Journal of Siberian Federal University. Mathematics & Physics 2025, 18(3), 331–336

EDN: JOFSFJ
УДК 535-15

# Spectrum Broadening of Femtosecond Radiation Pulse at 950 nm Wavelength in Material with Cubic Nonlinearity

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Received 11.09.2024, received in revised form 24.10.2024, accepted 25.01.2025

**Abstract.** The conditions for spectrum broadening of 60 fs radiation pulse at a central wavelength 950 nm in fused quartz plates due to phase self-modulation are studied experimentally and theoretically. The studies are conducted at radiation intensities from 50 to 400 GW/$cm^2$ and plates thicknesses from 1 to 10 cm. The experimental conditions and a calculation model based on solving a system of nonlinear Schrödinger equations in the approximation of a slowly changing wave are described. The possibility of compressing radiation pulse with a broadened spectrum in case of quadratic nonlinear phase compensation is estimated. It is shown that in case spectrum broadening it is possible to reduce the spectrally limited duration by no more than two times.

**Keywords:** femtosecond pulse, spectrum width, self-phase modulation, fused quartz.

**Citation:** S.V. Alekseev, V.F. Losev, Ya.V. Grudtsyn, A.V. Koribut, V.I. Yalovoy, Spectrum Broadening of Femtosecond Radiation Pulse at 950 nm Wavelength in Material with Cubic Nonlinearity, J. Sib. Fed. Univ. Math. Phys., 2025, 18(3), 331–336.
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## Introduction

Currently, powerful infrared (0.8 – 1 $\mu$m) laser systems are usually used to obtain powerful radiation pulses in the visible spectrum, the radiation pulses of which are converted into the second harmonic (SH) in nonlinear crystals. In Tomsk (IHCE SB RAS) together with Moscow group (FIAN), the alternative way of obtaining such pulses in visible region (475 nm) is being developed based on the THL-100 hybrid laser system. This way is associated with the initial production of femtosecond SH pulse in visible region with low energies (1 – 5 mJ) and subsequent increase in energy in a gas amplifier on XeF(C-A) molecules to the Joule level. The THL-100

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laser system, operating on this principle, is currently one of the most powerful systems in visible spectrum [1, 2]. The general interest in powerful pulses of radiation in visible spectrum is connected with the higher quantum energy, which allows to increase the efficiency of interaction of laser beam with various materials and environments. Powerful pulses also help to develop new applications, which include the creation of an X-ray laser in transparency window of water and the generation of powerful terahertz radiation [3–5].

One of the ways to increase the power of THL-100 laser system is to reduce the duration of output radiation pulse while maintaining the radiation energy. To do this, it is necessary to increase the spectral width of SH radiation pulse in the Ti:Sa complex and amplify it in active medium on XeF(C-A) molecules, which has a wide gain contour corresponding to a spectrally limited duration of 10 fs [6]. Since the femtosecond complex operates at the gain contour edge (950 nm) to match the SH wavelength with gas amplifier, it does not allow forming a radiation pulse shorter than 60 fs at the fundamental harmonic. To reduce the spectrally limited duration of radiation pulse, it is necessary to increase the width of its spectral contour by some artificial method. At present, the most common method of spectrum broadening for reducing the spectrally limited pulse duration is self-phase modulation in a medium with cubic nonlinearity.

This approach was proposed for high-power laser systems [7] and demonstrated at the PEARL facility (central wavelength 910 nm, pulse duration 65 – 75 fs, pulse energy up to 17 J, beam diameter 18 cm) [8]. The pulse spectrum at the laser output was broadened due to self-phase modulation in fused silica and then the pulse was compressed by chirped mirrors. It was demonstrated that with optimal choice of mirror dispersion a pulse with energy of 17 J can be compressed from 70 to 14 fs. This compression has undoubted merits: simplicity, low cost, negligible pulse energy losses, and applicability to any high-power laser.

This paper presents theoretical and experimental results of studies aimed at studying the possibility of broadening the radiation spectrum with a central wavelength of 950 nm depending on the glass block thickness and radiation intensity.

## 1. The equipment and methods

In the experiments, the femtosecond Ti:Sa laser complex operating at the gain contour edge (central wavelength is 950 nm) and serving as master oscillator (front end) for multiterawatt THL-100 laser system was used. The laser complex consists of master oscillator, stretcher, regenerative and two multi-pass amplifiers, and compressor on diffraction gratings. The output pulse at the fundamental harmonic has duration of 60 fs, beam diameter of 15 mm with intensity decay of $e^2$ times and energy of up to 10 mJ. The beam was directed without focusing onto polished fused quartz plates of different thickness (from 1 to 10 cm), where the spectrum was broadened due to self-phase modulation. After that, the radiation was recorded with an Ocean Optics HR4000 spectrometer (200 – 1100 nm, 0.7 nm). The laser radiation energy in the experiments was measured by Gentec-e maestro energy meter.

The model that takes into account the influence of phase self-modulation, group velocity dispersion and nonlinear response, nonlinear absorption, plasma formation and spatial effects associated with self-focusing was used in simulations. The model is based on solving the system of nonlinear Schrödinger equations in approximation of slowly varying wave [9] in cylindrical coordinate system and it has the following form:

$$\frac{\partial A}{\partial z} + iD\frac{\partial^2 A}{\partial \eta^2} + iD_\perp \Delta_\perp A + i\frac{ik_0^2 n_2}{n_0}\left(1 + \frac{i}{\omega_0}\frac{\partial}{\partial \eta}\right)\left(\int_0^\infty R(t')|A(\eta - t')|^2 \mathrm{d}t'\right) = 0 \qquad (1)$$

where $\eta$ is the dimensionless time in the traveling coordinate system, $z$ is the longitudinal coordinate, $D$ is the coefficient characterizing the second-order dispersion, $\omega_0$ is the average pulse frequency, $k_0$ is the wave number, *A(n, r, z)* is the complex envelope of the electric field (the initial distribution was Gaussian), *R(t)* is the nonlinear response function, including the fast and slow part, $n_0$ and $n_2$ are linear and nonlinear refractive coefficients. For numerical solution of nonlinear Schrödinger equations the conservative difference scheme with second order approximation was used both in the spatial coordinate and in time [10]. The simulations were carried out for laser beam intensity from 50 to 400 GW/$cm^2$. The thickness of fused quartz was varied in the same range as in experiments.

## 2. Results and discussion

The experimental study of spectrum broadening of first harmonic radiation was carried out at laser pulse energy of 8 mJ and radiation intensity at entrance to the material of 76 GW/$cm^2$. The initial width of laser radiation spectrum at half-maximum (FWHM) was 28 nm (Fig. 1).

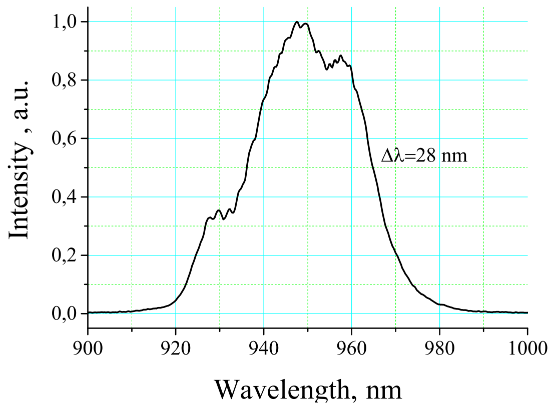


Fig. 1. Spectral contour of the fundamental harmonic radiation

The beam was passed through plates of fused quartz starting from 1 cm thickness and up to 10 cm. At the same time with thickness increase of the plates to 6 cm the spectrum width was increased, and with its further increase the spectrum width began to decrease due to Kerr nonlinearity. Here we present the spectrum only for the optimal regions. Thus, the spectrum width increased to 41.8 nm when using a 4 cm thick plate (Fig. 2a). When using a 6 cm plate, the spectrum width increased to 53.3 nm (Fig. 2b). That is, for these conditions, the spectrum width increased by 1.5 and 2 times, respectively. It is clear that, in general, the spectrum shifts to the short-wave side. Most likely, this is due to the fact that the temporal shape of laser radiation pulse is not Gaussian and the trailing edge is significantly steeper than the leading edge. Deep amplitude modulation of the radiation intensity is typical for spectral form behavior in phase self-modulation. In this case the duration of spectrally limited pulse is usually determined by the spectrum envelope at half amplitude. That is, we can hope that in our case the duration will be reduced by about two times.

Simulations close to the experiment showed that at intensity of 100 GW/$cm^2$ and 4 and 6 cm thick plates the spectrum width increases to 38.9 and 42.6 nm, respectively (Fig. 3a). In

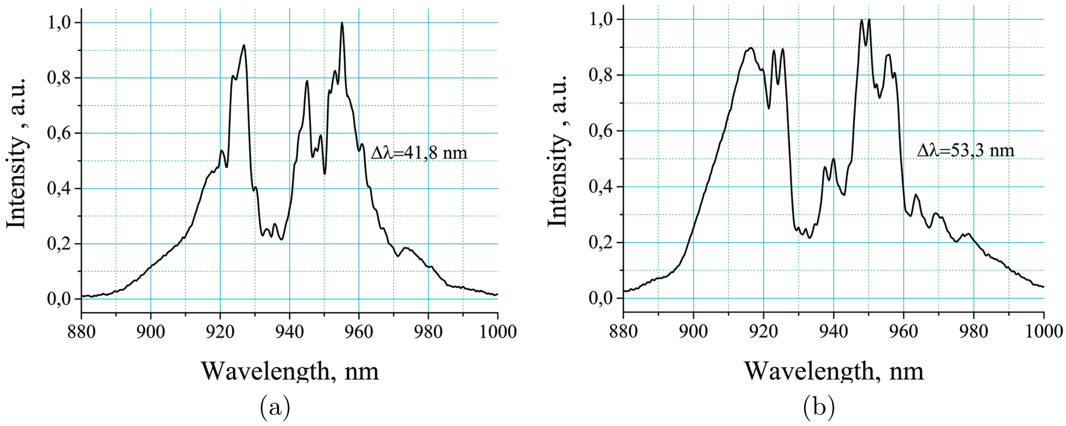


Fig. 2. Experimental spectral contours of broadened radiation obtained after passing through 4 m (a) and 6 cm (b) of fused quartz

this case, the broadening occurs symmetrically on both sides of the central wavelength. With increasing of intensity to 400 GW/$cm^2$ the spectrum width increased to 72 nm (Fig. 3b). In this case a significant width increase at the spectrum base was observed. Simulations showed that the spectrum broadening occurs mainly due to phase self-modulation. The simplified model did not allow obtaining a real picture of the spectrum with modulation.

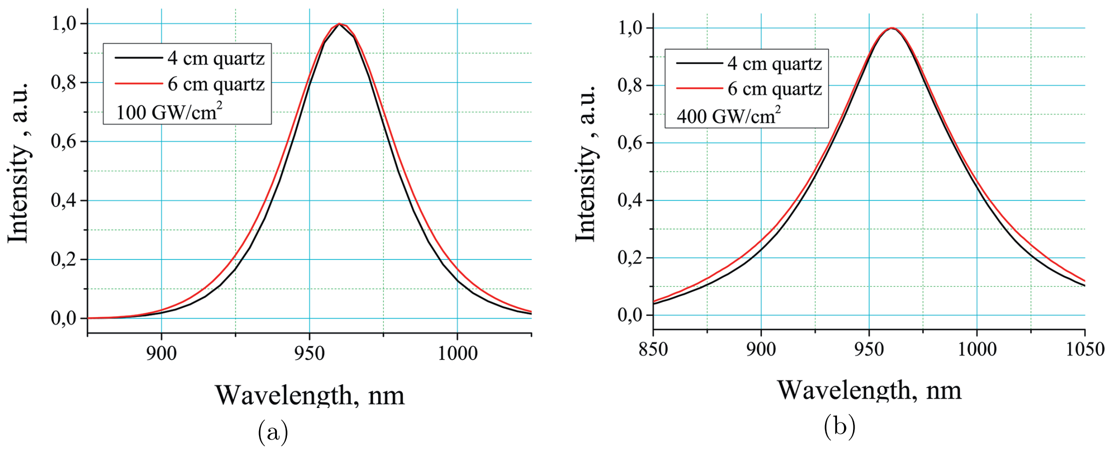


Fig. 3. Theoretical spectral contours of broadened radiation obtained after passing 4 (black) and 6 (red) cm of fused quartz at laser beam intensity of 100 (a) and 400 (b) GW/$cm^2$

To calculate the pulse duration of radiation with a broadened spectrum the beam was propagated in medium with negative dispersion. It was shown that pulse duration was reduced no more than twice. That is compensation for only the second-order dispersion is sufficient for Gaussian beam profile.

# Conclusion

Thus experimental and theoretical studies of the broadening possibility of radiation spectral contour of the fundamental harmonic at a central wavelength of 950 nm in fused quartz have been carried out and the possibility of compression of the radiation pulse with a broadened spectrum when compensating for the quadratic nonlinear phase was evaluated. It was shown that with increase of the material thickness from 1 to 6 cm in experiment and simulations the spectrum widens up to two times. However, the calculated shape of broadened spectral contour does not coincide with the experiment. To reconcile these data, both further refinement of the computational model (accounting for absorption and scattering) and greater approximation of the calculation conditions to the experiments (non-Gaussian pulse, the presence of cubic phase, etc.) are required. This is planned in our further research. Actually, in calculations, it is possible to reduce the spectrally limited duration up to two times. A similar pattern is expected in the experiment when compensating for the positive dispersion.

*The work was supported by the Ministry of Science and Higher Education of the Russian Federation (FWRM-2021-0014).*

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# Уширение спектра фемтосекундного импульса излучения на длине волны 950 нм в материале с кубической нелинейностью

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**Аннотация.** Экспериментально и теоретически исследуются условия уширения спектра 60 фс импульса излучения на центральной длине волны 950 нм в пластинах из плавленого кварца за счет фазовой самомодуляции. Исследования проводятся при интенсивности излучения от 50 до 400 ГВт/$cm^2$ и толщине пластин от 1 до 10 см. Описываются условия экспериментов и расчетная модель, основанная на решении системы нелинейных уравнений Шредингера в приближении медленно меняющейся волны. Оценивается возможность сжатия импульса излучения с уширенным спектром при компенсации квадратичной нелинейной фазы. Показывается, что при уширении спектра удается сократить спектрально ограниченную длительность не более двух раз.

**Ключевые слова:** фемтосекундный импульс, ширина спектра, плавленый кварц.