Raman random lasing in Ba(NO$_3$)$_2$ powder

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Abstract. Random lasing in Raman active material – barium nitrate powder – has been registered in two temporal regimes: nanosecond and picosecond. Stimulated Raman scattering (SRS) in Ba(NO$_3$)$_2$ for picosecond excitation has been shown to have much lower threshold and spectrum containing more components than for nanosecond excitation opposite to the case of a bulk medium. In picosecond regime SRS intensity increased with temperature decreasing and redistributed in favor of the higher order components. SRS pulse duration in picosecond regime was estimated to be in the range 16.5-22 ps. High conversion efficiency of SRS in barium nitrate powder in picosecond regime and its larger stability under laser impact than for bulk samples gives possibility to use it as an active material in Raman converters.

1. Introduction

Random lasing (RL) effect was for the first time described by Letokhov [1] in 1968. Random lasers are systems using highly disordered materials to obtain laser action. In these systems optical path is much longer than in bulk material and optical gain provides amplification that starts lasing. Random lasers continue to attract attention due to numerical practical applications and are actively studied till now [2–4]. Random lasing has been realized and investigated in a lot of different materials: powders of rare earths and semiconductors [5, 6], biological tissues [4], nanocomposites including nano or submicron particles [8]. Raman random laser is a special case of random laser, which uses stimulated Raman scattering (SRS) as a gain source [9, 10]. In Raman random laser stimulated inelastic scattering plays the role of a gain source while elastic scattering returns the radiation back into the media. The efficiency of the SRS process in dispersive media can be significantly increased due to the increasing of the interaction length in the process of the light diffusive propagation in such media. In addition, the local field effect in such media can also lead to a significant increase in efficiency of the SRS process. Taking into account all of the above, as well as the ease of production and high radiation resistance, disordered Raman active media are of great interest for various fields of photonics.

In our work we studied SRS in random material – barium nitrate Ba(NO$_3$)$_2$ powder. Spectrum of this material contains line, corresponding to the "breathing" mode with wavenumber $\nu(A_g) = 1047$ cm$^{-1}$, linewidth equal to 1.5 cm$^{-1}$, high gain coefficient ($g = 47\pm5$ cm/GW) and low SRS threshold. Due to slow relaxation of the vibrational excitations in Ba(NO$_3$)$_2$ (~30 ns) in the case of nanosecond excitation...
in barium nitrate crystals the SRS threshold is an order of magnitude less than for excitation by picosecond pulses [11]. Using excitation sources with different pulse duration we could study both steady-state and transient modes for the random media case.

2. Experimental

Ba(NO$_3$)$_2$ is characterized by a low moisture resistance and poor heat conductivity. Moderate mechanical properties of Ba(NO$_3$)$_2$ crystals can lead to its destruction in the process of pumping so using of this material in powder form could solve such problems. SRS spectra in barium nitrate powder for the first time were obtained in 1967 [12]. Later Ba(NO$_3$)$_2$ powder was used as disordered material for Raman random lasing effect study [13, 14].

In the SRS process two temporal regimes are possible: the first one is a steady state regime, when the pump pulse duration is much longer than the vibronic Raman mode dephasing time. The second case is a transient regime, when the pump pulse duration is smaller than the dephasing time and spectral width of pump laser is much broader than the Raman line homogeneous broadening.

To study SRS in barium nitrate powder in both temporal regimes we used two pumping sources: second harmonic of Q-switched Nd:YAG laser ($\lambda = 532$ nm, $\tau = 11$ ns, $E = 0.2$ J, frequency 10 Hz) and second harmonic of Nd:YAG mode-locked laser ($\lambda = 532$nm, $\tau = 30$ ps $E = 25$ mJ, frequency 10 Hz). The Ba(NO$_3$)$_2$ powder of micron-sized particles was placed in the cell. Thickness of the sample was 3 mm. Exciting laser beam was focused on the sample by the lenses with different focuses. Changing focus length and using filters we could change the power density of the exciting light on the sample. Stimulated scattering processes can depend on the sample temperature; therefore, we studied the effect of temperature on the SRS in random material. For temperature control a thermocouple was installed into the cell with a sample. Liquid nitrogen was used for cooling. A portable spectrometer has been used for SRS registration. When the pump energy overcomes the threshold value the random Raman lasing starts due to the excess of a gain over losses in disordered media. Both in picosecond and in nanosecond excitation we registered SRS in barium nitrate powder corresponding to the SRS-active mode at 1047 cm$^{-1}$ due to the internal totally symmetric vibrations of the quasi molecular NO$_3$ group.

At room temperature for picosecond pulses excitation we registered two Stokes and one anti-Stokes components. Thresholds of the components observation were 0.065 GW/cm$^2$, 3.5 GW/cm$^2$ and 14 GW/cm$^2$ for 1$^{st}$ Stokes, 2$^{nd}$ Stokes and anti-Stokes components correspondingly. For nanosecond pulses excitation at room temperature we registered only one Stokes component. Its observation threshold was 4 GW/cm$^2$.

The first Stokes component’s intensity dependence on the pump energy at room temperature for both temporal regimes is shown in figure 1.

![Figure 1. 1$^{st}$ Stokes component energy dependence on the pump pulse energy for picosecond and nanosecond excitation. 1 – picosecond excitation 2 – nanosecond excitation](image-url)
The threshold value for picosecond (transient) mode is lower than for nanosecond one opposite to the case of a bulk medium. The reason of this behaviour is the fact that virtual states involved in Raman transitions are much faster than electronic transitions connected with real atomic or molecular levels. Besides that, stay of photons in the dispersed medium due to multiple scattering is longer than in bulk material. These effects lead to the slow relaxation of the vibrational excitation in Ba(NO₃)₂ powder.

In the case of picosecond pumping pulse duration we studied temporal dynamics of SRS with the help of Fabry-Perot (F-P) interferometer with a variable distance between the mirrors. If pulse duration \( \tau_p \) is shorter than the delay time of the F-P \( \tau = 2d \cos \theta / c \) (\( d \)-separation between the mirrors), there is no overlapping and hence no interference effect, otherwise \( (\tau_p > \tau) \) we can see an interference (figure 2) [15]. So, varying the distance between mirrors it is possible to estimate the pulse duration. Interferograms, obtained at room temperature and at liquid nitrogen temperature with different distance between the mirrors in the case of picosecond excitation are presented in figure 2.

Duration of SRS pulse is estimated to be in the range 16.5÷22 ps. The case when the duration of the radiation pulse is shorter than the pump pulse for the picosecond regime is confirmed by the Monte Carlo method for conventional random lasers [16]. When \( d = 4 \) mm the interference pattern was absent at room temperature and appeared at the temperature of liquid nitrogen. So, increasing duration with temperature decreasing was observed.

Temperature investigations of the SRS in barium nitrate showed that SRS threshold do not depends significantly on the sample temperature. SRS intensity increased with temperature decreasing and redistributed in favor of higher order components. Temperature dependence of SRS components intensity is shown in figure 3.

![Figure 2. Fabry-Perot interferograms for pump and 1st Stokes component.](image)

![Figure 3. Temperature dependences for all the SRS components in Ba(NO₃)₂ for nano- and picosecond excitation.](image)
Similar temperature dependence of the SRS components intensity has been observed in calcite [17]. Investigations of the spatial distribution of emission on the sample surface for pump radiation elastic scattering, and for the 1st Stokes component showed that above the threshold speckle structure appeared in the SRS beam which is typical for random lasing effect and is characteristic of the source coherence [18, 19].

3. Conclusions
In our work we showed that SRS in Ba(NO$_3$)$_2$ powder can be excited both by nanosecond and picosecond laser pulses. SRS corresponding to the transient mode (picosecond regime) is much more effective than in steady-state mode (nanosecond regime). SRS threshold for picosecond regime is much lower than in nanosecond and more SRS components can be excited. SRS intensity dependence on the exciting light intensity is sharper in picosecond regime than in nanosecond. Temperature decreasing leads to the SRS intensity increasing especially in picosecond mode. We also observed SRS properties characteristic for random lasing. Thus, our experiments showed that disordered materials can be successfully used in Raman converters.

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