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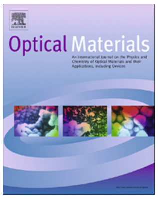

Spectral transformation of infrared ultrashort pulses in laser crystals

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ABSTRACT

The impact of peculiarities of transparent media on the self transformation of femtosecond pulses is considered. Spectral width and intensity distribution of anti-Stokes part of supercontinuum generated by infrared femtosecond pulses in selected single crystals were determined for different pulse energy density as a function of incident pulse wavelength changed in the 800–1600 nm region. Effect of material bandgap on the supercontinuum generation was examined based on experimental data for single crystals of gadolinium orthosilicate, gallium gadolinium garnet, lithium tantalate and lutetium vanadate. It was found that the anti-Stokes part of supercontinuum generated in Gd_2SiO_5 and $Gd_3Ga_5O_{12}$ crystals has a quite large spectral width with a short wavelength cut-off around 450 nm and contains a broad plateau.

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1. Introduction

The history of investigation related to the propagation and self-transformation of short laser pulses in transparent media is quite long. It began in the 1970 year, when Alfano and Shapiro demonstrated the transformation of picosecond laser pulses into a broad-band emission in glass samples [1]. Supercontinuum generation induced by femtosecond pulses has been observed for the first time in an ethylene glycol film in year 1983 [2] and in gases in year 1986 [3]. Numerous works on this subject that appeared during subsequent decade have been aimed at understanding the mechanisms of phenomena observed. Among these early works, a significant comprehensive investigation reported by Brodeur and Chin [4] has provided a comparison of experimental data acquired for various liquid and solid state media and an assessment of importance of particular mechanisms involved. Solid-state transparent media are of interest owing to their potential for application as sources of broad-band emission and parametric optical amplification. Materials investigated thus far encompass various glasses, among others BK-7 glass [6], SF10 and SF56 glasses [7], crystalline media as sapphire [8], barium fluoride BaF_2 [9], β-barium borate (BBO) [10] yttrium aluminum garnet (YAG), calcium fluoride (CaF_2) potassium gadolinium tungstate (KGW), gadolinium vanadate and yttrium vanadate [11]. Results reported in these papers have revealed that generalizations regarding continuum generation are not generally valid. In particular, it has been demonstrated that in contrast to predictions the continuum emission can be generated in media characterized by an energy gap significantly less than three times the incident photon energy. Spectral bandwidth of the Stokes emission and its energy distribution is not generally consistent with predicted exponential decrease with increasing wavelength. The threshold energy (power) for the continuum generation has been found to be strongly dependent on material peculiarities. Comparison of results gathered thus far is difficult since experimental conditions such as thickness of samples, focusing geometry, spatial location of the focus in the bulk sample, duration and energy of incident pulses, affect the phenomenon of the continuum generation. The “anomalies” mentioned above indicate that further investigation in this field is needed.

Intention of the present work is to get a more close insight into phenomena that accompany the propagation of ultrashort light pulse in inorganic crystals.

2. Theoretical background

When analyzing the experimental data Broader and Chin [4] were able to make generalizations about the supercontinuum generation, namely the occurrence of a bandgap threshold for continuum generation, an increase of continuum spectral width with increasing bandgap and a very strong Stokes–anti-Stokes asymmetry of the continuum. To account for these findings, not consistent with the theory of self-phase modulation (SPM), they proposed a mechanism involving a free-electron generation by multiphoton excitation (MPE). With this contribution the interplay between various mechanisms involved in the continuum generation is

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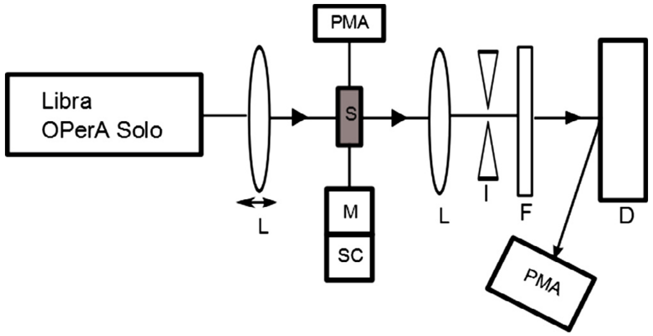
generally understood and can be summarized as follows. An ultra-short light pulse (in the range of tens or thousands of femtoseconds) propagating in an transparent medium is affected by an instantaneous Kerr nonlinearity that gives rise to temporal distribution of nonlinear phase denoted as the self-phase modulation (SPM) effect and the effect of self-focusing. Both these effects depend on 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 index of refraction induced by high energy density of electromagnetic field. The degree of spectral broadening of ultra-short pulses resulting from SPM has been analyzed theoretically in [5]. 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 on one hand and a plasma created induces a negative change in the index of refraction on the other hand. As a consequence, the self-focusing is arrested and a stable continuum generation involving a single filament can be obtained provided that the pulse energy is maintained below a break-down threshold for formation of multiple filaments. In real systems a competitive mechanism involving an avalanche ionization may provide an energy loss and should be considered, too. In this phenomenon, a seed electron arising from ionization of traps or impurities is promoted to the conduction band with an excess energy high enough to create a second free electron. Two free electrons thus created excite subsequent electrons and so on.

3. Experimental

Experiments were accomplished using a set-up depicted schematically in Fig. 1. Source of incident infrared light consists of femtosecond laser (Coherent Model "Libra") that delivers a train of 89 fs pulses at a centre wavelength of 800 nm and pulse energy of 1 mJ with a repetition rate regulated up to 1 kHz. To obtain light pulses at different wavelengths the laser is coupled to an optical parametric amplifier (Light Conversion Model "OPerA"). In this configuration the pulse wavelength can be varied between 230 and 2800 nm and the pulse energy is comprised between 6 and 150 μJ, depending on the spectral region. Excitation light was focused using a lens having a focal length of 30 cm. Location of the focus with respect to the sample was changed by moving the lens farther or closer to the sample. When a threshold for supercontinuum generation was exceeded the light emerging from a sample formed on a screen a uniformly illuminated circular picture, easily seen by a naked eye. With increasing energy density of the incident beam coloured rings due to the conical diffraction appeared around the central part of the beam. An iris was used to discriminate this unwanted phenomenon.



Libra: Ti:Al$_2$O$_3$ laser, 800 nm, 1kHz, 1mJ
OperA Solo: wavelength from 230 nm up to 2800 nm
S - Sample
L - lens f=30 cm
I - iris
F - filters
D - diffusing screen
M - grating spectrograph
SC - streak camera
PMA – photonic multichannel analyzer

Fig. 1. Experimental set-up.

Fundamental structural and optical features of the crystals investigated in this work are summarized below.

Gd$_2$SiO$_5$ (GSO) – monoclinic system, *P21/c* space group ($Z = 4$), $a = 9.1105$ Å, $b = 6.9783$ Å, $c = 6.8544$ Å, $\beta = 107.14$, Biaxial crystal with axis correlation: $Y||Y$, $(X, D_1) = 30.3°$ and $(Z, D_2) = 41.2°$. The set of refractive indices: $n_X = 1.871$, $n_Y = 1.884$ and $n_Z = 1.910$. Range of transparency: 0.21–5 μm. Deduced bandgap: 5.95 eV.

Gd$_3$Ga$_5$O$_{12}$ (GGG) – cubic system, *Ia−3d* space group ($Z = 8$), $a = 12.3829$ Å. The refractive index: 2.0 at the UV and 1.8 at the IR. The nonlinear refractive index n_2: 5.8×10^{13} cm^3 erg [12]. Range of transparency: 0.25–6.0 μm. Experimental bandgap: 5.33 eV [13].

LiTaO$_3$ (LTO) – trigonal/rhombohedral system, *R3c* space group ($Z = 6$), $a = 5.14$ Å, $c = 13.74$ Å). Uniaxial crystal. Refractive indices: $n_0 = 2.175$ and $n_e = 2.180$. Optical transparency: 0.28–5.5 μm. Deduced bandgap: 4.43 eV. The nonlinear refractive index n_2: 0.91×10^{-15} cm^2 W^{-1} at 1064 nm and 8.3×10^{-15} cm^2 W^{-1} at 532 nm (LNO [14]).

LuVO$_4$ (LVO) – tetragonal system (I4$_1$/amd space group, $Z = 4$) with $a = 7.0254$ Å and $c = 6.2347$ Å. Uniaxial crystals with refractive indices of $n_0 = 2.031$ and $n_e = 2.249$, and nonlinear refractive indices n_2 of 1.08×10^{-15} cm^2 W^{-1} ($E//c$) and 1.34×10^{-15} cm^2 W^{-1} ($E \perp c$) [15]. Optical transparency: 0.35–5.5 μm. Deduced bandgap: 3.60 eV.

4. Results and discussion

4.1. Spectral characteristics of supercontinuum generated by femtosecond pulses of infrared light at 800 nm

Almost all experimental data reported in literature on supercontinuum generation have been acquired upon illumination of samples with infrared pulses delivered by titanium–sapphire femtosecond lasers operating at or near 800 nm. In view of this, incident light pulses at 800 nm are employed at first aiming to consider the impact of crystalline media under study on spectral features of generated supercontinuum with a meaningful reference to the information gathered thus far in published works. In the following we will restrict our attention to the anti-Stokes part of supercontinuum stretching in the visible region of the spectrum. This restriction stems from sensitivity range of detection devices in our experimental set-up on one hand and from the fact that the generated supercontinuum can be easily observed by naked eye, on the other. When recording spectra the energy density in samples was varied by means of displacement of a focus of incident beam with respect to the sample. For all samples studied the spectra recorded were consistent with a characteristic behaviour established for other materials investigated thus far, namely the energy

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