Impact of femtosecond pulse duration on SRS amplification in KGW crystal

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Non-linear microscopy requires lasers operating within biological tissue's transparency window (1.1-1.3 µm) with minimal scattering losses [1-2]. However, the limited availability of femtosecond laser sources in this range prompts the utilization of non-linear light conversion methods. Stimulated Raman scattering (SRS) emerges as a solution by shifting wavelength within the Raman medium, thereby enabling compatibility with conventional lasers. Potassium gadolinium tungstate ($KGd(WO_4)_2$, KGW) crystal, with its unique properties including high Raman gain coefficient, large Raman shift, high damage threshold and moderate thermal conductivity [3], provides an exceptional platform for investigating and exploiting SRS phenomena.

In this study, we present numerical and experimental investigation of SRS amplification in KGW crystal, demonstrating efficient amplification of the seed with both transform-limited to 300 fs and chirped to 1 ps, 1030 nm pulses.

The numerical model of the Raman amplifier is based on semi classic approach of stimulated Raman scattering in transient regime. Slowly varying envelope approximation (SVEA) and flat-wave approximation are used for this purpose. Therefore, a spatial distribution of individual optical fields was neglected. The intensity was averaged over the transversal dimensions corresponding to Gauss beam propagation of both fields. The original set of equations was extended by terms describing effects of group velocity dispersion. The higher order dispersion effects were neglected. Linear and nonlinear absorption, as well as other nonlinear effects, are not considered within the model framework. The Fourier split-step method (Type I) is employed for solving the stimulated Raman scattering equations.

The Raman gain of each spectral component, and thus the excitation of the vibrational amplitude, depends on the resonance detuning due to the width of the Raman line. Assuming fundamental wave has a spectral profile $P(\omega)$ with the center frequency ω_P and dephasing time T_2 (2 ps) of Raman mode corresponds to spectral line assuming Lorentz line shape with a width of ω_R (5.4cm^{-1}), the spectrally resolved Raman gain is given by formula:

$$g(\omega - \omega_R) = g_0 R(\Delta) * P(\omega), \quad (1)$$

where $R(\Delta)$ is a Raman line as a function of detuning Δ and g_0 (3 cm/GW) is a steady state Raman gain. Raman line shape R is normalized to a peak value of 1, spectral curve of pump P is normalized to area of 1. The convolution operator * denotes spectral convolution, indicating that peak value of g corresponding to case of resonance causes a spectral narrowing of amplified Stokes spectrum.

For experimental validation of SRS amplification with different pulse types, an experimental setup was built as shown in Fig. 1. The laser (FemtoLux 30, Ekspla) operated at 800 kHz repetition rate, emitting 40 µJ, 1030 nm, 300 fs pulses with the possibility to chirp pulses up to 1 ps. The seed for an SRS amplifier was generated with a small fraction (350 nJ for 300 fs pulses and up to 1 µJ for chirped pulses) of the total energy by supercontinuum (SC) generation. For SC generation input beam with a diameter of 2.6 mm (at the 1/e^2 intensity level) was focused with a fused silica lens (f =+50 mm) onto the front face of uncoated and undoped 4 mm long KGW crystal. The rest part of energy was used to pump 10 mm length undoped KGW crystal for SRS amplification. Pump energy was controlled with attenuator which consist thin film polarizer and half-wave retardation plate. The seed and pump pulses were temporally synchronized using a delay line and spatially aligned for non-collinear interaction within a KGW crystal. The delay and small angle between (~0.6°) pump and seed pulses was adjusted to achieve the maximum gain in SRS amplifier with different type of pulses. The SRS amplification was characterized by measuring two spectra: the initial seed spectrum and the spectrum after amplification. By dividing the amplified spectra by the seed spectra, gain spectra were computed, revealing the gain of each spectral component. Subsequently, both spectra were integrated, and the ratio of the integrated spectra determined the amplification factor.

Input pulses duration was systematically adjusted from transform-limited 300 fs to chirped pulses up to 1 ps and used for SC generation and SRS amplification. Main SRS amplification results are shown in Fig. 2. Increasing the pump energy resulted in exponential growth of the SRS amplification. Conversely, stretching the pulse duration by applying negative chirp to the initial pulses led to a reduction of pump intensity, thereby decreasing the amplification factor. Meanwhile, the use of shorter pulses (300 fs) induced self-phase modulation (SPM) in the KGW crystal, resulting in a broadening of the pump spectra and subsequent decrease in pump intensity. SPM limited using of higher pump energies with transform-limited pulses and amplification did not match modelling results. Highest amplification factor (over 100) was achieved with chirped pulses up to 700 fs, where the highest pump intensity without SPM was retrieved.

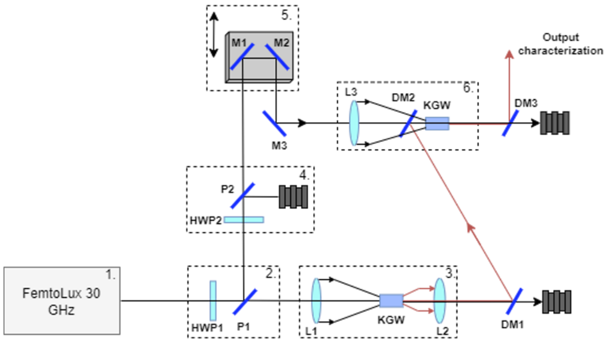

Fig. 1 Experimental setup main parts: 1. Laser source 2. Polarizing beam splitter 3. Seed generation (SC) for SRS amplification 4. Attenuator 5. Delay line 6. SRS amplifier

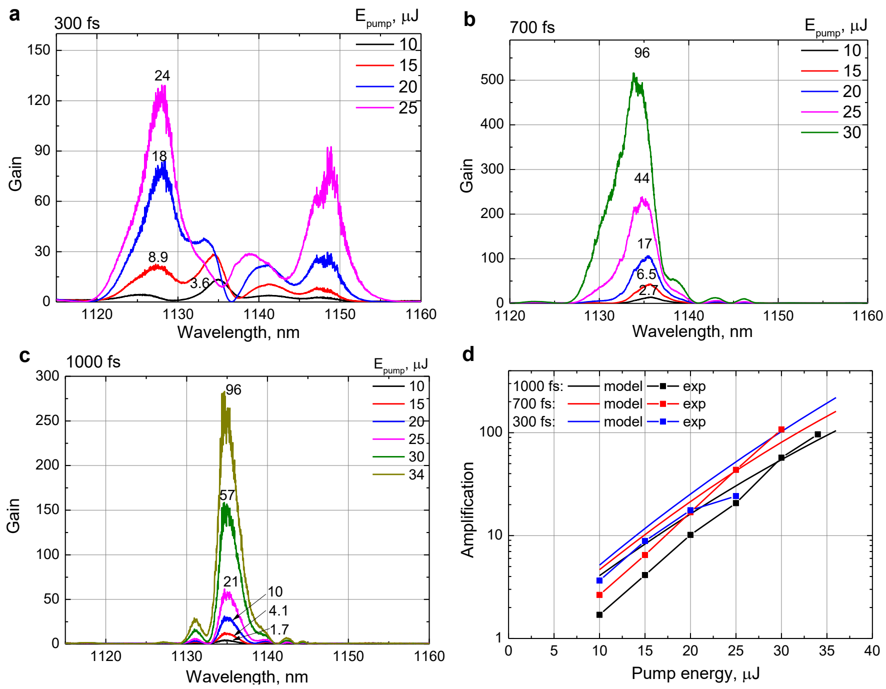

Fig. 2 SRS amplification results: a-c gain spectrum (number indicating amplification) dependence on pump energy for different pulses: a) 300 fs, b) 700 fs, c) 1 ps and d) amplification comparison with modelling results.

At low pump energies and chirped femtosecond pulses the results agree with the model and the main difference is not accounted optical losses. This suggests that in such a regime the peak intensity of the pump pulses is the main factor of amplification. However, for transform-limited and high energy pulses, experimentally observed self-phase modulation (SPM) deviates from theoretical predictions. These findings from both experimental results and numerical modeling provide valuable insights for future work to optimize and develop the compact and efficient laser source for biological imaging applications. More information and results about experimental setup will be presented at the conference.

References

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