Dependence of the short-wavelength cutoff in the mid-IR pulse spectrum on the interaction length in SiO$_2$ and CaF$_2$

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Abstract. This paper presents the results of experimental and numerical study of the anti-Stokes wing formation and the evolution of the short-wavelength cutoff in the 1900 nm pulse spectrum during its propagation and filamentation in fused silica and calcium fluoride. It is found that during the light bullet formation the short-wavelength cutoff in the supercontinuum spectrum is shifted to the anti-Stokes region with an increase in the nonlinear optical interaction length with the medium. However, change in the short-wavelength cutoff in the formed light bullet spectrum throughout its further propagation is insignificant.

1. Introduction
Filamentation of high-power femtosecond laser radiation in the anomalous group velocity dispersion regime leads to formation of a high-intensity wave packet, extremely compressed in space and time, — a light bullet [1]. Spatiotemporal transformation of the wave packet during the light bullet formation is accompanied by significant broadening of its spectrum — generation of a supercontinuum [2], the spectrum of which extends from near ultraviolet to mid-infrared region. The principal feature of the anti-Stokes region in the light bullet spectrum is the narrow wing, separated from the central region by a broad spectral minimum, which is formed as a result of destructive interference of supercontinuum radiation [3]. Supercontinuum generation by light bullets is a topic of great interest.

Investigations on the anti-Stokes wing of the supercontinuum spectrum showed that increase in central wavelength of the pulse leads to greater shift of the wing to the short-wavelength region whereas its spectral width decreases [4, 5]. The relationship between the shift of the anti-Stokes wing maximum and the pulse wavelength found in [6] on the basis of interference theory generalises experimental results obtained in different dielectric media with pulses of various frequencies. Short-wavelength cutoff in the spectrum of the mid-IR pulse propagating in the medium with anomalous group velocity dispersion depends on the radiation wavelength and band gap of the dielectric [7]. Experiments carried out in fused silica, LiF, CaF$_2$, and BaF$_2$ crystals with pulses at a wavelength tunable from 1350 to 4100 nm showed that the shift of the short-wavelength cutoff in the light bullet spectrum is linearly dependent on the order of multiphoton ionization process [8]. However, the evolution of the short-wavelength cutoff during the propagation and filamentation of the pulse in nonlinear optical medium has not yet been fully investigated. Therefore, the aim of this work consisted in the experimental and numerical study of the processes of the anti-Stokes wing formation and the short-wavelength cutoff shift during formation and propagation of the light bullet in fused silica and calcium fluoride.
2. Method

2.1. Experiment
The experimental study of the light bullet spectrum evolution during the filamentation of high-power femtosecond mid-IR laser radiation under anomalous group velocity dispersion conditions was conducted at the large-scale research facility "Multipurpose Femtosecond Laser Diagnostic Spectrometric Complex" at the Institute of Spectroscopy of the Russian Academy of Sciences. The femtosecond Ti:Sapphire Tsunami laser was used in the experiments; the laser pulses were sent to the Spitfire Pro regenerative amplifier, combined with the TOPAS parametric amplifier. The experimental setup diagram is shown in figure 1.

![Figure 1. Diagram of the experimental setup.](image)

Pulsed radiation at the wavelength of 1900 nm with the repetition rate of 1 kHz, the pulse energy of several μJ and FWHM duration of 70 fs, after the parametric amplification, was focused by a thin CaF$_2$ lens with focus length of 30 cm onto the entrance face of the wedge-shaped transparent dielectric sample. The FWHM beam width at the focus was 130μm. To study the relationship between the nonlinear optical interaction length in the dielectric and the pulse spectrum, the stage with the sample was moved perpendicularly to the direction of radiation propagation. This made it possible to change the interaction length while the input radiation parameters remained the same. To record the anti-Stokes region of the supercontinuum spectrum, the infrared part of which was cut off by the filter, the SL-40 Solar TII spectrometer, operating in range 200-1100 nm, was used.

2.2. Numerical simulation
For numerical simulation of pulse filamentation and formation of light bullets, the slowly evolving wave approximation was used [9]. This modification of slowly varying envelope approximation allows to extend the concept of the envelope in the description of the laser pulse propagation in the nonlinear medium to the case of the extremely short pulse duration, comparable to the carrier oscillation period. A scalar, axially symmetrical nonlinear envelope equation in the coordinate system moving with the pulse group velocity was considered [3]. Mathematical model used describes the effects of diffraction and dispersion of radiation, laser plasma generation during photo- and avalanche ionization in the dielectric, increment of medium refraction index caused by Kerr and plasma nonlinearities, as well as radiation attenuation due to inverse bremsstrahlung and ionization losses.

The wave packet parameters chosen for the numerical simulation were close to those in the actual laboratory experiments. The initial spatio-temporal distribution of the envelope amplitude $A(r,t,z=0)$ was considered as Gaussian:

$$A(r, t, z = 0) = A_0 \exp \left( - \frac{r^2}{2a_0^2} - \frac{\tau^2}{2\tau_0^2} \right).$$
3. Results

According to the numerical simulation results, there are several stages of the femtosecond pulse spectrum evolution with propagation distance during the filamentation and light bullet formation. At the beginning of the pulse propagation in the medium, the symmetric broadening of the initial spectrum occurs. Then the pulse spectrum undergoes sharp asymmetric broadening to the anti-Stokes side. This anti-Stokes broadening is directly connected with the light bullet formation and laser plasma generation; it occurs as a result of self-phase modulation of the light field caused by defocusing in self-induced laser plasma and self-steepening effect [1, 3]. Thus, the supercontinuum generation occurs. In the interval of the light bullet formation, the anti-Stokes broadening of the spectrum increases because of the growth of the pulse trailing edge steepness, and the short-wavelength cutoff in the spectrum is shifted to the anti-Stokes side with the increase in pulse propagation length.

Experimentally obtained spectra of the anti-Stokes wing, illustrating the shift of the short-wavelength cutoff during the process of the light bullet formation in fused silica and calcium fluoride, are presented in figure 2. When the radiation propagation distance in fused silica was approximately 4 mm, a third harmonic signal appeared in the visible region of the spectrum, and this position corresponds to $\Delta z = 0$ in figure 2A. An increase in the radiation propagation distance in fused silica ($\Delta z = 0.86$ mm) led to the appearance of the anti-Stokes spectral components in the visible region of the spectrum, which indicated the beginning of the light bullet formation. A further increase in nonlinear optical interaction length led to the formation of an isolated anti-Stokes wing with a maximum at the wavelength of 575 nm, which gradually shifted to the short-wavelength region with the increase of the pulse propagation distance. The cutoff wavelength, determined by the 0.1 level of the anti-Stokes wing spectral maximum, which at $\Delta z = 0.86$ mm was equal to 570 nm, decreased to 405 nm at $\Delta z = 1.2$ mm. After that it shifted to 450 nm at $\Delta z = 1.6$ mm and remained nearly unchanged with a further increase in the pulse propagation distance to $\Delta z = 2$ mm, as the light bullet was already formed. The same qualitative picture of the femtosecond pulse spectrum evolution and short-wavelength cutoff shift was observed in calcium fluoride (figure 2B; the position $\Delta z = 0$ also corresponds to the appearance of the third harmonic signal).

![Figure 2](image.png)

Figure 2. Anti-Stokes region of the experimental spectra obtained at different pulse propagation distances in fused silica (A) and calcium fluoride (B).

Figure 3 clearly illustrates the dynamic of the short-wavelength cutoff with the increase in the nonlinear optical interaction length in fused silica and calcium fluoride. In the numerical simulation, as well as in the laboratory experiment, the cutoff wavelength in the supercontinuum spectrum decreases with the increase in the nonlinear optical interaction length in the interval of the light bullet formation. During the further propagation of the light bullet, the cutoff remains practically unchanged. The interval of about 350 μm is the length of the light bullet formation where the anti-Stokes shift of the short-wavelength cutoff is about 100 nm per 100 μm of propagation length in figure 3A.
4. Conclusion
The general character of the short-wavelength cutoff shift process is the same for fused silica and calcium fluoride. Increase in the interaction length with the medium in the process of the light bullet formation in a transparent dielectric leads to the shift of the short-wavelength cutoff in the pulse spectrum to the anti-Stokes region, whereas the cutoff in the spectrum of the formed light bullet does not significantly change throughout its further propagation.

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