametric amplifier of white-light continuum (TOPAS-C) with a noncollinear difference frequency generator (NDFG). The FWHM duration of nearly transform-limited 3100 nm pulses was equal to 100 fs with a spectral width 200 nm (FWHM), the pulse energy could be varied from 0.2 to 16 μJ. The laser pulses were focused inside an isotropic LiF sample of 3 cm length by a thin CaF$_2$ lens with the focal length $F = 10$ or 15 cm, the geometrical focus was located at about 3 mm from the sample input face. During a coloration process the frequency rate of laser pulses was 100 Hz and the sample was moved in the direction perpendicular to the laser beam to insure one-shot exposure of every filament. After the end of coloration a spatial distribution of laser-induced CCs luminescence along filaments was scanned by microscopes Euromex Oxion 5× or Olympus IX71 10×. For LB duration measuring pulses after propagation through the sample were directed by a parabolic mirror at the entrance of the scanning mid-IR autocorrelator.

LiF has been selected in our experiments because of significantly brighter CC luminescence intensity in comparison with other alkali halide crystals that allowed us to observe a photoinduced transformation produced by a single mid-IR exciting pulse. In the considered wavelength range the GVD parameter $k_2$ is negative and varies from $-149$ fs$^2$ mm$^{-1}$ for 2600 nm to $-345$ fs$^2$ mm$^{-1}$ for 3350 nm. The anomalous GVD is necessary for a LB formation in a filament.

3. Experimental results

The photos of CCs luminescence in filaments produced by 3100 nm laser pulses, which demonstrate strictly periodic structure of single pulse filaments, are shown in figure 1. CC luminescence excited by 450 nm CW laser was recorded by the digital camera Nikon D800.

![Figure 1](image1.png)

Figure 1. Photoimage of CC structures induced in LiF under filamentation of a single laser pulse at different wavelengths. The laser pulse parameters: (a) wavelength 3350 nm, pulse energy 13.5 μJ; (b) wavelength 3100 nm, pulse energy 11.5 μJ; (c) wavelength 2600 nm, pulse energy 10.1 μJ. The laser pulse direction—from the left to the right.

One more characteristic feature of the observed CC microstructures is their dependence on the excitation wavelength (figure 2). A processing of more than 25 CC structures profiles in every set of experiments with different excitation wavelengths resulted in a conclusion that the structure period is constant at a given wavelength. One can see that a luminescence signal modulation decreases and a period increases with the excitation wavelength decrease. The CC structures periods are equal to 29.0, 31.1, and 36.5 μm for the wavelengths of 3350, 3100, and 2600 nm, respectively. A modulation was not observed at wavelengths less than 2600 nm.

The CC structure, induced in a filament, does not depend on the laser pulse intensity—it is period is the same in filaments, observed under the excitation by 3100 nm laser pulses with energy varying from 8.5 μJ (threshold of CCs formation) to 16.5 μJ. It also does not depend on focusing conditions and on the orientation of the sample with reference to the polarization direction of exciting laser beam.

![Figure 2](image2.png)

Figure 2. Luminescence intensity of CC structures (in arbitrary units) induced in LiF under filamentation of a single laser pulse at different wavelengths. The laser pulse parameters: (a) wavelength 3350 nm, pulse energy 13.5 μJ; (b) wavelength 3100 nm, pulse energy 11.5 μJ; (c) wavelength 2600 nm, pulse energy 10.1 μJ. The laser pulse direction—from the left to the right.

Average measured values of diameter (FWHM) of the structures induced by the laser pulses at 3350, 3100, and...
2600 nm are about 3.2, 3.0, and 1.9 μm, respectively, that is comparable to the wavelengths of the excitation pulses.

The measured autocorrelation functions (ACFs) point to the fact of a LB formation in process of filamentation in LiF: ACFs are shown in figure 3 for 2600 nm input pulse (without the sample) and for the paraxial part of the pulse separated by a diaphragm, mounted at the output face of the 1 cm sample. The energy fraction passed through the 50 μm pinhole decreased until 10% from the measured ACF, is about 38 fs that is about three oscillations in LB wave packet. With increasing the pulse energy till 12.7 μJ the LB formed inside the sample and delocalized after propagation to the output face. That is why the energy fraction passed through the diaphragm decreased until 10%. This behavior agrees with that observed in [5] for fused silica. Accordingly to [5] a LB shape differs significantly from the Gaussian. On account of this, the duration of a LB, estimated from the measured ACF, is about 26 fs that is about three oscillations for the selecting aperture 50 μm in diameter. But in this case we measured not only the duration of the LB but also essentially longer duration of a background surrounding the LB. The sensitivity of our autocorrelator was not sufficient to take a proper measurement with a lesser diaphragm aperture.

LB size can be estimated taking into account measured diameters of CC (see above). The energy gap between the strongly bound singlet exciton state and the valence band in LiF (12.8 eV [16]) is equal to the energy of more than 34, 32, and 26 mid-IR laser quanta for the wavelengths 3350, 3100, and 2600 nm, respectively. The energy of excitonic band is just equal to integer number of quantum energy for this wavelength so this fact evidences in favor of decisive contribution of direct exciton generation under multiphoton absorption [17]. This allow us to estimate the LB diameter from CC structure transverse size. Under assumption of K-photon absorption a LB with Gaussian beam profile generates excitons in an area the transverse size of which is in √K times lesser than LB diameter. From this estimation it follows that for wavelengths 3350, 3100, and 2600 nm the LB FWHM diameters are equal to 18.6, 17.0, and 9.6 μm, respectively.

4. Numerical simulation and discussion

Numerical simulations clearly illustrate the scenario of LBs field dynamics, which results in appearing of the observed CC density modulation. Our simulations were performed in the slowly-evolving-wave approximation (SEWA) that allows to accurately describe a wave packet propagation down to the single cycle regime [18]. In the pulse local frame τ = t − zνg (νg—pulse group velocity) a nonlinear envelope equation (NEE) [19] for the complex envelope amplitude A(r, z, τ) takes the form:

\[
\frac{\partial A}{\partial z} = -\frac{i}{2k_0} \hat{p}^{-1} \left[ \Delta A + \int_{-\infty}^{\infty} T \hat{A} e^{i\psi} d\Omega \right] - \frac{i\omega_0}{c_0} \hat{T} [\Delta n_A] + \hat{p}^{-1} [\Delta n_\psi A] - \frac{1}{2} \hat{r}^{-2} [\sigma N A] - \frac{1}{2} \alpha A.
\]

(1)

Here \(\hat{A}(r, z, \Omega)\) is the Fourier transform of \(A(r, z, \tau)\), \(\hat{T} = \hat{T} - \hat{U} (\partial \psi / \partial \tau)\) represents the linear dispersion function of the medium, \(\psi\) is the pulse carrier frequency, \(\Omega = \omega - \omega_0\). The frequency dependence of wave vector \(k(\omega)\) is determined by the Sellmier equation for LiF. The operator \(\hat{T} = 1 - \hat{U} (\partial \psi / \partial \tau)\) is responsible for the pulse self-steepening, \(\Delta n_\psi, \Delta n_A\) are increments of refraction index caused by Kerr and plasma nonlinearity, \(\sigma\) is the cross section for inverse Bremsstrahlung, and \(\alpha\) is the coefficient of photoionization losses. The photoionization rate is calculated from Keldysh formalism [20]. SEWA usability condition, according to which the difference between the phase \(\psi_{ph}\) and group \(\nu_g\) velocity must be small, holds in our simulations, because in LiF for considering mid-IR femtosecond pulses \(\psi_{ph} \sim \nu_g \sim 0.03 \nu_{ph}\), where \(\nu_{ph} = c_0 / n t_0 \lambda_0\).

The initial laser pulse parameters in simulations are close to the experimental values: pulse duration—100 fs (FWHM), energy—18.0, 15.5, and 10.5 μJ for pulses at 3350, 3100, and 2600 nm, respectively, that corresponds to the pulse peak power about 1.5 Pcr, where \(P_{cr}\) is the critical power for self-focusing.

The light field \(E(r, z, \tau)\) in LB wave packet can be represented as [18, 21]

\[
E(r, z, \tau) = \frac{1}{2} A \exp \left\{ i \omega_0 \tau + i \omega_\psi \left( \frac{1}{\nu_g} - \frac{1}{\nu_{ph}} \right) \right\} + c.c.,
\]

(2)

where \(A = |A(r, z, \tau)| \exp \{i \xi (r, z, \tau)\}\) is calculated pulse complex envelope. The equation (2) describes the evolution of the light field in the wave packet under the change of the envelope amplitude \(|A(r, z, \tau)|\), the envelope phase \(\xi (r, z, \tau)\), and the phase \(\varphi (z) = \omega_0 t_0 / c_0 \left( \frac{1}{\nu_g} - \frac{1}{\nu_{ph}} \right)\).

The envelope phase \(\xi (r, z, \tau)\) determines a spatiotemporal phase shift of the light field oscillations in the wave packet caused by nonlinearities, whereas \(\varphi (z)\) determines a temporal shift of the light field oscillations relative to the pulse envelope maximum due to a difference between the wave packet group velocity \(\nu_g\) and phase velocity \(\nu_{ph}\) at the carrier frequency \(\omega_0\), herewith \(\xi (r, z, \tau) \ll \varphi (z)\).

Figure 4 shows the light field \(E(\tau, z)\) of the wave packet with center wavelength 3100 nm at some characteristic distances. At the distance \(z = 7.26 \text{ mm} (\text{figure 4(a)})\) corresponding to
the initial stage of LB formation the pulse envelope contains several cycles of the carrier wave and the peak amplitude of the electric field increases about in five times. At the distance \( z \geq 7.56 \text{mm} \) (figures 4(b)–(d)) the pulse peak amplitude becomes exceed the value 10 \( A_0 \) (\( A_0 \) — initial peak amplitude) that is enough for the medium multiphoton and avalanche ionization and excitons generation [17]. In the anomalous GVD regime the light field intensity increases due to both spatial self-focusing and temporal pulse self-compression that leads to the formation of an extremely compressed LB [1, 10, 11].

The LB duration becomes about 10 fs (FWHM) that is about 10% of the initial pulse duration. So, the formed LB is a wave packet comprising near-single light field oscillation. LB propagates in the medium with a group velocity at distances of several hundred microns.

Due to the difference \( \Delta \nu = \nu_{ph} - \nu_{\nu} \) the carrier wave moves faster than the LB wave packet envelope. So, if at \( z_1 = 7.559 \text{mm} \) (figure 4(b)) a maximum of the carrier wave coincides with the maximum of the pulse envelope, at \( z_2 = 7.579 \text{mm} \) (figure 4(c)) the carrier wave shifts at quarter period and the resulting peak amplitude of the electric field in LB decreases by more than 20%. At \( z_3 = 7.596 \text{mm} \) (figure 4(d)) the pulse envelop maximum matches a minimum of the carrier wave and the electric field amplitude in the LB becomes maximal again. By this way under LB propagation as a near single-cycle wave packet, the light field amplitude ‘breathes’ due to the difference in the wave packet group velocity and the phase velocity of the carrier wave. These oscillations of electric field peak amplitude proceed throughout all the LB’s existence (about 500 \( \mu \text{m} \) as can be seen from experimental data of figure 2).

Relatively small oscillations of the light field peak amplitude result in a considerable oscillation of CC density due to a high order of direct multiphoton process of CC creation under mid-IR filamentation in LiF. This regular ‘breathing’ of the light field in LB due to change of \( \varphi(z) \) during a propagation in the medium results in CC periodic structure arising. Physical reason for the CC density modulation is a cyclic transformation of the light field in a near-single-cycle mid-IR LB travelling in LiF, caused by a periodical change of the phase shift between the carrier wave and the wave packet envelope due to a difference in the LB group and phase velocities.

The period of peak amplitude oscillations in LB of 3100 nm pulse obtained from the numerical simulation is equal to \( \Delta z = \frac{\lambda}{2n - 1} = 36 \pm 0.5 \mu \text{m} \). For the pulse at 3350 nm the LB breathing period \( \Delta z \) decreases to 34 ± 0.5 \( \mu \text{m} \) and for 2600 nm pulse \( \Delta z \) increases to 42 ± 1 \( \mu \text{m} \). These results qualitatively match the experimentally measured period of CC structure in LiF formed in mid-IR filament. The small quantitative difference is due to not exact fitting of LiF nonlinear parameters in the numerical model.

Numerical simulation shows also the light field \( E(r, \tau) \) distortions in the LB (figure 4). Along with a linear phase shift of the carrier wave relative to the LB envelope, which is described by the dispersion phase \( \varphi(z) \), the light field distortions arise due to spatio-temporal shift of the nonlinear phase \( \xi(r, \tau) \). Before the LB formation (figure 4(a)) the light field amplitude maximum is located on the axis of the wave packet that is in accordance with the Kerr phase incursion in the absence of laser plasma. Growing of the LB light field amplitude causes the ionization of the medium. Therefore, after the formation of the LB the maximum of the light field oscillations amplitude at the LB tail \( (\tau > 0) \) is shifted along \( r \) relatively to the pulse axis due to contribution in \( \xi(r, \tau) \) of defocusing plasma non-linearity, which is induced by LB leading front. We estimated the LB diameter from a radial width of the larger maximum of the light field oscillations. For the 3100 nm LB the diameter is about 14 \( \mu \text{m} \). This value is underestimated because it does not account the contribution on CC generation of the LB tail part with larger transverse size (figures 4(b)–(d)). Nevertheless, it is close to the experimental value 17 \( \mu \text{m} \), given above.

CC structures revealed in our experiments have a quite another origin than those reported beforehand under investigations in different media. The observed dependence of these structures on the excitation wavelength variation is opposite to a periodic filamentation in birefringent crystals, which is caused by the periodic change in the polarization of the pulse travelling in birefringent medium in combination with the cross-sectional difference in multiphoton absorption for the linear and circular polarizations [22]. A period of a single filament structure in these experiments increased with the excitation pulse wavelength increasing in contrast to our measurements for isotropic LiF crystal. In the filamentation regime close to self-guiding at normal GVD, which was investigated numerically in [23] for condensed matter and in [24] for air, the period of the light field oscillations caused by manifold refocusing decreases with decreasing of the laser pulse power in respect to \( P_\text{cr} \). In quasi-periodical LBs sequence that formed in process of filamentation at anomalous GVD the interval between the bullets

**Figure 4.** 3D pictures of the light field \( E(r, z, \tau) \) and its envelope \( \pm |A(r = 0, z, \tau)| \) on the axis of LB at several distances: (a) \( z_1 = 7.260 \) mm—initial stage of the LB formation; (b) \( z_2 = 7.559 \) mm—single-cycle LB envelope maximum matches a maximum of carrier wave; (c) \( z_3 = 7.578 \) mm—single-cycle LB envelope maximum matches zero of carrier wave; (d) \( z_4 = 7.596 \) mm—single-cycle LB envelope maximum matches a minimum of carrier wave. The laser (3100 nm) pulse direction—from the left to the right.
femtosecond pulse power was only slightly more than $CC$, period recorded in the present experiments where initial $\rho$ is not strictly periodic, and its value essentially exceeds the $L$ Laser Phys. Lett. 13 2016 065401 spectrum during femtosecond pulse filamentation under conditions of anomalous group-velocity dispersion in fused silica Quantum Electron. 43 326

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5. Conclusion

In conclusion, a regular ‘breathing’ of the light field in near-single-cycle LB propagating in isotropic LiF was firstly revealed by experimental observation and numerical investigation of strictly periodic CC structures created due to CEP effect during the interaction between LB and bulk solid materials in process of mid-IR filamentation. The length of these structures separate sections is about 30 μm and increases with laser pulse wavelength decreasing. It is shown that LB ‘breathing’ is a cyclic transformation of the light field amplitude caused by the phase shift between the carrier wave and the near-single-cycle wave packet envelope.

Impact of the CEP effect on nonlinear-optical interaction of a single-cycle laser pulse with media at characteristic interaction time comparable to the period of optical oscillation was discussed in [14, 18, 21]. Tracing of periodic CC structures in LiF is a way of registration of the absolute carrier wave phase change in a single-cycle mid-IR wave packet that allows spectral and temporal scaling in attosecond physics investigations on phase sensitivity of nonlinear-optical interactions [12].

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is not strictly periodic, and its value essentially exceeds the CCs period recorded in the present experiments where initial $\rho$ only slightly more than $P_o$, that suggests the single LB regime.

The stability of a photoinduced periodic CC structure confirms LB robustness, which is the result of intense light field self-organization under its nonlinear-optical interaction with medium in anomalous GVD condition [11].