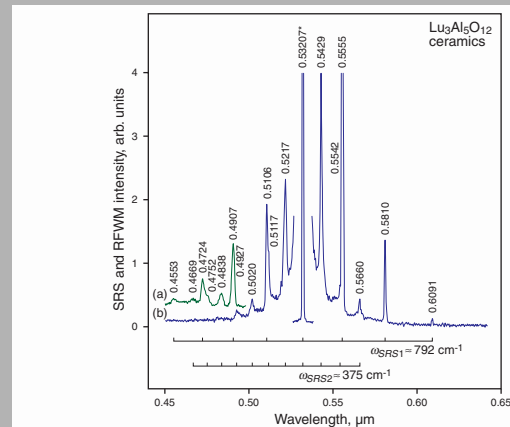


Abstract: In this paper are present results of the first investigation of stimulated Raman scattering (SRS) in the “garnet” $\text{Lu}_3\text{Al}_5\text{O}_{12}$ crystalline ceramics, which is promising host-material for activator lasants. With picoseconds laser excitation at $0.53207 \mu\text{m}$ wavelength was observed and analyzed high-order Stokes and anti-Stokes generation related to two $\chi^{(3)}$ -promoting vibration modes (≈ 792 and $\approx 375 \text{ cm}^{-1}$) of this novel SRS-active oxide material. A brief review of the pioneering papers on lasers based on $\text{Lu}_3\text{Al}_5\text{O}_{12}:\text{Ln}^{3+}$ single crystals and known SRS-active crystalline “garnet” materials is given in tabular form as well.



Fragments of SRS and RFWM spectrum of crystalline “garnet” ceramics $\text{Lu}_3\text{Al}_5\text{O}_{12}$ recorded at room temperature with picosecond pumping at the wavelength $\lambda_p = 0.53207 \mu\text{m}$

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Stimulated Raman scattering in “garnet” $\text{Lu}_3\text{Al}_5\text{O}_{12}$ ceramics – a novel host-material for Ln- and TM-lasant ions

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1. Introduction

Crystalline garnet-type (space group $O_h^{10} - Ia\bar{3}d$, no. 230) host-materials (single crystals and ceramics) for lanthanide (Ln) and transition-metal (TM) lasant ions form the most representative class of active media among of all known solid-state crystalline materials generating stimulated emission (SE). Currently, according to a recent review [1], more than 30 well-known oxide and fluoride gar-

net hosts together with SE-active Ln- and TM-activator ions represent about 100 laser crystals (crystalline host + certain lasant ions). These garnets are characterized by a variety of spectral-generation properties, among them the single-centered and multicentered, as well as the garnets with a partially disordered crystal-field of their lasant ions (mainly solid solutions). Structural possibilities of the garnet crystalline materials hit imagination, their c-

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Lasing Ln ³⁺ -ion	Ceramics	Single crystal ^{a)}		
		J' → J' SE channel (operating scheme, pumping) ^{b)}	Temperature, K	Co-activator ^{c)}
Nd ³⁺		⁴ F _{3/2} → ⁴ I _{9/2} (q-4-level, Xe) [11, 12]	77	
	^{d)}	⁴ F _{3/2} → ⁴ I _{11/2} 4-level, Xe, LD) [5, 11–14]	77–900 (Xe), 300 (LD)	
		⁴ F _{3/2} → ⁴ I _{13/2} (4-level, Xe, LD) [6, 7, 11, 15]	77, 300	
Ho ³⁺		⁵ S ₂ → ⁵ I ₅ (4-level, Xe) [16]	≈ 110	
	^{d)}	⁵ I ₇ → ⁵ I ₈ (q-4-level, Xe, LD) [8, 17–20]	77 (Xe), 300 (LD)	Er ³⁺ , Tm ³⁺ , Yb ³⁺
		⁵ I ₆ → ⁵ I ₇ (4-level, Xe) [21–24]	300	Yb ³⁺
		⁵ I ₆ → ⁵ I ₈ (q-4-level, Xe) [16, 23]	≈ 110	
Er ³⁺		⁴ S _{3/2} → ⁴ I _{13/2} (4-level, Xe) [21–23]	77, 300	
	^{d)}	⁴ I _{13/2} → ⁴ I _{15/2} (q-4-level, Xe) [8]	77	
		⁴ S _{3/2} → ⁴ I _{9/2} (4-level, Xe) [21–23]	300	
		⁴ I _{11/2} → ⁴ I _{13/2} (4-level, Xe) [21–27] (f-fl, Xe) [28]	300	Ho ³⁺ , Tm ³⁺ , Yb ³⁺ , Cr ³⁺
Tm ³⁺		³ H ₄ → ³ H ₆ (q-4-level, Xe, LD) [8, 29]	77 (Xe), 300 (LD)	
Yb ³⁺	^{d)}	² F _{5/2} → ² I _{7/2} (q-4-level, Xe, LD) [30–34]	77–175 (Xe), 77–313 (LD)	Nd ³⁺ , Cr ³⁺
Er ³⁺ +Tm ³⁺		⁴ I _{11/2} → ⁴ I _{13/2} (Er ³⁺) ~> ³ H ₄ → ³ H ₆ (Tm ³⁺) (cr-casc, Xe) [35, 36]	≈ 110	
Tm ³⁺ +Ho ³⁺		³ H ₄ → ³ H ₅ (Tm ³⁺) ~> ⁵ I ₇ → ⁵ I ₈ (Ho ³⁺) (cr-casc, Xe) [37]	≈ 110	Cr ³⁺

^{a)} In tables and text of the article we used citations only of publications from refereed journals.

^{b)} The notations here are: q-4-level is the quasi-four-level operating laser scheme; Xe is the Xe-flashlamp pumping; 4-level is the four-level operating laser scheme; LD is the laser-diode pumping; f-fl is the feed-flowing operating laser scheme; cr-casc is the cross-cascaded operating laser scheme; wave arrow indicates the non-radiative intermanifold transition (for details, see [2]).

^{c)} Here Ln³⁺ co-activators used as the sensitizing or deactivating ions that improve conditions for the occurrence of laser action.

^{d)} One can predict that SE generation at wavelengths of this intermanifold laser transition will be obtained in one of the first.

Table 1 Intermanifold (J' → J') SE transition of Ln³⁺ ions in Lu₃Al₅O₁₂:Ln³⁺ single crystals and some conditions and operating regimes of their laser oscillations, as well as the expected lasing in crystalline “gemstone” Lu₃Al₅O₁₂:Ln³⁺ ceramics

dodecahedral (D₂), *a*-octahedral (C_{3i}), and *d*-tetrahedral (S₄) crystallographic positions may occupy unlike co activator ions with different valences from (1⁺) to (6⁺) (see Table 1.15 in [2]). For many years, and today the most used laser garnets are Y₃Al₅O₁₂:Ln³⁺ (see, e.g. [3, 4]). Some of them, especially Y₃Al₅O₁₂:Nd³⁺ are commercial for a long time. Second in richness of spectral-generation properties are the laser garnets Lu₃Al₅O₁₂:Lu³⁺ [1].

They were introduced almost 40 year ago [5–8]. First among these were the Lu₃Al₅O₁₂ crystal doped with laser ions Nd³⁺ [5], Ho³⁺, Er³⁺, and Tm³⁺ [8]. To excite their pulse SE generation in those early years were used Xe-flashlamps as the pump sources. Following lasing Ln³⁺-ion in Lu₃Al₅O₁₂ crystal was Yb³⁺ [9, 10]. In last years this laser crystal have received substantial attention due to its high quantum efficiency (very

Stokes and anti-Stokes $\chi^{(3)}$ -lasing components			SRS-promoting vibration mode, cm ⁻¹	
Wavelength, μm^a	Line	SRS and RFWM attribution ^{b)}	ω_{SRS1}	ω_{SRS2}
0.4553	ASt ₄₋₁	$^*\omega_p + 4\omega_{SRS1} = [(\omega_p + 3\omega_{SRS1}) + (\omega_p + 3\omega_{SRS1}) - (\omega_p + 2\omega_{SRS1})] = 2\omega_{AST3-1} - \omega_{AST2-1} = \omega_{AST4-1}$	≈ 792	
0.4669	ASt ₇₋₂	$^*\omega_p + 7\omega_{SRS2} = [(\omega_p + 6\omega_{SRS2}) + (\omega_p + 6\omega_{SRS2}) - (\omega_p + 5\omega_{SRS2})] = 2\omega_{AST6-2} - \omega_{AST5-2} = \omega_{AST7-2}$		≈ 375
0.4724	ASt ₃₋₁	$^*\omega_p + 3\omega_{SRS1} = [(\omega_p + 2\omega_{SRS1}) + (\omega_p + 2\omega_{SRS1}) - (\omega_p + \omega_{SRS1})] = 2\omega_{AST2-1} - \omega_{AST1-1} = \omega_{AST3-1}$	≈ 792	
0.4752	ASt ₆₋₂	$^*\omega_p + 6\omega_{SRS2} = [(\omega_p + 5\omega_{SRS2}) + (\omega_p + 5\omega_{SRS2}) - (\omega_p + 4\omega_{SRS2})] = 2\omega_{AST5-2} - \omega_{AST4-2} = \omega_{AST6-2}$		≈ 375
0.4838	ASt ₅₋₂	$^*\omega_p + \omega_{SRS2} = [(\omega_p + 4\omega_{SRS2}) + (\omega_p + 4\omega_{SRS2}) - (\omega_p + 3\omega_{SRS2})] = 2\omega_{AST4-2} - \omega_{AST3-2} = \omega_{AST5-2}$		≈ 375
0.4907	ASt ₂₋₁	$^*\omega_p + 2\omega_{SRS1} = [(\omega_p + \omega_{SRS1}) + (\omega_p + \omega_{SRS1}) - \omega_p] = 2\omega_{AST1-1} - \omega_p = \omega_{AST2-1}$	≈ 792	
0.4927	ASt ₄₋₂	$^*\omega_p + 4\omega_{SRS2} = [(\omega_p + 3\omega_{SRS2}) + (\omega_p + 3\omega_{SRS2}) - (\omega_p + 2\omega_{SRS2})] = 2\omega_{AST3-2} - \omega_{AST2-2} = \omega_{AST4-2}$		≈ 375
0.5020	ASt ₃₋₂	$^*\omega_p + 3\omega_{SRS2} = [(\omega_p + 2\omega_{SRS2}) + (\omega_p + 2\omega_{SRS2}) - (\omega_p + \omega_{SRS2})] = 2\omega_{AST2-2} - \omega_{AST1-2} = \omega_{AST3-2}$		≈ 375
0.5106	ASt ₁₋₁	$^*\omega_p + \omega_{SRS1} = [\omega_p + \omega_p - (\omega_p - \omega_{SRS1})] = \omega_{AST1-1}$	≈ 792	
0.5117	ASt ₂₋₂	$^*\omega_p + 2\omega_{SRS2} = [(\omega_p + \omega_{SRS2}) + (\omega_p + \omega_{SRS2}) - \omega_p] = 2\omega_{AST1-2} - \omega_p = \omega_{AST2-2}$		≈ 375
0.5217	ASt ₁₋₂	$^*\omega_p + \omega_{SRS2} = [\omega_p + \omega_p - (\omega_p - \omega_{SRS2})] = \omega_{AST1-2}$		≈ 375
0.53207	λ_p	ω_p	–	–
0.5429	St ₁₋₂	$\omega_p - \omega_{SRS2} = \omega_{St1-2}$		≈ 375
0.5542	St ₂₋₂	$^*\omega_p - 2\omega_{SRS2} = \omega_{St2-1} - \omega_{SRS2} = \omega_{St2-2}$		≈ 375
0.5555	St ₁₋₁	$\omega_p - \omega_{SRS1} = \omega_{St1-1}$	≈ 792	
0.5660	St ₃₋₂	$^*\omega_p - 3\omega_{SRS2} = \omega_{St2-2} - \omega_{SRS2} = \omega_{St3-2}$		≈ 375
0.5810	St ₂₋₁	$^*\omega_p - 2\omega_{SRS1} = \omega_{St1-1} - \omega_{SRS1} = \omega_{St2-1}$	≈ 792	
0.6091	St ₃₋₁	$^*\omega_p - 3\omega_{SRS1} = \omega_{St2-1} - \omega_{SRS1} = \omega_{St3-1}$	≈ 792	

^{a)} Measurement accuracy for separate lines is $\pm 0.0003 \mu\text{m}$ and for the overlapping lines is $\pm 0.0005 \mu\text{m}$.

^{b)} Possible cascade SRS and RFWM processes are asterisked.

Table 2 Spectral composition of Raman-induced Stokes and anti-Stokes generation of “garnet” Lu₃Al₅O₁₂ fine-grained ceramics recorded at room temperature with the second harmonic radiation pumping at $\lambda_p = 0.53207 \mu\text{m}$

small energy difference between absorption pump photons and lasing photons) that causes low thermal load under $0.97 \mu\text{m}$ continuous wave (CW) laser-diode (LD) pumping. Since the Lu₃Al₅O₁₂:Yb³⁺ garnet have excellent thermo-mechanical characteristics it is preferred for high power lasers, especially with a cryogenic cooling when well realized quasi-4-level operating regime. Of course, the above-mentioned, to large extent also applies to Y₃Al₅O₁₂:Yb³⁺ garnet laser materials (for crystals and ceramics) that are virtually identical with both the spectroscopic [10] and thermal properties (see, e.g. [11]). But, here we should also note an important feature, ion properties (e.g., ionic radius and mass, the configuration of the electron shell), Yb³⁺ and Lu³⁺ are very close (they

are immediate neighbors on the Periodic Table) compared with Yb³⁺ and Y³⁺, which are far from each other in this table. Among experts there is a perception that it has a positive effect in growing high quality lutetium garnets with necessary concentration of the Ln³⁺ lasant ions. It can be confidently asserted, that in the coming years the Lu₃Al₅O₁₂:Ln³⁺ laser ceramics begin their rapid expansion both in research laboratories and in industry, that is, to repeat the situation with the Y₃Al₅O₁₂:Ln³⁺ laser ceramics at the beginning of this century, We hope that, their creation and development will help accumulated over the years of considerable knowledge about Lu₃Al₅O₁₂:Ln³⁺ laser crystals. Therefore, we deemed it appropriate to present in Table 1 historical stages of the first lasing ex-

Crystalline “garnet” materials		Lasant ions	ω_{SRS} , cm ⁻¹	g_{srs}^{St1} , cm/GW
Composition ^{a)}	Type			
{Ca ₃ }[Ga ₂](Ge ₃)O ₁₂	crystal	Nd ³⁺ , Er ³⁺ , d-c ^{b)}	≈ 807 [48]	≈ 0.5 ^{c)}
{Ca ₃ }[Nb,Ga] ₂ (Ga ₃)O ₁₂	crystal	Nd ³⁺ , Er ³⁺ , Yb ³⁺	≈ 500 [49]	–
Y ₃ Al ₅ O ₁₂	crystal	Pr ³⁺ , Nd ³⁺ , Ho ³⁺ , Er ³⁺ , Tm ³⁺ , Yb ³⁺ , Cr ³⁺	≈ 370 [50]	≈ 0.1 ^{d)}
	ceramics	Nd ³⁺ , Yb ³⁺	≈ 370 [51]	≈ 0.1 ^{d)}
{YGd ₂ }[Sc ₂](Al ₂ Ga)O ₁₂	ceramics	Yb ³⁺	≈ 358 [52]	≈ 0.15 ^{d)}
{Gd ₃ }[Sc ₂](Ga ₃)O ₁₂	crystal	Nd ³⁺ , Ho ³⁺ , Er ³⁺ , Tm ³⁺ , Yb ³⁺ , Cr ³⁺	≈ 351 [49]	–
Gd ₃ Ga ₅ O ₁₂	crystal	Pr ³⁺ , Nd ³⁺ , Ho ³⁺ , Er ³⁺ , Yb ³⁺ , Cr ³⁺ , Ni ²⁺	≈ 354 [49]	–
Tb ₃ Ga ₅ O ₁₂	crystal	–	≈ 360 [53]	≥ 0.3 ^{c)}
Lu ₃ Al ₅ O ₁₂	ceramics	e)	≈ 792, ≈ 375	≥ 0.2 ^{f)}

- a) The formula for the two-cation compounds presented in the usual form, while for more complex garnets it indicated the distribution of cations over the crystallographic positions of garnet structure: where the braces, square brackets, and parentheses mark the cations occupying the c, a, and d positions, respectively.
- b) d-c means a defect-center.
- c) Evaluated with the picosecond pumping at 0.53207 μm wavelength.
- d) Evaluated with the picosecond pumping at 1.06415 μm wavelength.
- e) Potential crystalline host-material for Ln and TM lasant ions.
- f) Evaluated with the picosecond pumping at 0.53207 μm wavelength for the first Stokes lasing related to $\omega_{SRS1} \approx 792 \text{ cm}^{-1}$.

Table 3 SRS-active garnet crystalline materials (single crystals and ceramics)

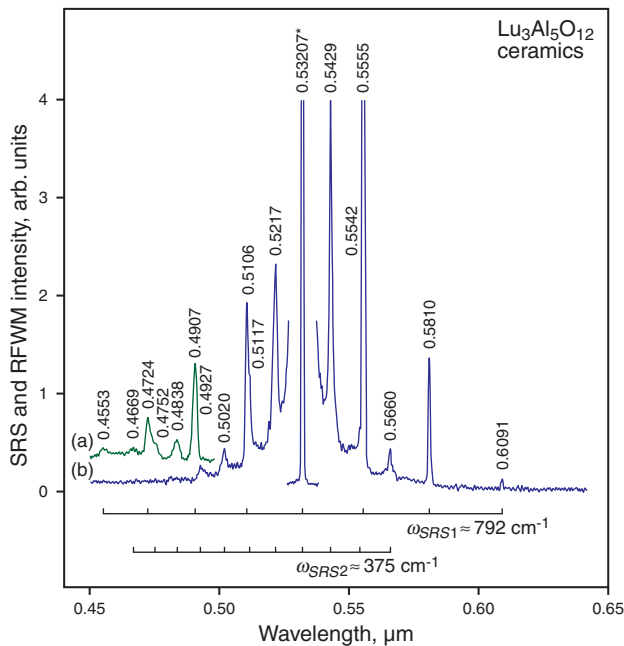


Figure 1 (online color at www.lphys.org) Fragments of SRS and RFWM spectrum of crystalline “garnet” ceramics Lu₃Al₅O₁₂ recorded at room temperature with picosecond pumping at the wavelength $\lambda_p = 0.53207 \mu\text{m}$. The wavelength of all lines (pump line asterisk) are given in μm, their intensities are shown without correction for the spectral sensitivity of the used spectroscopic system with a Si-CCD sensor. The spacing of the Stokes and anti-Stokes lasing lines related to SRS-promoting vibration modes $\omega_{SRS1} \approx 792 \text{ cm}^{-1}$ and $\omega_{SRS2} \approx 375 \text{ cm}^{-1}$ of ceramic studied are indicated by the horizontal scale brackets. The fragment (a) recorded under higher pump power using a BG3 glass Schott filter

periments with Lu₃Al₅O₁₂:Ln³⁺ crystals. In this table, which has a review character also included several references of recent publications on Lu₃Al₅O₁₂:Ln³⁺ crystalline lasers with LD pumping. Our confidence in the expected publications of Lu₃Al₅O₁₂:Ln³⁺ ceramic lasers reinforces the series of recent articles on scintillator-oriented (e.g. medical imaging) applications of transparent Pr³⁺-ion doped Lu₃Al₅O₁₂ ceramics (see, e.g. [38, 39]). This work was promoted by the above-mentioned vision and is dedicated to the results of the study of nonlinear-optical $\chi^{(3)}$ -properties of “garnet” Lu₃Al₅O₁₂ fine-grained ceramics as a perspective host-matrix for Ln- and TM-lasant ions. Samples for our study were made by VSN method, which was used for the fabrication of well-known Y₃Al₅O₁₂:Ln³⁺ laser ceramics (see, e.g. [40]).

2. Stimulated and spontaneous Raman scattering

The investigation of the different manifestations of Raman-induced $\chi^{(3)}$ -nonlinear optical interactions in isotropic “garnet” Lu₃Al₅O₁₂ ceramics was performed in single-pass excitation scheme applying the experimental setup, which we repeatedly used in our studies on the SRS-spectroscopy of inorganic and organic crystalline materials (see, e.g. [41–44]). In this work, as in our previous similar study it was used as the pump source a Xe-flashlamp-pumped Nd³⁺:Y₃Al₅O₁₂ mode-locked picosecond laser with a double-pass Nd³⁺:Y₃Al₅O₁₂ amplifier. Its ≈ 110-ps radiation at 1.06415 μm wavelength (main inter-Stark SE transition of the lasing $^4F_{3/2} \rightarrow ^4I_{11/2}$ channel of Nd³⁺ ions) and up to ≈ 5 mJ energy was transformed by an internal KTiOPO₄-doubler (with an efficiency of ≈ 25%)

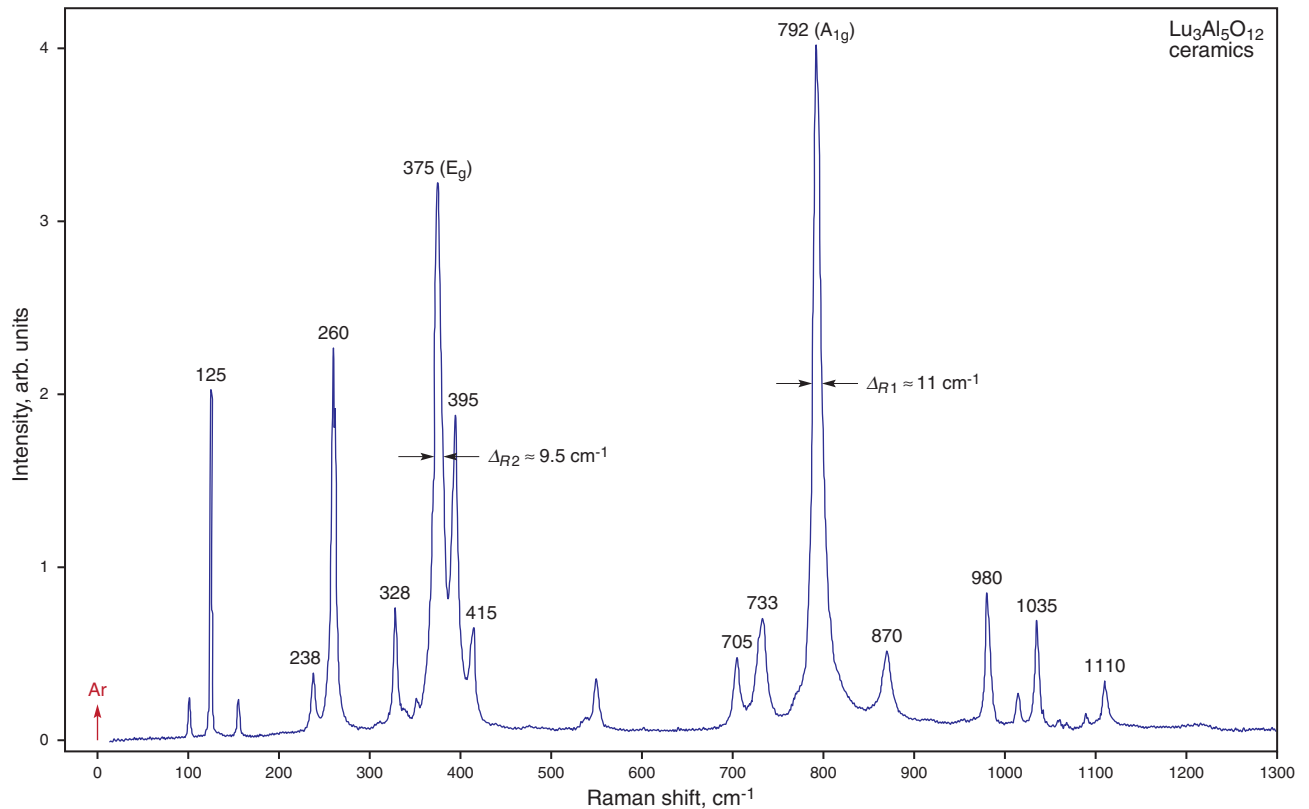


Figure 2 (online color at www.lphys.org) The first order spontaneous Raman scattering (A_{1g} , E_g)-spectrum of fine-grained $\text{Lu}_3\text{Al}_5\text{O}_{12}$ ceramics recorded at room temperature under Ar-ion laser excitation at $0.488 \mu\text{m}$ (indicated by a vertical arrow). The frequency of some Raman shifted lines are given in cm^{-1}

into the second harmonic generation with pulse duration of about 80 ps. The nearly Gaussian pump beam of this pump radiation $\lambda_p = 0.53207 \mu\text{m}$ wavelength is focused into the $\text{Lu}_3\text{Al}_5\text{O}_{12}$ ceramic 20-mm long sample with a spherical plane-convex lens, resulting in a beam waist diameter of about $160 \mu\text{m}$. The spectral composition of the multi-wavelength $\chi^{(3)}$ -lasing of the title ceramics by its SRS and Raman-induced four-wave mixing (RFWM) parametric cascade processes is analyzed with a spectrometric system based on a scanning grating monochromator (McPherson Model 270) equipped with a Hamamatsu linear image Si-CCD sensor (S3923-1024Q), which provide sufficient spectral sensitivity in the visible range. One of the recorded SRS-RFWM spectra is shown in Fig. 1. The results of the identification of its Stokes and anti-Stokes components are listed in Table 2. As can be seen from these experimental data the $\text{Lu}_3\text{Al}_5\text{O}_{12}$ ceramic “garnet” under picosecond excitation at $\lambda_p = 0.53207 \mu\text{m}$ wavelength is manifested two SRS-promoting vibration modes $\omega_{SRS1} \approx 792 \text{ cm}^{-1}$ and $\omega_{SRS2} \approx 375 \text{ cm}^{-1}$. In our experiments, we can also roughly estimate the steady-state Raman gain coefficient g_{ssR}^{St1-1} of the title ceramics for its first-Stokes lasing component at $\lambda_{St1-1} = 0.5555 \mu\text{m}$ wavelength related to stronger vibronic transition with

$\omega_{SRS1} \approx 792 \text{ cm}^{-1}$ (see Fig. 2). We are able to do this because the excitation regime of SRS generation in our experiment is well satisfy the necessary condition $\tau_p \gg T_2 = (\pi \Delta_{R1})^{-1} \approx 0.96 \text{ ps}$ (here T_2 and Δ_R are the phonon relaxation (dephasing) time and the linewidth of the corresponding Raman shifted line in the spontaneous Raman scattering spectrum, see Fig. 2). As in our previous works (see, e.g. [41, 42]), for this aim we used the sufficiently tested method based on the well known ratio $g_{ssR}^{St1-1} I_p^{thr} l_{SRS} \approx 30$ (see, e.g. [45]) together with comparative measurements of the “threshold” pump intensity I_p^{thr} (accordance to the conventional definition adopted in [46, 47]) of the confidently detectable first Stokes lasing signal for our $\text{Lu}_3\text{Al}_5\text{O}_{12}$ ceramics and for a reference isotropic crystalline material with approximately equal SRS-active length ($l_{SRS} \approx 20 \text{ mm}$). Based on the data of this rough comparative evaluations, we concluded that the value of the steady-state Raman gain coefficient g_{ssR}^{St1-1} for $\text{Lu}_3\text{Al}_5\text{O}_{12}$ ceramics not less than 0.2 cm/GW . In recent years SRS-effect was observed also in several “garnet” crystals and ceramics with ordered and partly disordered structure, most of them are also host-materials for Ln and TM lasant ions. To provide a place of ceramic stud-

ied among those “garnet” crystalline materials the second small overview composed, which is presented as Table 3.

As can be seen, the listed here $\chi^{(3)}$ -active crystalline materials, like ceramics $\text{Lu}_3\text{Al}_5\text{O}_{12}$, despite the fact that they appear different SRS-promoting vibration modes, their steady-state Raman gain coefficients are almost equal and low. In the case of large-scale SE-active elements (in high-power generators or amplifiers) on the base of these garnet materials, the risk of parasitic SRS at high densities of laser radiation in them will extremely low. It certainly is a positive factor.

Now, briefly about the nature of the two SRS-promoting vibration modes of crystalline ceramics $\text{Lu}_3\text{Al}_5\text{O}_{12}$. Eighty atoms in the primitive cell of the O_h^{10} -cubic garnet structure give $3N_{Br} = 240$ degrees of freedom, which (in accordance with the factor-group analysis and symmetry degeneracy [54]) give rise to 97 lattice optical vibration modes. They could be characterized (at $\mathbf{k} = 0$, center of Brillouin zone) by the following set of irreducible representations:

$$\Gamma_N = 3A_{1g} + 8E_g + 14F_{2g} + 5A_{1u} + 5A_{2u} + 5A_{2g} + 10E_u + 14F_{1g} + 16F_{2u} + 17F_{1u},$$

among which 25 modes ($3A_{1g} + 8E_g + 14F_{2g}$) are Raman active and should be appear in the spontaneous Raman scattering spectra. Because our ceramics is a dense conglomeration of randomly oriented microcrystalline grains with the size of most grains of about one micron, we have been sufficient to measure only one spectrum, which is shown in Fig. 2. Comparative analysis of our spectrum with the data the study of Raman-active phonons of $\text{Lu}_3\text{Al}_5\text{O}_{12}$ single crystals (see, e.g. [55–57]) allowed us to conclude that both the SRS-promoting vibrations of the $\text{Lu}_3\text{Al}_5\text{O}_{12}$ ceramics related to A_{1g} and E_g modes of its AlO_4 molecular tetrahedral units. With high probability can be argued that the highest frequency vibration A_{1g} ($\approx 792 \text{ cm}^{-1}$) corresponds to its breathing mode and the lower frequency vibration E_g ($\approx 375 \text{ cm}^{-1}$) connects to the rotation of the molecular unit. Here it is pertinent to note that the observed width of the lines $\Delta\nu_{R1} \approx 11 \text{ cm}^{-1}$ and $\Delta\nu_{R2} \approx 9.5 \text{ cm}^{-1}$ can be associated with the imposition of neighboring lines with close vibronic frequencies related to the optical modes (A_{g1} , E_g , F_{2g}) [55–57]. It is perfectly acceptable, because in our measurements, a relatively wide Ar-laser beam is excited in $\text{Lu}_3\text{Al}_5\text{O}_{12}$ ceramics of several differently oriented its one-micron-sized crystalline grains.

3. Conclusion

We have discovered stimulated Raman scattering of the $\text{Lu}_3\text{Al}_5\text{O}_{12}$ -based crystalline ceramics. The spectral components of its multipole Stokes and anti-Stokes picosecond $\chi^{(3)}$ -nonlinear lasing were identified and attributed to two SRS-promoting vibration modes $\omega_{SRS1} \approx 792 \text{ cm}^{-1}$

and $\omega_{SRS2} \approx 375 \text{ cm}^{-1}$, which directly originate from internal atom motions of the AlO_4 molecular tetrahedral units. There is no doubt that the title ceramics is a promising crystalline host-material for lasing ions. Therefore, we hope that our work in a modest degree of push search investigation to create real $\text{Lu}_3\text{Al}_5\text{O}_{12}$ laser ceramics.

Knowing the experience in the development of lasers on the base of single crystals $\text{Lu}_3\text{Al}_5\text{O}_{12}$ doped with Ln^{3+} ions, we can expect the rapid development of ceramics $\text{Lu}_3\text{Al}_5\text{O}_{12}:\text{Ln}^{3+}$, capable to generate at inter-Stark transitions of $J' \rightarrow J'$ SE channels of lasing ions that projected in Table 1.

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References

- [1] A.A. Kaminskii, Laser Photon. Rev. **1**, 93 (2007).
- [2] A.A. Kaminskii, Crystalline Lasers: Physical Processes and Operating Schemes (CRC Press, Boca Raton, FL, 1996).
- [3] W. Koechner, Solid-State-Laser Engineering (Springer, Berlin, 1976, 1980, 1988, 1991, 1996, 2000, 2005).
- [4] M.J. Weber (ed.), Handbook of Laser Science and Technology (CRC Press, Boca Raton, FL, 2000).
- [5] A.A. Kaminskii, P.V. Klevtsov, Kh.S. Bagdasarov, A.A. Maier, A.A. Pavlyuk, A.G. Petrosyan, and M.V. Provotorov, JETP Lett. **16**, 387 (1972).
- [6] A.A. Kaminskii and S.E. Sarkisov, Inorg. Mater. (USSR) **9**, 457 (1973).
- [7] A.A. Kaminskii, S.E. Sarkisov, P.V. Klevtsov, Kh.S. Bagdasarov, A.A. Pavlyuk, and A.G. Petrosyan, Phys. Status Solidi (a) **17**, K75 (1973).
- [8] A.A. Kaminskii, Kh.S. Bagdasarov, A.G. Petrosyan, and S.E. Sarkisov, Phys. Status Solidi (a) **18**, K31 (1973).
- [9] Kh.S. Bagdasarov, G.A. Bogomolova, D.N. Vylegzhanin, A.A. Kaminskii, A.M. Kevorkov, A.G. Petrosyan, and A.M. Prokhorov, Sov. Phys. Dokl. **19**, 358 (1974).
- [10] G.A. Bogomolova, D.N. Vylegzhanin, and A.A. Kaminskii, Sov. Phys. JETP **42**, 440 (1976).
- [11] A.A. Kaminskii, G.A. Bogomolova, Kh.S. Bagdasarov, and A.G. Petrosyan, Opt. Spectrosc. (USSR) **39**, 643 (1975).
- [12] A.A. Kaminskii, in: Spektroskopiya Kristallov (Nauka, Moscow, 1975), p. 92, in Russian.

- [13] X.D. Xu, X.D. Wang, J.Q. Meng, Y. Cheng, D.Z. Li, S.S. Cheng, F. Wu, Z.W. Zhao, and J. Xu, *Laser Phys. Lett.* **6**, 678 (2009).
- [14] X.D. Xu, S.S. Cheng, J.Q. Meng, D.Z. Li, D.H. Zhou, L.H. Zheng, J. Xu, W. Ryba-Romanowski, and R. Lisiecki, *Opt. Express* **18**, 21370 (2010).
- [15] A.A. Kaminskii, N.B. Karlov, S.É. Sarkisov, O.M. Stelmakh, and V.E. Tushk, *Quantum Electron.* **6**, 1371 (1976).
- [16] A.A. Kaminskii, V.A. Fedorov, A.G. Petrosyan, A.A. Pavlyuk, I. Boom, P. Reiche, and D. Schulz, *Inorg. Mater.* **15**, 1180 (1979).
- [17] V. Kushawaha, Y. Chen, Y. Yan, and L. Major, *Appl. Phys. B* **62**, 109 (1996).
- [18] N.P. Barnes, E.D. Filer, F.L. Naranjo, W.J. Rodriguez, and M.R. Kokta, *Opt. Lett.* **18**, 708 (1993).
- [19] A.A. Kaminskii, K. Kurbanov, and A.G. Petrosyan, *Phys. Status Solidi (a)* **98**, K57 (1987).
- [20] A.A. Kaminskii, A.G. Petrosyan, V.A. Fedorov, S.E. Sarkisov, V.V. Ryabchenkov, A.A. Pavlyuk, A.A. Lyubchenko, and I.V. Mochalov, *Sov. Phys. Dokl.* **26**, 846 (1981).
- [21] A.A. Kaminskii, T.I. Butaeva, A.M. Kevorkov, V.A. Fedorov, A.G. Petrosyan, and M.M. Gritsenko, *Inorg. Mater. (USSR)* **12**, 1238 (1976).
- [22] A.A. Kaminskii, A.O. Ivanov, S.E. Sarkisov, I.V. Mochalov, V.A. Fedorov, and L. Li, *Sov. Phys. JETP* **44**, 516 (1976).
- [23] A.A. Kaminskii, T.I. Butaeva, A.O. Ivanov, I.V. Mochalov, A.G. Petrosyan, G.I. Rogov, and V.A. Fedorov, *Sov. Tech. Phys.* **2**, 308 (1976).
- [24] A.A. Kaminskii and A.G. Petrosyan, *Inorg. Mater. (USSR)* **15**, 543 (1979).
- [25] A.M. Prokhorov, A.A. Kaminskii, V.V. Osiko, M.I. Timoshchekin, E.V. Zharikov, T.I. Butaeva, S.E. Sarkisov, A.G. Petrosyan, and V.A. Fedorov, *Phys. Status Solidi (a)* **40**, K69 (1977).
- [26] A.A. Kaminskii, A.G. Petrosyan, G.A. Denisenko, T.I. Butaeva, V.A. Fedorov, and S.E. Sarkisov, *Phys. Status Solidi (a)* **71**, 291 (1982).
- [27] M.A. Andriasyan, N.V. Vardanyan, and R.B. Kostanyan, *Quantum Electron.* **12**, 804 (1982).
- [28] A.A. Kaminskii and A.G. Petrosyan, *Sov. Phys. Dokl.* **24**, 363 (1979).
- [29] J.D. Kmetec, T.S. Kubo, T.J. Kane, and C.J. Grund, *Opt. Lett.* **19**, 186 (1994).
- [30] Kh.S. Bagdasarov, G.A. Bogomolova, D.N. Vylegzhanin, A.A. Kaminskii, A.M. Kevorkov, A.G. Petrosyan, and A.M. Prokhorov, *Sov. Phys. Dokl.* **19**, 358 (1974).
- [31] G.A. Bogomolova, D.N. Vylegzhanin, and A.A. Kaminskii, *Sov. Phys. JETP* **42**, 440 (1976).
- [32] T. Kasamatsu, H. Sekita, and Y. Kuwano, *Appl. Opt.* **38**, 5149 (1999).
- [33] A. Brenier, Y. Guyot, H. Canibano, G. Boulon, A. Ródenas, D. Jaque, A. Eganyan, and A.G. Petrosyan, *J. Opt. Soc. Am. B* **23**, 676 (2006).
- [34] J. Dong, K. Ueda, and A.A. Kaminskii, *Opt. Lett.* **32**, 3266 (2007).
- [35] A.A. Kaminskii, *Bull. Acad. Sci. USSR. Fiz. Ser.* **45**, 106 (1981).
- [36] A.A. Kaminskii, A.G. Petrosyan, and V.A. Fedorov, *Sov. Phys. Dokl.* **26**, 309 (1981).
- [37] A.A. Kaminskii, A.G. Petrosyan, and K.L. Ovanesyan, *Inorg. Mater.* **19**, 1098 (1984).
- [38] Y. Shi, M. Nikl, X.Q. Feng, J.A. Mares, Y.Q. Shen, A. Beitelrova, R. Kucerkova, Y.B. Pan, and Q. Liu, *J. Appl. Phys.* **109**, 013522 (2011).
- [39] T. Yanagida, A. Fukabori, Y. Fujimoto, A. Ikesue, K. Kamada, J. Kataoka, Y. Yokota, A. Yoshikawa, and V. Chani, *Phys. Status Solidi (c)* **8**, 140 (2011).
- [40] Japanese patents 10-101333 and 10-101411 (1998).
- [41] A.A. Kaminskii, L. Bohatý, P. Becker, P. Held, H. Rhee, H.J. Eichler, and J. Hanuza, *Laser Phys. Lett.* **6**, 335 (2009).
- [42] A.A. Kaminskii, L. Bohatý, P. Becker, H.J. Eichler, H. Rhee, and J. Hanuza, *Laser Phys. Lett.* **6**, 872 (2009).
- [43] A.A. Kaminskii, L. Bohatý, P. Becker, H.J. Eichler, and H. Rhee, *Laser Phys. Lett.* **7**, 142 (2010).
- [44] P. Becker, L. Bohatý, J. Liebertz, H.-J. Kleebe, M. Müller, H.J. Eichler, H. Rhee, J. Hanuza, and A.A. Kaminskii, *Laser Phys. Lett.* **7**, 367 (2010).
- [45] W. Kaiser and M. Maier, in: F.T. Arecchi and E.O. Schultz-Dubois (eds.), *Laser Handbook*, Vol. 2 (North-Holland, Amsterdam, 1972), p. 1077.
- [46] K.K. Lai, W. Schüsslbauer, H. Silberbauer, H. Amler, U. Bogner, M. Maier, M. Jordan, and H.-J. Jodl, *Phys. Rev. B* **42**, 5834 (1990).
- [47] A.A. Kaminskii, P. Becker, L. Bohatý, K. Ueda, K. Takaichi, J. Hanuza, M. Maczka, H.J. Eichler, and G.M.A. Gad, *Opt. Commun.* **206**, 179 (2002).
- [48] A.A. Kaminskii, H.J. Eichler, J. Fernandez, J. Findeisen, R. Balda, and A.V. Butashin, *Phys. Status Solidi (b)* **207**, R3 (1998).
- [49] A.A. Kaminskii, H.J. Eichler, K.-I. Ueda, J. Fernandez, J. Findeisen, and R. Balda, *Quantum Electron.* **28**, 939 (1998).
- [50] A.A. Kaminskii, H.J. Eichler, K. Ueda, S.N. Bagaev, G.M.A. Gad, J. Lu, T. Murai, H. Yagi, and T. Yanagitani, *Phys. Status Solidi (a)* **181**, R19 (2000).
- [51] A.A. Kaminskii, H.J. Eichler, K. Ueda, S.N. Bagaev, G.M.A. Gad, J. Lu, T. Murai, H. Yagi, and T. Yanagitani, *JETP Lett.* **72**, 499 (2000).
- [52] A.A. Kaminskii, S.N. Bagaev, K. Ueda, H. Yagi, H.J. Eichler, A. Shirakawa, M. Tokurakawa, H. Rhee, K. Takaichi, and T. Yanagitani, *Laser Phys. Lett.* **6**, 671 (2009).
- [53] A.A. Kaminskii, H.J. Eichler, P. Reiche, and R. Uecker, *Laser Phys. Lett.* **2**, 489 (2005).
- [54] D.L. Rousseau, R.P. Bauman, and S.P.S. Porto, *J. Raman Spectrosc.* **10**, 253 (1981).
- [55] J.-J. Song, P.B. Klein, R.L. Wadsack, M. Selders, S. Mroczkowski, and R.K. Chang, *J. Opt. Soc. Am.* **63**, 1135 (1973).
- [56] K. Papagelis, J. Arvanitidis, G. Kanellis, G.A. Kourouklis, and S. Ves, *Phys. Status Solidi (b)* **211**, 301 (1999).
- [57] K. Papagelis, G. Kanellis, S. Ves, and G.A. Kourouklis, *Phys. Status Solidi (b)* **233**, 134 (2002).