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# Spectral features of the Stokes part of supercontinuum generated by femtosecond light pulses in selected oxide crystals: A comparative study

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The peculiarities of the Stokes part of supercontinuum (SC) generated by femtosecond light pulses at wavelength 800 nm in single crystals of $Gd_2SiO_5$ (GSO), $Ca_4GdO(BO_3)_3$ (GCOB), $Gd_3Ga_5O_{12}$ (GGG), $LiTaO_3$ (LTO) and $LuVO_4$ (LVO) were investigated. Spectral bandwidth and intensity of SC were measured as a function of energy of incident 100 fs pulses employing a grating spectrograph coupled with an InGaAs detector and spatial characteristics of the beam inside crystal samples were monitored perpendicularly to the direction of propagation and recorded using an optical microscope coupled with a camera. It was found that spectral widths of the Stokes part of SC increase markedly with increasing energy of incident pulses for all crystals under study. For fixed focusing conditions the spectral widths of generated SC in GSO, GCOB and GGG wide band-gap crystals are relatively large with cut-off wavelengths close to 1500 nm. Bandwidths of SC generated in LVO and LTO crystals, characterized by band-gaps $E_g$ inferior to three times incident photon energy, are markedly smaller with cut-off wavelengths of 1300 nm and 1150 nm, respectively. Increase of incident pulse energy affects SC spectra giving rise to plateau-like regions stretching to ca 1000 nm.

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## 1. Introduction

Supercontinuum generation (SC) induced by femtosecond pulses has been observed first in an ethylene glycol in year 1983 [1] and in gases in year 1986 [2]. Subsequent works on this subject have aimed to understand the mechanisms of these observed phenomena. A comprehensive investigation reported by Brodeur and Chin [3] has provided a comparison of experimental data acquired for various liquid and solid state media and an assessment of the importance of the particular mechanisms involved. It has been pointed out in this early study that an ultra-short light pulse (in the range of tens or thousands of femtoseconds) propagating in a transparent medium is affected by an instantaneous Kerr nonlinearity that gives rise to temporal distribution of nonlinear phase denoted as the self-phase modulation (SPM) and by effect of self-focusing. Both these effects depend on the material's index of refraction $n = n_0 + n_2 I$, where $n_0$ denotes a linear index of refraction and $n_2$ denotes a nonlinear refraction coefficient induced by a high energy density of electromagnetic field. The relevance of the self-focusing effect is corroborated by a coincidence of experimentally observed power thresholds for continuum generation with estimated critical powers for self-focusing. Self-focusing brings about a creation of filaments - regions with transversal diameters within tens of micrometers encompassing a significant part of the pulse energy. The effect of self-focusing is counterbalanced by a defocusing effect of multiphoton excitation (MPE) which promotes electrons from valence band VB to conduction band CB thereby producing free electrons in the filament region. This process depletes the field energy and plasma is created which induces a negative change in the index of refraction. As a consequence, the self-focusing is stopped and a stable continuum generation can be obtained. The relation between the phenomena of SC and filamentation has been considered in numerous subsequent experimental and theoretical studies, e.g. in Refs. [4–6]. Results then gathered provided an evidence that the SC is related to an interplay between self-phase modulation, self-focusing, self-steepening, four-wave mixing, multiphoton absorption and plasma formation. Recently an interest in phenomena of SC is increasing abruptly. In fact, hundreds of scientific papers presenting results of basic

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research and practical issues in this topic have been published during last decade. An excellent review by Dubietis et al. [7], just published, summarizes a progress and presents a state-of-the art in this field.

Considerable attention directed to SC in wide band-gap solid-state media is stimulated by their potential for application purposes as sources of broad-band emission and parametric optical amplification. Materials investigated thus far encompass glasses, e.g. fused silica [3], BK-7 glass [8], or crystals, e.g. LiF [3], $CaF_2$ [3], $BaF_2$ [8–10], sapphire [11], yttrium aluminium garnet (YAG) potassium gadolinium tungstate (KGW), gadolinium vanadate $GdVO_4$ and yttrium vanadate $YVO_4$ [12]. In studies mentioned above the characteristics of the anti-Stokes (visible part) of SC in normal group velocity dispersion (GVD) regime have been considered mainly. Nevertheless, spectral features of the Stokes part of SC in YAG crystals have been then studied, too [12]. More recently, investigation of SC (including its Stokes part) in barium fluoride, calcium fluoride and fused silica, generated at different incident wavelengths corresponding to normal and anomalous GVD regimes has been reported [13]. Detailed experimental and theoretical investigation of an infrared extension of SC in sapphire has provided generalizations relevant for better understanding of temporal dynamics that leads to supercontinuum generation [14]. Supercontinuum generation has been achieved also in sapphire and YAG crystals with picosecond light pulses [15]. An ultrabroadband SC at 2 μm in sapphire, fused silica, $CaF_2$ and YAG in the regime of anomalous GVD has been demonstrated [16].

In the present work we report results gathered during a comparative study of spectral features of the Stokes part of supercontinuum generated by femtosecond pulses at wavelength 800 nm in $Ca_4GdO(BO_3)_3$ (GCOB), $Gd_2SiO_5$ (GSO), $Gd_3Ga_5O_{12}$ (GGG), $LiTaO_3$ (LTO) and $LuVO_4$ (LVO) single crystals manufactured by the Czochralski technique. The choice of the crystals represents a trade-off between an intention to gather media which differ markedly in energy gap and nonlinear refraction coefficient on one hand, and on the other hand securing the ready availability of real materials of the highest structural and optical quality. All these crystals are of interest for application in optics and optoelectronics. Therefore their manufacture has been mastered and their fundamental structural and optical properties of relevance to the present study are available.

Intention of the work arises from two reasons. First, to get a new information on infrared extension of SC in crystals characterized by relatively narrow bandgaps, especially those that have bandgap values smaller than three times the pump photon energy and according to predictions [4] are not be able to show SC. To our knowledge results presented in this work have not been reported before. Second, a detailed knowledge on spectral features of SC may help to understand unclear mechanisms of nonresonant up-conversion phenomena in rare earth-doped crystals excited by infrared femtosecond pulses [17–19]. Results gathered in the present work will be discussed with a reference to peculiarities of the anti-Stokes part of SC in GSO, GGG, LTO and LVO crystals reported in Ref. [20].

## 2. Experimental

Experiments were accomplished using a femtosecond laser (Coherent Model “Libra”) emitting 100 fs light pulses at 800 nm with a repetition rate of 1 kHz. The highest pulse energy was 1 mJ. Fig. 1 depicts a scheme of experimental set-up employed. The incident pulse energy was attenuated using a set of calibrated neutral density filters $F_1$ (ThorLabs) and the energy of transmitted light was monitored with an energy meter (Coherent Model Field $Max_{II}$). The attenuated incident beam passed through a circular

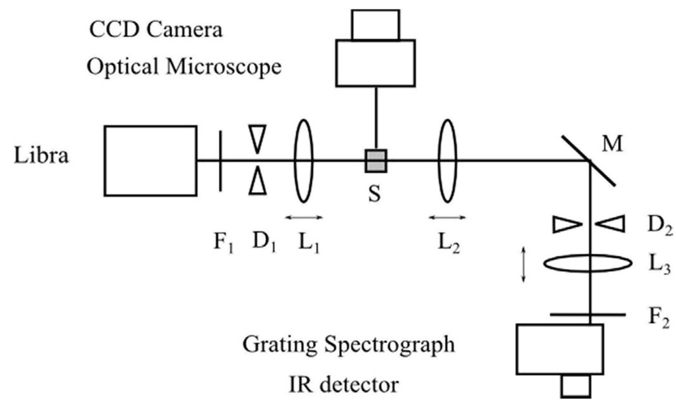


**Fig. 1.** Scheme of experimental set-up: $D_1$, $D_2$ - diaphragms, $F_1$ - variable density attenuator, $F_2$ - long pass 850 nm filter, M - mirror, $L_1$, $L_2$, $L_3$ - lenses, S - sample.

diaphragm $D_1$ and was then focused with a $L_1$ lens. Resulting SC beam was spatially filtered with a diaphragm $D_2$ to discriminate a conical emission and the intense light around pump wavelength was attenuated with a 850 nm long pass filter. Spectral characteristics of the Stokes part of SC generated in crystal samples were analysed with a grating spectrograph (Princeton Instr. Model Acton 2500i) coupled with an InGaS detector (Acton Model ID 441-C). Spatial characteristics of the beam inside samples were monitored perpendicularly to the direction of propagation and recorded using an optical microscope (Nikon Model Optiphot) coupled with a camera.

To make the comparison of data meaningful the samples in the form of oriented polished cubes with identical dimensions ($4 \times 4 \times 4$ $mm^3$) were fixed on a motorized table, and the change of samples during measurement was accomplished by moving the table in the plane perpendicular to the propagating beam. Thus, the position of the sample from the focus was kept constant for all the crystals. Selected structural and optical features of the crystals studied are summarized in Table 1.

## 3. Results and discussion

Except for the cubic GGG system the remaining investigated crystals were anisotropic and showed a more or less pronounced birefringence. It has been shown [26] that due to the occurrence of ordinary and extraordinary rays in birefringent crystals both the filamentation and spectral characteristics of SC depend on the angle between optical axis of the crystal and polarization of incident light pulse. In particular, in the case of uniaxial sapphire crystal the spectral width and intensity of blue side of SC spectrum was the highest when the angle between plane of polarization of the light pulse and the crystal axis was 0° or 90° [26]. Bearing this in mind, our samples were oriented in such a way that the beam propagated along the optical axis in LTO and LVO crystals and along the crystallographic b-axis (Y dielectric axis) in GSO and GCOB crystals.

Initially the pulse energy density on the surface of samples was varied in order to determine thresholds for irreversible damage of crystals. For this measurement the incident non attenuated beam was focused employing a lens with a focal length f = 150 mm and samples were located well before a focus, thereby they were illuminated by a weakly converging beam. Next, the distance between the focus and samples was decreased gradually and samples were examined carefully to reveal the occurrence of an internal or surface damage. It was observed that surface damage occurred incidentally for LVO and LTO samples when the diameter of light beam

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