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# Material dispersion effect on the oscillations of a single-cycle wave packet

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Analytically, numerically and experimentally the effect of the material dispersion of the dielectric on the oscillation period of the electric field in a single cycle wave packet (a light bullet) is investigated. Wave packet oscillates due to periodic phase shift between the envelope of the pulse and its carrier wave. The influence of nonlinear shift of phase and group velocities on the analytical estimation of the oscillation period of a light bullet with a different carrier wavelength is considered.

**Keywords:** group velocity, phase velocity, material dispersion, absolute phase shift, single-cycle wave packet, filamentation, light bullet.

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The development of ultrafast metrology of electronic processes in atomic systems and biological objects is inextricably linked with the optics of bipolar and unipolar pulses, the duration of which is close to one period of optical oscillations [1–3]. The nonlinear optical action of extremely short bipolar pulses on the objects under study is determined not only by the amplitude of the light field, but also by the phase of its oscillations [4–8]. The maximum electric field strength in the cosine mode of a bipolar pulse, at which the maximum of its envelope coincides with the maximum of optical oscillations, is significantly higher than in the sine mode, at which the field turns to zero at the maximum of the envelope. The difference in the magnitude of the maximum electric field strength of the cosine and sine modes of the pulse increases with a decrease in the number of optical oscillations in it and is most significant for single-cycle pulses of the near and medium IR range, at which the atomic scale of time resolution in spectroscopy is achieved. At the same time, during the propagation of a single-cycle bipolar pulse in a medium with material dispersion, a periodic change of cosine and sine modes occurs due to a phase shift between its envelope and carrier frequency (Carrier-Envelope Phase, CEP) caused by a difference in phase and group velocities [1,2,8].

One of the possible methods for obtaining single-cycle wave packets is the formation of light bullets (LBs) during filamentation of femtosecond laser pulses in the volume of a transparent medium under conditions of anomalous group velocity dispersion (GVD) [9]. The LB is a wave packet that, during diffraction and anomalous GVD in the absence of any guiding structures, is extremely compressed in time and space due to self-phase modulation in a medium with Kerr nonlinearity [10]. The light field of the LB is localized in its core, the duration of which is one or two periods of optical oscillations, the diameter — of the order of five wavelengths [11]. The LB's core contains about 10% of the energy of the wave packet, the peak strength of the electric field corresponds to the intensity of $\sim 50\,\mathrm{TW/cm^2}$. During the LB propagation, the spatial, temporal and energy parameters of its core oscillate synchronously, which causes a periodic change in the efficiency of the nonlinear optical interaction of the LB with the medium [8,12]. In transparent dielectrics, the oscillation period of the LBs of the middle IR spectral range is several tens of micrometers and decreases with increasing carrier wavelength.

In this paper, the effect of the material dispersion of the dielectric on the period of oscillations of a single-cycle wave packet caused by the phase shift between the envelope of the pulse and the carrier frequency during propagation in the volume of the medium is investigated. The contribution of nonlinear changes in the phase and group velocities of the LBs to the change in the oscillation period of the maximum of the electric field strength modulus in the LB with different carrier wavelengths is considered.

The generally accepted concepts of a carrier wave and an envelope are generalized to wave packets containing several optical oscillations based on the analysis of the square of the electric field strength modulus, for which the time integral of the space-time distribution corresponds to the intensity of a narrow-band wave packet [11]. The envelope of the wave packet is a smooth curve connecting the maxima of the square of the modulus of the electric field strength, the carrier is a field oscillations under this curve. Then a single-cycle pulse is one whose envelope duration, determined, for example, by the level of $e^{-1}$, is equal to the period of optical oscillations.

When a single-cycle pulse propagates in a dispersing medium, the carrier wave shifts relative to the envelope due to the difference in its phase and group velocities. For the period $\Delta z$, with which the cosine mode in the pulse is restored when the carrier wave is dephased relative to the envelope, in the approximation $\frac{\lambda}{n}\frac{\partial n}{\partial\lambda}\Big|_\lambda \ll 1$ the estimation received [13]

$$\Delta z(\lambda) = \frac{1}{2}\left(\frac{\partial n}{\partial\lambda}\Big|_\lambda\right)^{-1}, \tag{1}$$

where $\lambda$ is the carrier wavelength. According to a more precise expression, the phase correction period of the carrier and the envelope of the pulse is determined by the formula [7,12]

$$\Delta z(\lambda) = \frac{\lambda V_{gr}(\lambda)V_{ph}(\lambda)}{2c_0[V_{ph}(\lambda) - V_{gr}(\lambda)]}, \tag{2}$$

where $V_{gr}(\lambda), V_{ph}(\lambda)$ — group and phase velocities at the carrier wavelength $\lambda$, $n(\lambda)$ — refractive index, $c_0$ — the velocity of light in vacuum.

It can be seen that the oscillation period is $\Delta z(\lambda)$ is determined primarily by the difference in phase and group velocities, the dependences of which on the wavelength are shown in Fig. 1, *a* in a wide spectral range covering the regions of normal, zero and anomalous GVD. Curves of the dependence of the oscillation period $\Delta z$ of the maximum modulus of the electric field strength $|E|_{max}$ on the wavelength calculated for a single-cycle pulse by the formula (2) for three dielectrics, are shown in Fig. 1, *b*. The smallest difference between the phase and group velocities is achieved at wavelengths near the zero value of the group velocity dispersion parameter ($k_2 = \partial^2 k/\partial\omega^2$), where the greatest value of the oscillation period is observed $\Delta z$. The vertical segments in Fig. 1, *a* indicate the wavelength of the zero group velocity dispersion $\lambda^{k_2=0}$: for LiF $\lambda^{k_2=0} = 1.2\,\mu$m, for $CaF_2$ — $1.5\,\mu$m, for $BaF_2$ — $1.9\,\mu$m. In areas of normal and anomalous GVD, the difference between $V_{gr}(\lambda)$ and $V_{ph}(\lambda)$ increases, and the oscillation period $\Delta z$ decreases with increasing detuning from the wavelength $\lambda^{k_2=0}$ (fig. 1, *b*). In barium fluoride $V_{gr}(\lambda)$ and $V_{ph}(\lambda)$ are closest, and the oscillation period is $\Delta z(\lambda)$ is significantly higher than in lithium and calcium fluorides.

Analytical estimation (2) of oscillation period $\Delta z(\lambda)$ is in good agreement (Fig. 1, *b*) with the results of numerical simulation of propagation of a model single-cycle Gaussian wave packet with a harmonic carrier in a dispersive medium:

$$E(r, t, z = 0) = E_0 \exp\left(-\frac{r^2}{2r_0^2} - \frac{t^2}{2\tau_0^2}\right)\cos\left(\frac{2\pi c_0}{\lambda}t\right), \tag{3}$$

where $\tau_0 = \lambda/c_0$, $r_0 \approx 10\lambda$.

The LBs are formed as a result of Kerr self-focusing of a femtosecond laser pulse in the volume of a transparent dielectric, which develops in concert with its compression over time during self-phase modulation under conditions of anomalous GVD [10]. During the formation of the LBs, the rapid increase in the intensity of the collapsing wave packet during its spatio-temporal compression is limited by the defocusing of radiation in the plasma induced by an intense light field. The large gradient of the light wave phase both in space and in time caused by the increment of the refractive index at the Kerr and plasma nonlinearities leads to the superbroadening of the frequency and angular spectra of the wave packet. According to the numerical study of the LBs [11], performed by solving the unidirectional pulse propagation equation (UPPE) [14,15], the spatio-temporal distribution of the light field and the spectrum of the formed LB are qualitatively different from the Gaussian wave packet (3). This causes difficulties in determining the envelope of the LB and the carrier wave. Nevertheless, considering the most intense spectral component in the LB as a carrier, and the curve connecting the neighboring maxima of the square of the electric field strength modulus as an envelope allows us to generalize the definitions introduced for a single-cycle wave packet to it, and introduce, in particular, the concepts of cosine and sine modes of the LBs. The maximum of the modulus of the electric field strength $|E|_{max}$ and the parameters of the LBs core oscillate due to the dispersion shift of the envelope and carrier phases during its propagation, similar to the field strength in a Gaussian pulse with a harmonic carrier. For the oscillation period $\Delta z(\lambda)$ it is possible to apply the estimate (2), written for a Gaussian pulse with a harmonic carrier in a linear dispersive medium [8]. At the same time, the deviation of the analytical estimate $\Delta z$ of the oscillation period of $|E|_{max}$ of LB obtained by (2) with tabular values $V_{gr}(\lambda), V_{ph}(\lambda)$, from the values of $\Delta z$ determined from the numerical solution of the unidirectional pulse propagation equation (UPPE), reaches 10% for the dielectrics under consideration (Fig. 2).

The LB's oscillations were experimentally investigated in a single-pulse regime, when fluctuations in the energy and absolute phase of the pulse light field do not affect the measurement results. In LiF, the oscillations of the LB's parameters were recorded by the laser coloration method [16] by change in the density of induced color centers along the filament, in $CaF_2$ — by change in the luminescence energy of the induced laser plasma. The spectral dependence of the oscillation period $\Delta z(\lambda)$ obtained in the experiment is close to the analytical estimate (Fig. 2). At the same time, the nature of the experimental dependence $\Delta z(\lambda)$ is noticeably different from the analytical one.

We investigated the influence of nonlinear changes in the group $V_{gr}(\lambda)$ and phase $V_{ph}(\lambda)$ velocities, on the oscillation period of the LB's parameters, which were determined in numerical analysis by the shift of its envelope and carrier relative to the coordinate system traveling with the group

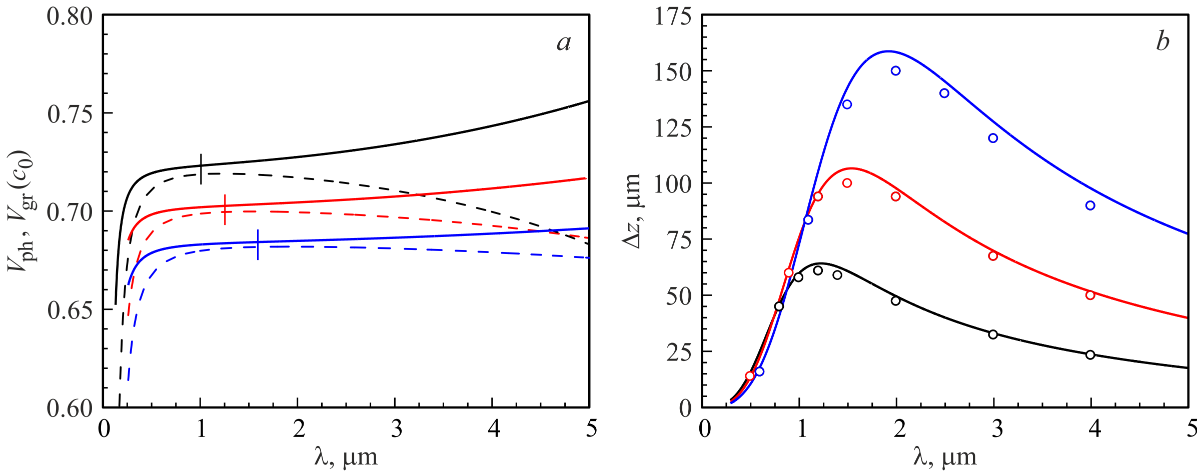


**Figure 1.** Spectral dependencies ($a$) of phase $V_{ph}(\lambda)$(solid curves) and group $V_{gr}(\lambda)$ (dashed) velocities in LiF (black curve), $CaF_2$ (red) and $BaF_2$ (blue) and ($b$) of the oscillation period $\Delta z(\lambda)$ of the maximum of the electric field strength modulus $|E|_{max}$ in a Gaussian pulse, determined by (2) (solid curves) and obtained from numerical simulation (circles).

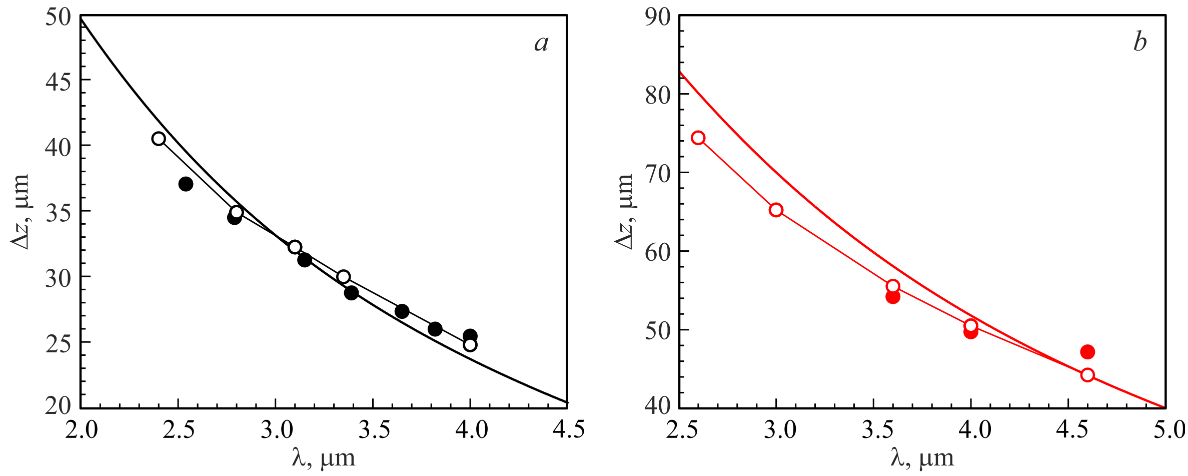


**Figure 2.** Spectral dependence of the oscillation period $\Delta z(\lambda)$ of the electric field strength modulus of LB ($a$) in LiF, ($b$) $CaF_2$, obtained experimentally $\Delta z(\lambda)$ (shaded characters), analytically $\Delta z(\lambda)$ by (2) with tabular values $V_{gr}(\lambda)$ and $V_{ph}(\lambda)$ (solid curve) and analytically by (2) taking into account the nonlinear change of group $\delta V_{gr}/V_{gr}$ and phase $\delta V_{ph}/V_{ph}$ LB velocities (empty characters connected by a curve).

velocity of the initial wave packet. As follows from the analysis of the dynamics of the wave packet, the relative decrease in the phase $\delta V_{ph}/V_{ph}$ and group $\delta V_{gr}/V_{gr}$ velocities in LiF and $CaF_2$ is small and does not exceed 0.4% in the considered wavelength range. At the same time, the magnitude of the oscillation period is sensitive to small changes in velocities:

$$\frac{\delta(\Delta z)}{\Delta z} = C(\lambda)\left[\frac{\delta V_{gr}}{V_{gr}} + \frac{\delta V_{ph}}{V_{ph}}\right]. \quad (4)$$

Here $C(\lambda)$ — sensitivity function of $\delta(\Delta z)/\Delta z$ to $\delta V_{ph}/V_{ph}$ and $\delta V_{gr}/V_{gr}$, which in the wavelength range $\lambda = 2{-}5\,\mu m$ takes values in the range from 20 to 200 for the materials under consideration, which indicates a significant influence of changes in group and phase velocities on the magnitude of the oscillation period $\Delta z(\lambda)$. As a result, a change in the period $\delta(\Delta z)/\Delta z$ associated with a nonlinear change in the phase $\delta V_{ph}/V_{ph}$ and group $\delta V_{gr}/V_{gr}$ LB velocities, reaches several percent. At the same time, the nature of the spectral dependence $\Delta z(\lambda)$, obtained analytically with corrections for nonlinear changes in phase and group velocities, is closer to that measured for LBs.

In a single-cycle wave packet and LB formed in the volume of a transparent dielectric during filamentation of a femtosecond pulse under conditions of anomalous group velocity dispersion, the maximum value of the electric field strength modulus periodically changes during propagation due to a phase shift between the envelope and carrier waves. The dependence of the oscillation period of the electric strength in the wave packet is determined by the material dispersion of the dielectric, reaching a maximum in the region of zero GVD and decreasing during detuning in the region of normal and anomalous GVD. For the period of field strength oscillations in the LB, the analytical estimate obtained for a Gaussian pulse with a harmonic carrier in a linear dispersive medium is applicable with an accuracy of 10%. The nonlinear deviation of the phase and group velocities of the LB, which is no more than 0.4%, strongly affects the oscillation period of its parameters, the value of which varies by 5–10%. The spectral dependence of the oscillation period of the LBs, obtained analytically with corrections for nonlinear changes in its phase and group velocities, coincides with the measured one.

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### Conflict of interest

The authors declare that they have no conflict of interest.

## References

[1] Th. Brabec, F. Krausz. Rev. Mod. Phys., **72** (2), 545 (2000). DOI: 10.1103/RevModPhys.72.545
[2] F. Krausz, M. Ivanov. Rev. Mod. Phys., **81** (1), 163 (2009). DOI: 10.1103/RevModPhys.81.163
[3] R.M. Arkhipov, M.V. Arkhipov, N.N. Rosanov. Quantum Electronics, **50** (9), 801 (2020). DOI: 10.1070/QEL17348.
[4] G.G. Paulus, F. Grasbon, H. Walther, P. Villoresi, M. Nisoli, S. Stagira, E. Priori, S. De Silvestri. Nature, **414** (6860), 182 (2001). DOI: 10.1038/35102520
[5] E. Goulielmakis, M. Schultze, M. Hofstetter, V.S. Yakovlev, J. Gagnon, M. Uiberacker, A.L. Aquila, E.M. Gullikson, D.T. Attwood, R. Kienberger, F. Krausz, U. Kleinberg. Science, **320** (5883), 1614 (2008). DOI: 10.1126/science.1157846
[6] M. Uiberacker, Th. Uphues, M. Schultze, A.J. Verhoef, V. Yakovlev, M.F. Kling, J. Rauschenberger, N.M. Kabachnik, H. Schröder, M. Lezius, K.L. Kompa, H.-G. Muller, M.J.J. Vrakking, S. Hendel, U. Kleineberg, U. Heinzmann, M. Drescher, F. Krausz. Nature, **446**, 627 (2007). DOI: 10.1038/nature05648.
[7] L. Bergé, C.-L. Soulez, C. Köhler, S. Skupin. Appl. Phys. B, **103**, 563 (2011). DOI: 10.1007/s00340-011-4513-4
[8] E.D. Zaloznaya, V.O. Kompanets, A.D. Savvin, A.E. Dormidonov, S.V. Chekalin, V.P. Kandidov. Laser Phys. Lett., **19**, 075402 (2022). DOI: 10.1088/1612-202X/ac7134
[9] Y. Silberberg. Opt. Lett., **15** (22), 1282 (1990). DOI: 10.1364/OL.15.001282
[10] V.P. Kandidov, E.D. Zaloznaya, A.E. Dormidonov, V.O. Kompanets, S.V. Chekalin. Quantum Electronics, **52** (3), 233 (2022). DOI: 10.1070/QEL18000.
[11] E.D. Zaloznaya, A.E. Dormidonov, V.O. Kompanets, S.V. Chekalin, V.P. Kandidov. JETP Lett., **113** (12), 787 (2021). DOI: 10.1134/S0021364021120146.
[12] A.V. Kuznetsov, V.O. Kompanets, A.E. Dormidonov, S.V. Chekalin, S.A. Shlenov, V.P. Kandidov. Quantum Electronics, **46** (4), 379 (2016). DOI: 10.1070/QEL16038].
[13] L. Xu, Ch. Spielmann, A. Poppe, T. Brabec, F. Krausz, T.W. Hänsch. Opt. Lett., **21** (24), 2008 (1996). DOI: 10.1364/OL.21.002008
[14] M. Kolesik, J.V. Moloney. Phys. Rev. E, **70** (036604), (2004). DOI: 10.1103/PhysRevE.70.036604
[15] V.Yu. Fedorov, M. Chanal, D. Grojo, S. Tzortzakis. Phys. Rev. Lett., **117** (043902), (2016). DOI: 10.1103/PhysRevLett.117.043902
[16] S.V. Chekalin, V.O. Kompanets. Opt. Spectrosc., **127**, 88 (2019). DOI: 10.1134/S0030400X19070051.