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Role of Multiphoton Ionization in the Short-Wavelength Broadening of the Spectrum of a Light Bullet in the Middle Infrared Range

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The short-wavelength cutoff of the anti-Stokes wing of the supercontinuum in the spectrum of a light bullet at filamentation of femtosecond radiation at a wavelength tunable in a wide range of the middle infrared range in fused silica and fluorides has been studied for the first time. A physical model for the formation of the spectrum of the light bullet has been proposed on the basis of the spectral broadening caused by the phase self-modulation of the light field on the trailing edge of the light bullet at the generation of a plasma in the process of multiphoton ionization. Within this model, a function has been obtained to determine the wavelength shift of the short-wavelength cutoff as a function of the multiphoton order. This function is in agreement with experimental results.

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1. INTRODUCTION

The broadening of the laser pulse spectrum in crystals and glasses detected for the first time in [1, 2] is a tool for obtaining supercontinuum, i.e., broadband coherent pulsed radiation. At the filamentation of femtosecond laser radiation, supercontinuum is formed because of the phase self-modulation of the light field in a nonlinear medium, which is enhanced by defocusing in the induced laser plasma and by the appearance of an envelope shock wave (*self-steepening*) [3, 4]. The supercontinuum spectrum at the plasma generation in the filament of a femtosecond pulse becomes asymmetric, and its anti-Stokes broadening is much larger than Stokes broadening [5]. Experiments on the filamentation of pulses at the fundamental wavelength, as well as at the second and third harmonics of radiation of a Ti:sapphire laser in various condensed media [6] showed that the anti-Stokes broadening of the supercontinuum spectrum is proportional to the ratio of the band gap width U_i to the energy of the radiation photon $\hbar\omega$. At filamentation under the conditions of normal group velocity dispersion, the spectrum has a unimodal character and the intensity of spectral components decreases monotonically with a decrease in their wavelength [4, 7]. At the filamentation of radiation in the middle infrared range, which is located in the region of anomalous group velocity dispersion of most of the transparent dielectrics, the supercontinuum spectrum in the anti-Stokes region is significantly nonmonotonic [7–11]. Studies of filamentation of pulses in fused silica at the variation of the wavelength in the range of 800–2300 nm showed that light bullets with a high localization of the light field in the space and time are formed at anomalous group velocity dispersion [12–14]. The formation of light bullets with a high intensity gradient is accompanied by the "emission" of anti-Stokes radiation caused by the strong phase self-modulation of the light field at the trailing edge of a light bullet [15–17]. In the process of propagation of light bullets, an isolated wing of the supercontinuum is formed; the shift of this wing toward the anti-Stokes region increases and the spectral width decreases with increasing pulse wavelength [15, 18–20]. The dispersion equation for the shift of the maximum of the anti-Stokes wing depending on the pulse wavelength obtained in [21] generalizes the results of known experiments on the study of the supercontinuum spectrum at the filamentation of radiation in the middle infrared range [8, 9, 15, 18, 20, 22–25].

The relation of the short-wavelength edge (short-wavelength cutoff) of the spectrum of the anti-Stokes wing of the supercontinuum to the width of the band gap of a dielectric at the filamentation of radiation in the middle infrared range was discussed in [8, 23, 26]. The shift of this edge toward shorter wavelengths with increasing multiphoton order in the process of plasma generation was observed and discussed in [13, 20, 25]. However, the characteristics of the formation of the short-wavelength cutoff in the spectrum of light bullets of the middle infrared range and their relation to the multiphoton ionization remain unstudied.

In this work, we report the results of the experimental and analytical studies of processes determining the short-wavelength cutoff of the anti-Stokes wing of the supercontinuum in the spectrum of light bullets of the middle infrared range. The spectrum of the anti-Stokes wing of the spectrum of light bullets at filamentation in fused silica and fluorides is recorded for the first time for femtosecond pulses at a wavelength tunable from 1350 to 4100 nm. A physical model for the formation of the short-wavelength cutoff in the spectrum of light bullets is proposed on the basis of the phase self-modulation of the light field on the trailing edge of the light bullet in the plasma in the process of multiphoton ionization in a dielectric.

2. EXPERIMENT

The supercontinuum spectrum at the filamentation of pulses in fused silica and LiF, CaF_2, and BaF_2 crystals was experimentally studied on the ISAN femtosecond spectrometric complex [21]. The wavelength of pulses λ_0 was varied from 1350 to 4100 nm. Laser pulses with a FWHM duration of 60 to 120 fs were focused by a thin CaF_2 lens with a focal length of 20 cm into a spot with a diameter of 50–120 μm on the front face of samples 30 mm in thickness. The energy of pulses was varied in the range of 0.5–18 μJ in order to reach the single light bullet regime at filamentation in dielectrics under consideration. Spectra were recorded by an ASP-100MF fiber spectrometer and an ASPIRHS spectrometer (Avesta Ltd.) in the spectral ranges of 200–1100 and 1200–2500 nm, respectively. In order to record integral spectra of the anti-Stokes band of the supercontinuum, radiation was focused on a thin diffusive scatterer placed immediately in front of the spectrometers. Modification of fused silica, CaF_2, and BaF_2 at the parameters of radiation under consideration was not observed, and spectra were recorded at a pulse repetition frequency of 1 kHz. Filamentation in LiF is accompanied by the formation of long-lived color centers, which qualitatively change the structure of the supercontinuum spectrum [27]. For this reason, when recording spectra in LiF, a sample was displaced at each pulse normally to the direction of pulse propagation.

3. EXPERIMENTAL RESULTS

In our experiments performed with fluorides and fused silica, we detected an increase in the short-wavelength shift and a decrease in the spectral width of the anti-Stokes band with an increase in the wavelength of the pump pulse of the middle infrared range for any material of the sample. This is clearly seen in Fig. 1, where the positions of the maximum, as well as the long- and short-wavelength cutoffs at a level of 0.01 (see the inset), of the visible band in the supercontinuum spectrum measured for LiF are shown. A similar effect was previously observed in fused silica also in [15, 18]. According to the presented data, the dependence of the position of the short-wavelength cutoff λ_{cut} on the wavelength of the pump pulse is different from that for the position of the maximum of the anti-Stokes wing determined by both the spectral broadening and interference effects at the formation of the spectrum of the anti-Stokes wing [21].

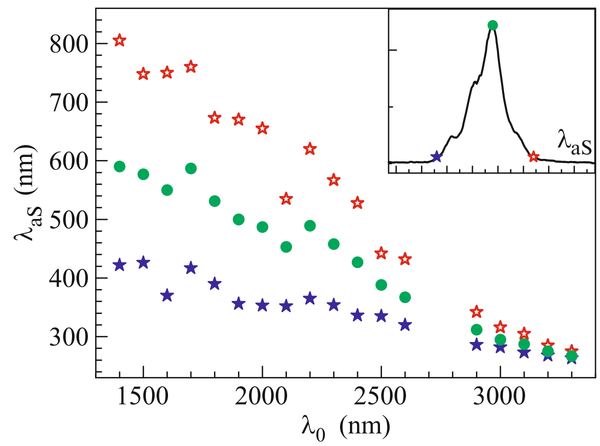

Fig. 1. (Color online) Wavelengths of the maximum and edges (see the inset) of the spectral band of the anti-Stokes wing in the spectrum of supercontinuum of the light bullet measured in LiF at the variation of the wavelength of the pump pulse from 1350 to 3300 nm.

Processing the measured spectra of the anti-Stokes band of the supercontinuum at the filamentation of femtosecond pulses in fused silica, CaF_2, BaF_2, and LiF, we determined the dependence of the shift $\Delta\lambda_{aS} = \lambda_0 - \lambda_{cut}$ of the short-wavelength cutoff λ_{cut} on the multiphoton order $K = [U_i/\hbar\omega_0 + 1]$, where ω_0 is the central frequency of pump radiation and [] means the integer part of the number (Fig. 2). At a monotonic decrease in λ_{cut} in the spectrum of light bullets with increasing λ_0 (Fig. 1), its shift $\Delta\lambda_{aS} = \lambda_0 - \lambda_{cut}$ increases almost linearly with increasing parameter K (Fig. 2). The experimentally obtained dependence $\Delta\lambda_{aS}(K)$ makes it possible to assume that multiphoton ionization plays a significant role in the formation of the short-wavelength cutoff.

4. PHYSICAL MODEL

Experimental results are analyzed within a model according to which the pulse spectrum broadening at the formation of light bullets is due to the phase self-modulation of the light field at the nonlinear interaction with a medium. In the approximation of the unidirectional propagation, the light field $E(r,t,z)$ in a nonlinear medium has the form

$$E(r,t,z) = E_0(r,t,z) \times \exp\{i(\omega_0(t - t_0) - k_0 z + \varphi_{nl}(r,t,z) + \text{const})\}, \quad (1)$$

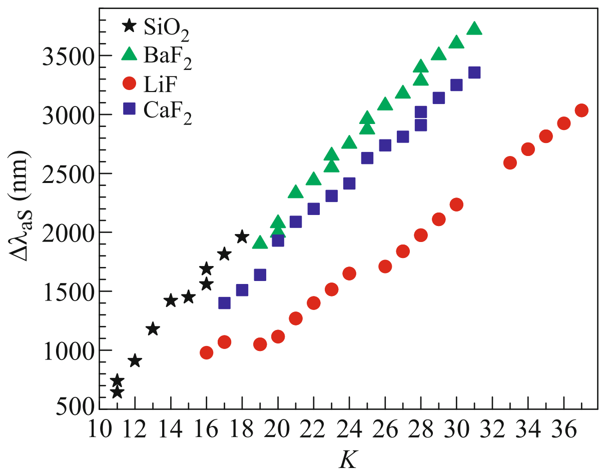

Fig. 2. (Color online) Shift of the short-wavelength cutoff $\Delta\lambda_{aS}$ versus the multiphoton order K obtained from the experimentally measured spectra for fused silica, CaF_2, BaF_2, and LiF.

where $E_0(r,t,z)$ is the complex amplitude; ω_0 and $k_0 = 2\pi/\lambda_0$ are the frequency and wavenumber of the pump pulse, respectively; and t_0 is the time layer of the pulse. Let $t_0 = 0$ correspond to the central layer of the pulse. Taking the first two terms in the expansion of the nonlinear phase incursion $\varphi(r,t,z)$ near the time layer t_0

$$\varphi_{\rm nl}(r,t,z) = \varphi_{\rm nl}(r,t_0,z) + \left.\frac{\partial\varphi}{\partial t}\right|_{t_0}(t-t_0), \qquad (2)$$

the light field $E(r,t,z)$ can be represented in the form

$$E(r,t,z) = E_0(r,t,z) \times\exp\{i((\omega_0+\Delta\omega)(t-t_0)-k_0z+\text{const})\}. \qquad (3)$$

Here, $\Delta\omega(r,t_0,z) = \left.\frac{\partial\phi}{\partial t}\right|_{t_0}(r,t_0,z)$ is the nonlinear increase in the frequency of the light field in the time layer t_0 of the pulse. The nonlinear phase incursion $\varphi_{\rm nl}(r,t,z)$ in the light bullet is determined by the Kerr nonlinearity $\Delta n_{\rm K}(r,,z)$ and the nonlinearity of the induced laser plasma $\Delta n_{\rm pl}(r,t,z)$:

$$\varphi_{\rm nl}(r,t,z) = -k_0\int_0^z(\Delta n_{\rm K}(r,t,z') + \Delta n_{\rm pl}(r,t,z'))dz', \qquad (4)$$

where z is the mean free path of light bullets. On the leading edge of the light bullet ($t_0 < 0$), where the energy density in the laser plasma is negligibly low and the Kerr nonlinearity dominates ($|\Delta n_{\rm K}| \gg |\Delta n_{\rm pl}|$), the nonlinear frequency shift is given by the formula

$$\Delta\omega(r,t_0<0,z) = -k_0n_2\int_0^z\frac{\partial I}{\partial t}(r,t_0<0,z')dz', \qquad (5)$$

where n_2 is the cubic nonlinearity coefficient of the medium. Since the time derivative of the intensity at $t_0 < 0$ is $\partial I/\partial t > 0$, $\Delta\omega(r,t_0<0,z) < 0$, which corresponds to the Stokes shift of the frequency of the light field at the leading edge of the light bullet.

At the trailing edge of the light bullet ($t_0 > 0$), where the contribution from the laser plasma to a change in the refractive index prevails, the nonlinear phase incursion $\varphi_{\rm nl}(r,t_0>0,z)$ is determined by an increase in the real part of the refractive index of the plasma, which can be represented in the form

$$\Delta n_{\rm pl}(r,t,z) = -\frac{1}{2}\left(\frac{\Omega}{\omega_0}\right)^2\left(\frac{N_e(r,t,z)}{N_0}\right). \qquad (6)$$

Here, $N_e(r,t,z)$ is the electron density at the front of the plasma channel formed at the tail of the light bullet, N_0 is the density of neutrals, and $\Omega^2 = e^2N_0/(\varepsilon_0m_e)$ is the square of the plasma frequency at the unit degree of ionization. Correspondingly, the nonlinear frequency shift at the trailing edge of the light bullet $\Delta\omega_{\rm aS}(r,t_0>0,z)$ is

$$\Delta\omega_{\rm aS}(r,t_0>0,z) = \frac{k_0}{2}\left(\frac{\Omega}{\omega_0}\right)^2 \times\int_0^z\frac{\partial}{\partial t}\left(\frac{N_e(r,t_0>0,z')}{N_0}\right)dz'. \qquad (7)$$

Since $\partial N_e(r,t_0>0,z)/\partial t > 0$, the frequency shift is positive $\Delta\omega_{\rm aS}(r,t_0>0,z) > 0$, and the trailing edge of the light bullet is thereby a source of anti-Stokes components of the supercontinuum.

The time dependence of the electron density $N_e(r,t,z)$ is generally determined by photoionization, avalanche ionization, and recombination processes in the plasma:

$$\frac{\partial N_e(r,t,z)}{\partial t} = W(I)(N_0-N_e(r,t,z)) + \nu_iN_e(r,t,z) - \beta N_e^2. \qquad (8)$$

The characteristic frequency ν_i of inelastic collisions of electrons with neutrals is given by the formula

$$\nu_i = \frac{1}{U_i}\frac{e^2|E_0|^2}{2m_e(\omega_0^2+\nu_c^2)}\nu_c, \qquad (9)$$

where m_e is the electron mass, e is the elementary charge, U_i is the width of the band gap, and ν_c is the frequency of elastic collisions. In a dielectric, e.g., with $U_i \approx 10$ eV and $\nu_c \approx 10^{14}$ s^{-1} for radiation at a wavelength of $\lambda_0 = 3$ μm with an intensity of $I = 10^{13}$ W/cm^2, the frequency ν_i is estimated as 10^{15} s^{-1}. Because of the plasma defocusing and appearance of a shock wave enveloping the light bullet, the duration of its trailing edge, which is a source of anti-Stokes components of the supercontinuum, decreases to several femtoseconds [14]. Under these conditions at a relative electron density in the laser plasma below one-tenth percent ($N_e < 10^{-3}N_0$), the contribution of avalanche ionization to a change in the electron density at the trailing edge of the light bullet can be neglected. Recombination processes with a characteristic time of several picoseconds do not affect the electron density N_e.

The peak intensity of the light field in the light bullet increases from 10^{10}–10^{11} W/cm^2 to a saturation intensity of 10^{13}–10^{14} W/cm^2 (*intensity clamping*). In fused silica and fluorides at the wavelength of the middle infrared range, the adiabaticity parameter $\gamma = \omega_0\sqrt{2m_eU_i}/(e|A|)$ [28] at a saturation intensity of 10^{14} W/cm^2 is below unity, which corresponds to the prevalence of tunneling ionization. However, at the trailing edge of the light bullet, where the intensity is about 10^{13} W/cm^2, the adiabaticity parameter is $\gamma \geq 1$ and photoionization occurs in a regime intermediate between tunneling and multiphoton ionization. The experimentally obtained dependence of the shift of the cutoff wavelength $\Delta\lambda_{aS} = \lambda_0 - \lambda_{cut}$ on the multiphoton parameter in the spectrum of the light bullet (Fig. 2) implies that multiphoton ionization dominates in a change in the electron density at the trailing edge of the light bullet, where the anti-Stokes components of the supercontinuum are generated. In this case, the time dependence of the electron density is described by the equation

$$\partial N_e/\partial t = \sigma_K I^K N_0, \quad (10)$$

where σ_K is the multiphoton ionization cross section and I is the intensity at the trailing edge of the light bullet. The short-wavelength cutoff frequency ω_{cut} in the anti-Stokes region of the supercontinuum spectrum is determined by the maximum rate of increase in the electron density, which is achieved just at multiphoton rather than tunneling ionization. It is noteworthy that, according to the experiments performed with various materials with a pulse at a wavelength of 1300 nm with a duration of 110 fs [29], nonlinear absorption in broadband dielectrics increases with increasing intensity to 5×10^{13} W/cm^2 as $I^{K'}$, where the parameter K' is slightly smaller than the multiphoton parameter K. In the accepted approximations, the anti-Stokes broadening of the spectrum $\Delta\omega_{aS}$ determined by Eqs. (7) and (10) under the assumption that $\partial N_e/\partial t$ does not vary at the mean free path of the light bullet is given by the formula

$$\Delta\omega_{aS} = \frac{\pi}{\lambda_0}\left(\frac{\lambda_0}{\Lambda}\right)^2 z\sigma_K I^K. \quad (11)$$

Since the cross section σ_K depends weakly on the intensity and the mean free path of the light bullet

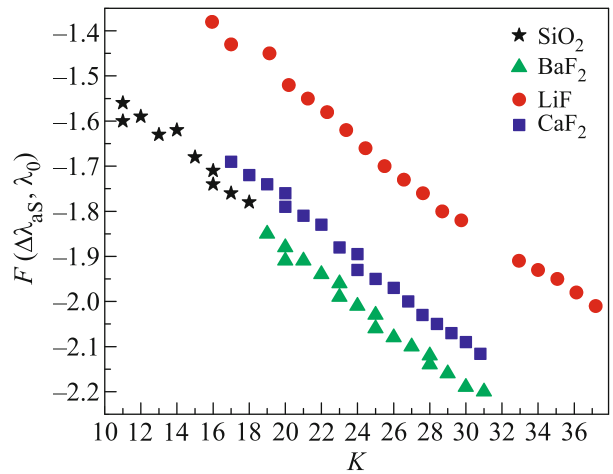

Fig. 3. (Color online) Function $F(\Delta\lambda_{aS}, \lambda_0)$ calculated from experimental data versus the multiphoton order K.

depends weakly on the wavelength λ_0, passing from the frequency shift $\Delta\omega_{aS}$ to the short-wavelength cutoff shift $\Delta\lambda_{aS} = \lambda_0 - \lambda_{cut}$, one can find a function $F(\Delta\lambda_{aS}, \lambda_0)$ proportional to K:

$$F(\Delta\lambda_{aS}, \lambda_0) = \log\left(\frac{\Delta\lambda_{aS}\Lambda^2}{\lambda_0^3}\right) \propto K + \text{const},$$

where $\Lambda = 2\pi c/\Omega$. In solid dielectrics, $N_0 \approx 10^{22}$ cm^{-3} and the plasma wavelength at complete ionization is $\Lambda \approx 0.34$ μm.

The linear dependence of the function on the multiphoton order K is confirmed by plots obtained from measurements performed for all studied materials at the variation of the wavelength of the pulse of the middle infrared range from 1350 to 4100 nm (Fig. 3). To calculate the function $F(\Delta\lambda_{aS}, \lambda_0)$ determining the short-wavelength cutoff in fused silica, we used the experimental data presented in [18]. It is noteworthy that, according to Fig. 1, at an increase in the wavelength of the pump pulse, when an increase in the contribution of tunneling ionization should be expected, the role of multiphoton ionization remains decisive for the entire anti-Stokes wing of the supercontinuum.

The found linear dependence of the function $F(\Delta\lambda_{aS}, \lambda_0)$ on the multiphoton order K certainly indicates that the cutoff wavelength λ_{cut} of the anti-Stokes wing of the supercontinuum generated by light bullets of the middle infrared range is determined by the multiphoton generation of the dynamic front of the plasma channel by the light field at the trailing edge of the light bullet. Thus, the analysis of the experimental data shows that the shift of the supercontinuum spectrum toward shorter wavelengths or, in other words, its short-wavelength cutoff is determined by a high rate of multiphoton ionization of the plasma created by the trailing edge of the light bullet.

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