Letter

Carrier-envelope phase effect on light bullet dynamics

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Abstract
A light bullet (LB) is a wave packet of a few optical cycles that is extremely compressed in space and time, which is formed in the bulk transparent dielectric during femtosecond filamentation under anomalous group velocity dispersion. We demonstrate for the first time that the carrier-envelope phase shift during propagation of a near single-cycle LB causes synchronous oscillations of the spatial, temporal and energy parameters of its core with the period decreasing with increasing carrier wavelength. When analyzing the structure of color centers and induced plasma channels in fluorides, it was experimentally found that LB parameter oscillations lead to a periodic change in the nonlinear optical interaction with the dielectric.

Keywords: filamentation, light bullet, carrier-envelope phase

(Some figures may appear in colour only in the online journal)

1. Introduction
The development of ultrafast optics opens up new vistas in the sphere of ultrafast metrology, the characteristic time of which is determined not by the pulse envelope, but by the period of light field oscillation \cite{1, 2}. Ultrafast metrology includes the study of inner-shell electronic transitions, charge-transfer in biological molecules and upon chemical processes as well as other issues of electron-optical interaction. These events proceed on a time scale of the light oscillation period and are incredibly sensitive to absolute phase of a few-cycle light wave. Evolution of an atomic dipole moment, dynamics of light-field-induced electron tunneling, electron energy spectrum—depend not only on envelope amplitude, but also on carrier wave phase in the case of exposure to a few-cycle optical pulse \cite{3–5}. Moreover, this dependence becomes more significant with a reduction in number of optical cycles in a wave packet down to a single cycle. Filamentation of femtosecond pulses in transparent dielectric under anomalous group velocity dispersion (AGVD) condition causes compression of a wave packet both in time and in space. As a result, an intense few-cycle light bullet (LB) with high electric field localization forms \cite{6–8}. Spatiotemporal light field distribution in an LB is dictated by joint manifestation of Kerr and plasma nonlinearities and therefore is rather sophisticated, as well as frequency-angular spectrum, including high harmonics and broadband supercontinuum \cite{9–11}. One estimates the radius of high intensity LB core as two-three carrier wavelengths, its duration—as less than two optical cycles \cite{10–12}.

Parameters of an LB have been quantitatively determined in \cite{12} by analyzing the distribution of electric field strength simulated with the use of unidirectional pulse propagation equation (UPPE) \cite{13}.

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Absolute phase of the light carrier crucially affects processes accompanying femtosecond filamentation in transparent dielectric. The effect of initial carrier-envelope phase (CEP) on tunneling ionization was found in [14] by recording periodic change in intensity and phase of spectral components during filamentation of an 1800 nm pulse in fused silica. Numerical study of the influence of the initial CEP on the nonlinear distortions of the rapidly-oscillating carrier wave during filamentary compression of a pulse down to a few cycles was carried out in noble gas under normal group velocity dispersion condition [15] when a pulse compression is performed by a mechanism different from the mechanism of LBs formation [16]. The ‘breathing’ of a few-cycle LB, caused by CEP, was recorded in [17] with laser coloration method. In the pulse with a stable CEP when the power increases two LBs occurs which propagate with different group velocities but hold CEP stable [18].

Nevertheless, the influence of CEP on the spatiotemporal and energetic parameters of LB, which determine the efficiency of its nonlinear optical interaction with a medium, has not been investigated yet.

In this paper we show that CEP shift during propagation of a near single-cycle LB formed in Gaussian pulse in transparent dielectric at AGVD causes synchronous strictly periodic oscillations of the radius, duration and energy of its core. The dependence of oscillation period of an LB core parameters at the carrier wavelength is investigated numerically, on the basis of analysis of electric field strength obtained by solution of unidirectional equation of LB propagation in LiF, CaF$_2$, BaF$_2$. The periodic structures induced by LB due to strictly periodic oscillations of its parameters during LB propagation in transparent dielectrics has been experimentally observed as modulation of long lived color centers (CCs) density in LiF and electron concentration of plasma channels in LiF, CaF$_2$, BaF$_2$. By this way a periodic change in the efficiency of LBs nonlinear-optical interaction with the dielectric has been demonstrated. The analytical estimate written for a Gaussian pulse with a harmonic carrier wave can be used to find out the LB oscillation period as a function of wavelength.

2. Numerical investigations

2.1. Methods

We used UPPE [13] and computer code [19] in numerical investigation of LB formation and propagation in LiF, CaF$_2$ and BaF$_2$. Equation for spectral component of the electric field strength $\tilde{E}(\omega, k_r, z)$ in the nonlinear dispersive medium has the following form:

$$\frac{\partial \tilde{E}(\omega, k_r, z)}{\partial t} = i k_z(\omega) \tilde{E}(\omega, k_r, z) + \frac{\omega}{2k_z(\omega)c_0^2} \left[i\omega P_{nl} - J_a - J_f \right],$$

(1)

where $P_{nl}$ is the nonlinear polarization, $J_a$ and $J_f$ describes the current of multiphoton absorption and the current of free electrons, respectively. Here, $k_z^2(\omega) = \omega^2 n^2(\omega)/c_0^2 - k^2$, $n(\omega)$ is the dispersion of the dielectric described by the Sellmeier formula; $c_0$ is the speed of light in vacuum, $\varepsilon_0 = 8.85 \times 10^{-12}$ F m$^{-1}$ is vacuum permittivity.

Equation (1) describes pulse propagation under diffraction and dispersion, changes in the refractive index of the medium caused by Kerr and plasma nonlinearities, losses by Bremsstrahlung and photoionization [12, 13].

Electron concentration in laser plasma $N_e$ is dictated by the process of field ionization at a rate of $W_f(|E|^2)$, taking into account both multiphoton and tunneling effects [20], as well as process of avalanche ionization at a rate $W_A$ due to inelastic collisions of electrons with neutral atoms, and is described by the equation:

$$\frac{\partial N_e}{\partial t} = W_f \left(|E|^2\right) \left(1 - \frac{N_e}{N_0}\right) + W_A N_e,$$

(2)

where $N_0$ is the neutral atom density.

The bandgap $U_i$, which determines the $W_f(\eta^2)$, is taken equal to $U_i = 13.6$ eV in LiF, $U_i = 10.0$ eV in CaF$_2$ and $U_i = 9.1$ eV in BaF$_2$ [21]. The frequency of an inelastic collisions of electrons with neutral atoms is $\sim 10^{15}$ s$^{-1}$ for condensed media. A term taking into account the recombination of free electrons with the characteristic times by several orders of magnitude exceeding the duration of the femtosecond pulse is absent in equation (2).

In order to study formation and dynamics of an LB during filamentation in fluorides we considered bandwidth-limited collimated Gaussian wave packet

$$E(r, t, z = 0) = E_0 \exp\left(-\frac{r^2}{2r_0^2} - \frac{t^2}{2t_0^2}\right) \cos(\omega_0 t),$$

(3)

where $E(r, t, z)$ is an electric field strength, $r_0$, $2t_0$—radius and duration of a wave packet at $e^{-1}$ level of squared field strength; $\omega_0 = 2\pi c_0/\lambda_0$—frequency at carrier wavelength $\lambda_0$. Initial duration of a multicycle wave packet at the wavelength varying in the range $\lambda_0 = 2400$–$5700$ nm was $2t_0 = 120$ fs, initial radius—$r_0 = 30$ μm. Peak electric field strength $E_0$ at all wavelengths corresponded to peak power $P = 1.5 P_{cr}$, where $P_{cr}(\lambda_0)$—critical power for stationary self-focusing.

2.2. Formation of an LB with a high-field core

Propagation of a wave packet in a bulk Kerr medium under AGVD condition initiates a compression of the wave packet in space and in time due to self-phase modulation caused by cubic nonlinearity (figure 1(a)). An amplitude of electric field strength $E(r, t, z)$ triples during LB formation, therefore, generation of laser plasma takes place. Defocusing in induced plasma gives rise to aberrational distortions when light field sharply decreases on the beam axis. Moreover, local maximum of field strength becomes displaced from the axis which signifies the formation of a ring structure at the pulse tail because of its divergence in plasma (figure 1(b)). Deformations of a wave packet temporal profile gradually come over whole cross-section plane.
Collection of a few optical cycles with high electric field amplitude is an LB (figure 1(b)). According to [12] we determine parameters of an LB by region of high field localization $S_{HF}$—core of an LB. Squared absolute value of field strength in this region satisfies a condition

$$|E(r,t,z)|^2 |_{r,t \in S_{HF}} \geq e^{-1} \max_r |E(r=0,t,z)|^2. \quad (4)$$

High-field region has a shape with time axis of symmetry. For a many cycle Gaussian wave packet the border of the core is an elliptical curve in $(r,t)$ plane. Under compression of a wave packet the high field region monotonically compresses both in space and in time. Its shape remains like initial one up to an LB formation. The core shape of already formed LB is dramatically distorted (figure 2 (top row)). Due to defocusing in induced laser plasma an $S_{HF}$ of LB extremely compresses in time domain and becomes cone-like at the trailing front. Maximum of an electric field shifts toward trailing edge of a wave packet, which testifies the decrease in its group velocity under condition of Kerr nonlinearity.

Maximum radius of the LB core determines the local radius $r_c$ of a wave packet. Temporal size of $S_{HF}$ on the beam axis ($r = 0$) shows local duration $2\tau_c$. Along with the local parameters we consider effective radius $r_{eff}$ and effective duration $2\tau_{eff}$ of a wave packet. These quantities take account of electric field distribution in LB core:

$$r_{eff} = \left( \frac{\int_{S_{HF}} r^2 |E(r,t,z)|^2 dS}{W_{HF}(z)} \right)^{\frac{1}{2}}, \quad (5a)$$

$$\tau_{eff} = \left( \frac{\int_{S_{HF}} t^2 |E(r,t,z)|^2 dS}{W_{HF}(z)} \right)^{\frac{1}{2}}, \quad (5b)$$

where $W_{HF}(z) = \int_{S_{HF}} |E(r,t,z)|^2 dS$ is an energy in a region of high field localization.

Introduced parameters are the generalization of characteristics of a quasi-harmonic wave packet to an LB consisting of a few cycles. At $z = 0$ mm local $r_c$ and effective $r_{eff}$ radii are the same as radius $r_0$ of an initial wave packet (1), local $2\tau_c$ and effective $2\tau_{eff}$ duration—the same as duration $2\tau_0$.

2.3. LB parameters oscillation

Core of already formed propagating LB periodically contracts and expands synchronously in space and in time (figure 2 (top row)). Peak value of squared field strength on the beam axis $\max_t |E(r=0,t,z)|^2$ also periodically changes with distance (figure 2 (bottom row)).

It reaches maximum when $S_{HF}$ contracts down to minimal size (figures 2(a), (d) and (c), (f)) and decreases with $S_{HF}$ expansion (figures 2(b) and (e)). Along with periodic change in shape and sizes of LB core, parameters of LB oscillate too. Compression of the core accompanies by synchronous decrease in duration, radius and energy ($W_{HF}$) of LB but by corresponding increase in peak field strength. To the contrary, with core expansion the duration, radius and energy of an LB increases, but peak field strength decreases. The result is that all core parameters: duration, radius, peak field strength and energy localized in the core periodically change with LB propagation (figure 3). Energy of LB core oscillates in phase opposition with electric field strength, but in phase with its sizes which determine spatiotemporal volume of high field region. Mean energy localized in a core of an LB is about 10% of the total wave packet energy $W_{pulse}$ (figure 3(b)).

During LB propagation in LiF all its core parameters oscillate with the same period $\Delta z$, which does not change over the path length. Any deviation of the oscillation period of any parameter from the value averaged over all temporal, spatial and energy parameters of LB is lower than 3%. With an increase in carrier wavelength $\lambda_0$ the oscillation period $\Delta z$ of LB parameters monotonically decreases (figure 4). The same dependence takes place for LB formed in CaF$_2$ and BaF$_2$.

Strongly periodic change in the parameters of the single-cycle LB core due to CEP shift on its path length is qualitatively different from nonmonotonic distribution of fluence along the filament due to multiple refocusing which is the
Figure 2. (Top row) Isolines of squared electric field strength with \(E_0^2\) interval. Core of a light bullet is a filled region, its border—dashed curve; (bottom row) squared electric field strength on the beam axis \(|E(r = 0, t, z)|^2\). Shown distributions correspond to nearest maximum and minimum values of \(\text{max}|E(r = 0, t, z)|^2\): (a), (d) \(z = 1.780\) mm; (b), (e) \(z = 1.795\) mm; (c), (f) \(z = 1.810\) mm. Pulse at \(\lambda_0 = 3350\) nm propagates from right to left in LiF.

Figure 3. Oscillations of light bullet parameters in LiF: (a) local \(r_e\) and effective \(r_{\text{eff}}\) radii, local 2\(\tau_e\) and effective 2\(\tau_{\text{eff}}\) durations normalized to the wavelength \(\lambda_0\) and the period \(T_0\) respectively; (b) squared peak strength of electric field on the beam axis \(|E_{\text{peak}}(r = 0)/E_0|^2\) and relative energy \(W_{\text{HF}}/W_{\text{pulse}}\) in the core of a light bullet. Pulse wavelength is \(\lambda_0 = 3350\) nm.

Figure 4. Spectral dependence of oscillation period \(\Delta z\) of light bullet parameters obtained in computer simulation (empty markers), analytically (6) (solid line), experimentally by measuring color centers density in LiF and electron concentration in BaF\(_2\) and CaF\(_2\) (filled markers). Experimental errors are in all cases not more than the size of corresponding symbol.

Figure 5. Sine- and cosine-shaped electric waves on beam axis: (a) in a light bullet at (black line) \(z = 1.795\) mm, (red line) \(z = 1.810\) mm, (b) in a wave packet similar to a Gaussian one at (black line) \(z = 1.150\) mm, (red line) \(z = 1.165\) mm; (dotted lines) LB envelope approximation. The 3350 nm pulse propagates in LiF from right to left.

For the physical interpretation of strictly periodic change in the parameters of a few-cycle LB let us extend to it a concept of CEP of a few-cycle pulse. According to [1], CEP shift in a propagating Gaussian few-cycle pulse with harmonic carrier wave causes a periodic change in resulting wave amplitude. Spatiotemporal distribution of a field strength in an LB qualitatively stands out from corresponding distribution in a Gaussian pulse or wave packet at the start of compression. LB spectrum is broadband and together with carrier frequency there are high harmonics and high-frequency components of supercontinuum. The notion of the pulse envelope is formally inapplicable for a few-cycle LB. We created this envelope using \(\text{max}|E(r = 0, t, z)|^2\) (figure 5). It allowed us to describe axial light field strength in the LB at two close distances as sine- and cosine-shaped waveforms (figure 5(a)). Thus, the light field on the axis of 3350 nm LB has a sine-shaped waveform at \(z_1 = 1.795\) mm and cosine-shaped waveform at \(z_2 = z_1 + \Delta z/2\). These waveforms correspond to 90\(^\circ\) CEP shift upon LB propagation over a distance equal to half oscillation period \(\Delta z/2 = 15\) \(\mu\)m. It should be noted that at the beginning of wave packet compression during filamentation its shape is similar to many cycle Gaussian one. In this case there is a negligible CEP impact on the resulting amplitude of a light field (figure 5(b)).
For a pulse with the envelope containing a few cycles of harmonic carrier wave, there is an estimate from [1] of the period $\Delta z$ of the resulting field amplitude oscillation [15, 23]:

$$\Delta z(\lambda_0) = \frac{\lambda_0 V_{gr}(\lambda_0)}{2n(\lambda_0)(V_{ph}(\lambda_0) - V_{gr}(\lambda_0))},$$

(6)

where $V_{ph}(\lambda_0)$, $V_{gr}(\lambda_0)$—tabulated data on phase and group velocity for a wavelength $\lambda_0$. The analytical dependence (equation (6)) is in a good agreement with the simulated periods of oscillations of LBs formed in LiF, BaF$_2$, CaF$_2$ (figure 4) despite a key distinction between an LB and a Gaussian few-cycle pulse. Thus, one can use a simple estimate of period (equation (6)) to calculate the oscillation period for parameters of an LB with complex spatiotemporal distribution of a light field and with a broad frequency-angular spectrum. The deviation of $\Delta z(\lambda_0)$ caused by a decrease in the LB group velocity $V_{gr}(\lambda_0)$ due to the Kerr nonlinearity does not exceed 10% for considered wavelengths.

The oscillations of the LB parameters lead to the periodic character of its nonlinear interaction with the dielectric. Figure 6 shows numerically calculated plasma channels formed by the LB in BaF$_2$ at $\lambda_0 = 4700$ nm and in LiF at $\lambda_0 = 3500$ nm.

The obtained distributions of free electron concentration in plasma channel have a periodic structure corresponding to an oscillating change in the LB electric field strength. The period of change in an electron concentration is $\Delta z = 90 \mu m$ in BaF$_2$ in figure 6(a) and $\Delta z = 31 \mu m$ in LiF in figure 6(b), which corresponds to oscillation periods shown for the same conditions in figure 4 with empty markers.

3. Experimental investigations

In present work the effect of oscillation of LB parameters on its nonlinear interaction was for the first time studied experimentally both by laser coloration method and by detection of plasma channels glowing that allowed us to investigate different dielectrics.

The Mid IR femtosecond pulses at wavelength $\lambda_0$, tunable in the range from 2400 to 5700 nm, which lies in the AGVD region for dielectrics under consideration were generated by the travelling-wave optical parametric amplifier of superfluorescence with the noncollinear difference frequency generator. TM polarized 130 fs mid IR pulses with energy of about 30 $\mu J$ were focused by silver-coated concave mirror with a focal length $f = 200$ mm inside a LiF, CaF$_2$ and BaF$_2$ sample at a distance of several millimeters from its input face. Propagation of a laser pulse in the medium under AGVD condition initiates a concerted compression of light field in space and in time due to self-phase modulation caused by cubic nonlinearity up to formation of a single-cycle LB. In our experiment, nonlinear action of formed oscillating single-cycle LBs on transparent dielectrics LiF, CaF$_2$ and BaF$_2$ was recorded by two different methods in a single laser pulse exposure regime, which completely eliminates measurement errors caused by the shot-to-shot irreproducibility of laser pulse parameters. That is why possible fluctuations of CEP of our laser were not important. To implement a single-pulse exposure regime the sample was displaced in the direction perpendicular to the laser beam after each shot.

The first one is the laser coloration method [17, 23, 24] based on the well-known effects of staining of LiF crystals as a result of formation of long-lived structures of luminescent CCs to visualize the trace of a laser beam [25]. To analyze the spatial distribution of the luminescence intensity of the recorded CCs filamentary structure that reproduced the density of the laser-induced electronic excitations in the filament we used a monochrome 8Mp Charge Coupled Device (CCD) camera 8051 M (Thorlabs) with $10 \times$ NA 0.3 objective with illumination of the induced CCs at the absorption wavelength by a CW laser radiation at 450 nm and detection of luminescence for long time after the writing of these structures. The scattered pump radiation was cut off by an auxiliary yellow-green filter. Some samples of spatial distribution of CCs density in LiF recorded by this way in LiF are shown in figures 7(a) and (b).

In the second method a structure of plasma channels induced by LB in LiF, CaF$_2$ and BaF$_2$ was registered online by its self-luminescence straight during LB propagation. These experiments were performed with the use of the same camera with a LOMO $8 \times$ NA 0.2 objective that provided 1.5 $\mu m$ resolution for a 500 nm plasma luminescence signal. To implement a single laser pulse exposure regime the camera exposure time was set at 0.9 ns. Typical traces of electron concentration spatial distribution along the plasma channel induced by LB obtained by this way in CaF$_2$ are shown in figures 7(c) and (d).

It should be noted that in both cases the length of CCs structures and plasma channels is 300–500 $\mu m$, which corresponds to LB path length where light field localization is
high [17]. Moreover the observed traces of CCs structures and plasma channels clearly demonstrate periodic modulation of signal intensity along the track. It is important that in LiF the oscillation period obtained from CCs structures measurement is equal to one recorded from plasma channels at the same wavelength. In both cases periodic variations of the signal intensity from microstructures induced by multiphoton processes in all dielectrics give evidence of regular change in LB parameters because of the CEP. This is due to the compression of the infrared pulse into a bullet with a duration close to the period of optical oscillations [8, 17, 23], i.e. indicates the formation of a single-cycle LB. Experimentally measured spatial period of this modulation oscillation \( \Delta z \), caused by CEP decreases with increasing carrier wavelength in a good accordance with calculations (figure 4). It should be noted that all experimental symbols in figure 4 are in the existence domain of near single-cycle LB. Data for BaF\(_2\), CaF\(_2\) are more scarce than those for LiF due to less domain for short wavelengths and larger filamentation threshold for long wavelengths region.

Results of our experiments show that nonlinear-optical interaction of an LB with dielectric is determined not only by temporal but also by spatial and energetic parameters oscillating during LB propagation.

4. Conclusion

As a result of the conducted investigation, we discovered that CEP affects all parameters of an LB core which is characterized by high localization of a light field both in space and in time. In contrast to the case of a few-cycle Gaussian pulse with a flat wave front [1], upon LB propagation, CEP causes periodic variation not only of amplitude of electric field strength but also of its spatiotemporal localization. We found out that the estimate of oscillation period in dependence to wavelength obtained for a Gaussian pulse with harmonic carrier is valid for an LB with complex spatiotemporal field distribution and broadband frequency-angular spectrum. Through the analysis of CCs density in LiF and electron concentration in LiF, CaF\(_2\), BaF\(_2\) induced by an LB, we revealed that oscillation of LB parameters (caused by CEP) leads to periodic modulation of nonlinear-optical interaction of a light bullet with dielectric. Considering oscillations of spatial and energetic parameters of an LB is significant for quantitative analysis of LB nonlinear influence upon medium.

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