Laser Exciting of Bound Photonic States and High Frequency Gravitational Waves in Media during Combinational Light Processes

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Abstract. Multiple Stokes and anti-Stokes satellites (Comb Parametric Raman Processes) were observed in liquids and crystals when excited by picosecond laser pulses of a YAG:Nd$^{3+}$ solid-state laser with generation wavelengths of 1064 and 532 nm. The Stokes combination satellites of the infrared range were detected as a result of their conversion into the visible spectral region. The bound two-photonic states, corresponding to high frequency gravitational waves, in media due to strong photon-photon interactions is predicted.

1. Introduction
After the appearance of intense laser light sources, the phenomenon of stimulated Raman scattering (SRS) of light in various dielectric media was discovered [1,2]. In contrast to spontaneous Raman scattering (RS), in spectrum of SRS a sharp increase in the intensity of one of the strongest spontaneous Stokes RS line was observed. With a further increase of the spectral intensity of the exciting laser radiation, the several Stokes and anti-Stokes satellites in liquids or crystals appear. Its intensity becomes close to the intensity of the exciting laser radiation [2–4]. Thus Combinational Parametric Raman Scattering (CPRS) processes take place. In this case the simultaneous destruction of two quanta of exciting radiation and Stokes and anti-Stokes pairs birth [5–7] is realized. At the first stage of SRS researches the giant pulses of ruby or yttrium aluminum garnet lasers with a duration of about 10 ns and sufficiently high energy in each pulse (0.1–1 J) as the source of exciting light have been used. When such radiation was strongly focused on a dielectric medium, the destruction of the samples took place, preventing the possibilities of CPRS observing in many media. Recently multi-frequency Raman scattering processes were observed in many liquids [8,9] and crystals [10–12] under picosecond laser emission.

Parametric excitation of high frequency gravitational waves in cosmic objects has been predicted in several works [13-16]. We have analyzed the opportunity to observe the resemble effect in dielectric media on the base of CPRS. If the frequency of exciting laser emission is $\omega_0$, the creation of scalar bound two-photonic state with frequency ($2\omega_0$) in dielectric media is waiting. Such bound states correspond to high frequency scalar gravitational wave. The conditions for bound two-photonic states appearing in dielectric media are the strong photon-photon interaction by means of virtual phonons exchange between interacting photons during CPRS.
High quality Q-factor bound two-photonic states may exist at the presence of Fermi Resonance. In this case, the scalar exciton states with energy, close to the bound two-photonic states with frequency are located in spectra of media. Elementary excitations of vacuum, corresponding to bound two-photonic states, is known as paraphotons or hidden photons [17-20]. For detection of high frequency scalar gravitational waves, i.e paraphotons, the same media, in which bound two-photonic states were excited, should be used.

In this paper the experimental results of CPRS spectra recording in various condensed dielectric media (CaCO$_3$, quartz, NaBrO$_3$, Ba(NO$_3$)$_2$ and others), excited by ultra short (60-80 ps) YAG:Nd$^{3+}$ laser pulses, are presented. The different experimental schemes for the generation and detection of scalar high frequency gravitational waves and paraphotons during CPRS is discussed

2. Experimental technique

The investigations of CPRS processes in various condensed dielectric media excited by ultrashort (60-80 ps) YAG:Nd$^{3+}$ laser pulses have been made. The second optical harmonic of a YAG:Nd$^{3+}$ laser with an emission wavelength $\lambda = 532$ nm and the main generation line of this laser with a wavelength $\lambda = 1064$ nm for the excitation of the SPRS spectra in crystals (Figure 1) and liquids (Figure 2) have been used.

![Figure 1. Schematic diagram of an experimental setup for recording the SPRS spectra in condensed media when the second optical harmonic of a YAG:Nd$^{3+}$ laser is excited ($\lambda = 532$ nm); 1 — YAG:Nd$^{3+}$ laser; 2 — lens; 3 — nonlinear optical crystal; 4 — laser emission; 5 - fiber tip; 6 — quartz fiber; 7 — mini spectrometer; 8 — computer.](image1)

![Figure 2. Schematic diagrams of experiments for the excitation of SPRS by means of YAG:Nd$^{3+}$ laser in liquids; 1 — laser radiation source; 2 — lens; 3 - cuvette with liquids; 4 - laser emission; 5 - fiber tip; 6 — notch filter; 7 — mini spectrometer; 8 - computer.](image2)
a repetition frequency of 10 Hz with an average power of 10-100 mW and an energy in each pulse of 1-20 mJ. Rapid registration of the spectra of the SPRS was carried out with a compact FSD-8 fiber-optic spectrometer with a multi-element receiver providing digital processing of the spectra in the range of 200-1100 nm with a resolution about 1 nm.

3. Experimental results and discussion

Liquids (light and heavy water, ethanol, glycerin) and single crystals (barium nitrate, calcite, sodium bromate, KGW) as well as crystalline powders (LiOH, LiOD) were investigated. Equidistant frequency combs were detected in the form of a large number (2-8) of Stokes and anti-Stokes satellites, extending from the far infrared region to the ultraviolet range (see Figure 3, a-f). In accordance with the synchronization conditions for the elementary processes of the four-particles SPRS, the energy and quasimomentum conservation laws must be satisfied. In the simplest case of the decay of two quanta of exciting radiation into the corresponding Stokes and anti-Stokes components, we have:

$$2\omega_L = \omega_{1S} + \omega_1A; \quad 2k_L = \vec{k}_{1S} + \vec{k}_{1A};$$
$$2\omega_L = \omega_{2S} + \omega_2A; \quad 2k_L = \vec{k}_{2S} + \vec{k}_{2A};$$
$$2\omega_L = \omega_{3S} + \omega_3A; \quad 2k_L = \vec{k}_{3S} + \vec{k}_{3A};$$
$$2\omega_L = \omega_{4S} + \omega_4A; \quad 2k_L = \vec{k}_{4S} + \vec{k}_{4A}. \quad (1)$$

The conditions of synchronism for another processes take the form:

$$2\omega_L = \omega_{1S} + \omega_L; \quad 2k_1S = \vec{k}_{2S} + \vec{k}_L;$$
$$2\omega_L = \omega_{2S} + \omega_1S; \quad 2k_2S = \vec{k}_{3S} + \vec{k}_{3A};$$
$$2\omega_L = \omega_{3S} + \omega_2A; \quad 2k_3S = \vec{k}_{4S} + \vec{k}_{2A};$$
$$2\omega_L = \omega_{4S} + \omega_A; \quad 2k_4S = \vec{k}_L + \vec{k}_{2A};$$
$$2\omega_2A = \omega_1A + \omega_3A; \quad 2k_2A = \vec{k}_{1A} + \vec{k}_{3A};$$
$$2\omega_3A = \omega_2A + \omega_4A; \quad 2k_3A = \vec{k}_{2A} + \vec{k}_{4A}. \quad (2)$$

The SPRS was observed in ethanol by pumping radiation from a second-harmonic of a YAG:Nd$^{3+}$ laser (see Figure 3(a,b)). For the “forward scattering” (see Figure 3a), one anti-Stokes and two Stokes lines were recorded with an average frequency shift is $\Delta \nu = 2923$ cm$^{-1}$. From Figure 3(a), it can be seen that the half width of the blue anti-Stokes mode is much larger than that of the first and second Stokes lines. The registration of the SPRS spectrum with the reflection (backward scattering) geometry (see Figure 3(b)) leads to a change in the number and shape of the observed lines. The first Stokes line in Figure 3(b) has more half width than in Figure 3(a) as a result of the formation of a two-component band with frequencies of 2836 and 2921 cm$^{-1}$ instead of one narrow line at a frequency of 2947 cm$^{-1}$ (see Figure 3(a)). On both sides of the high-intensity first and second Stokes satellites, the low intensity wings were detected (see Figure 3(b)).

Excitation of the calcite single crystal by laser radiation with a wavelength of $\lambda = 532$ nm leads to the generation of SPRS with four Stokes and three anti-Stokes Raman satellites in the visible and near-infrared ranges (see Figure 3(c,d)). All observed lines belong to the full-dimensional oscillation $A_{1g}$ with a frequency shift $\Delta \nu = 1086$ cm$^{-1}$.

When a SPRS was excited in a Ba(NO$3$)$_2$ crystal by a picosecond pulsed YAG:Nd$^{3+}$ laser with a pump wavelength of $\lambda = 1064$ nm, a large number of combinational satellites were recorded (see Figure 3(e,f)), located in a wide spectral range, in the near-IR and visible regions of the spectrum. According to Figure 3(e,f) in the spectrum of multifrequency SPRS in barium nitrate crystal, the eight anti-Stokes components appeared with the frequency shift $\Delta \nu = 1047$ cm$^{-1}$ (taking into account the measurement error of $\pm 50$ cm$^{-1}$). This corresponds to fully symmetric
Figure 3. Normalized spectra of SPRS in ethanol (a,b), calcite (c,d), excited by the second optical harmonic of a YAG:Nd$^{3+}$ laser with an emission wavelength of $\lambda = 532$ nm, and a barium nitrate crystal (e,f) during the excitation of secondary radiation by YAG:Nd$^{3+}$ laser line with a wavelength $\lambda = 1064$ nm.

internal vibrations of nitrate ions [NO$_3$]. In multifrequency SPRS processes, each anti-Stokes component (see Figure 3(e,f)) corresponds to some Stokes satellite, placed in the infrared region of the spectrum.

Figure 4 shows the SPRS spectra of barium nitrate and glycerol obtained during the experiment on a device with a frequency doubler of scattered radiation The spectrum of SPRS of barium nitrate in Figure 4(a) consists of four anti-Stokes lines, obtained as a result of the sample being excited by a laser source, and combination components, which appeared as a result
Figure 4. The spectra of the SPRS in barium nitrate (a) and glycerol (b), excited by the main YAG:Nd\(^{3+}\) laser line with an emission wavelength of \(\lambda = 1064\) nm with conversion of infrared Stokes modes to the visible region of the spectrum.

As a result, modes were detected corresponding to twice the frequencies of the first (\(\nu = 7250\) cm\(^{-1}\)) and the second (\(\nu = 5170\) cm\(^{-1}\)) Stokes lines, which are in the infrared region. As a result of the scattered radiation passing through the frequency doubler, additional combination satellites are formed, corresponding to the combination of exciting radiation with the first Stokes line (\(\nu = -8282\) cm\(^{-1}\)) and the sum of the frequencies of the first and second Stokes (\(\nu = -6208\) cm\(^{-1}\)) components of the spectrum. Figure 4(b) shows the spectrum of the SPRS of glycerol with the conversion of the first Stokes (\(\nu = -6448\) cm\(^{-1}\)) component to the red region of the visible range. Thus, the SPRS satellites have been registered from the IR range.

The excitation of a large number of Stokes and anti-Stokes components (see Figure 3) when pumping single crystals and liquids with ultrashort pulsed laser radiation in the visible and near infrared ranges provides the possibility of obtaining lasing frequency bands in a wide spectrum: from far infrared to ultraviolet. Spare biphoton Stokes-antiStokes states, emerging during SPRS, may be transformed into bound photonic states. Such result is predicted by the theory [21,22] under conditions of the strong photon-phonon interaction, if the frequency of exciting laser emission is \(\omega_0\), the created scalar bound two-photonic state is described as scalar gravitational
Figure 5. Schematic diagram of an experimental setup for recording of high frequency gravitational waves in the case SPRS processes in crystals; 1 — YAG:Nd<sup>3+</sup> laser, 2 — lenses, 3 — nonlinear crystal; 4 — laser emission; 5 — opaque stopper, 6 — fiber tip, 7 — quartz fiber; 8 — mini spectrometer; 9 — computer.

Figure 6. Schematic diagram of an experimental setup for recording of high frequency gravitational waves in the case SPRS processes in liquids; 1 — YAG:Nd<sup>3+</sup> laser, 2 — lenses, 3 — nonlinear liquids; 4 — laser emission; 5 — opaque stopper, 6 — fiber tip, 7 — quartz fiber; 8 — mini spectrometer; 9 — computer.

wave with frequency 2ω<sub>0</sub>. Thus in this nonlinear processes the high frequency gravitational waves may be excited in dielectric media. The preferable conditions for bound two-photonic states existence are the Fermi resonance presence. In this case, the scalar exciton states with energy, close to the bound two-photonic states with frequency 2ω<sub>0</sub>, are in electronic spectra of media. After bound two-photonic states creating in media due to pulsed laser excitation, the corresponding scalar gravitational waves should propagate in vacuum (spare space). Elementar excitations of such waves is known as paraphotons or hidden photons. For detection of high frequency scalar gravitational waves, the same media (in which bound two-photonic states were excited) is proposed in the experimental schemes, presented at Figures 5,6.

4. Conclusion
The experimental results of investigations of CPRS in various condensed dielectric media (CaCO<sub>3</sub>, quartz, NaBrO<sub>3</sub>, Ba(NO<sub>3</sub>)<sub>2</sub> and others), excited by ultra short (60-80 ps) YAG:Nd<sup>3+</sup> laser pulses, are presented. The second optical harmonic of a YAG:Nd<sup>3+</sup> laser with wavelength λ = 532 nm as well as the laser emission with a wavelength λ = 1064 nm were very effective for the excitation of the CPRS spectra in discussed dielectric media. The opportunity of high frequency gravitational wave generation and detection in dielectric media was analyzed.
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