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NEW MANIFESTATION OF NONLINEAR PROPERTIES OF ACENTRIC TRIGONAL $LaBGeO_5$ AND ORTHORHOMBIC $Gd_2(MoO_4)_3$ LASER CRYSTAL-HOSTS: MULTIPLE STOKES AND ANTI-STOKES SRS AND SHG DUE TO CERENKOV-TYPE PHASE MATCHING

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We have discussed new optical potentialities in acentric $Gd_2(MoO_4)_3$ and $LaBGeO_5$ laser crystal-hosts having simultaneously relatively high $\chi^{(2)}$ and $\chi^{(3)}$ nonlinear susceptibilities. In particular, we show experimental results on efficient multiple Stokes and anti-Stokes stimulated Raman scattering and Raman induced four-wave mixing, as well as second harmonic generation with Cerenkov-type phase matching.

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1. Introduction

The structural diversity provides a wide variety of physical properties of laser crystals. The resultant inexhaustible spectroscopic potential constitutes a gold mine of opportunities to develop and create new operating schemes and principles for excitation of their stimulated emission in different experimental conditions [1]. Among about three hundred of insulating laser crystal-hosts ever known, acentric compounds with high quadratic nonlinear susceptibility ($\chi^{(2)}$) are distinguished by a unique set of properties. Thus, they have already provided one of the most elegant optical advances in laser physics, i.e. the self-frequency doubling phenomenon. A not less interesting nonlinear interaction, the self-stimulated Raman scattering, is also exhibited by laser crystals having considerable cubic nonlinear susceptibility ($\chi^{(3)}$). Recently we showed that two acentric $LaBGeO_5$ and $Gd_2(MoO_4)_3$ laser-crystal hosts offer simultaneously relatively high ($\chi^{(2)}$) and ($\chi^{(3)}$) nonlinearities, which allow one to apply them both in second harmonic generators of one-micron emission of neodymium lasers [2,3] and in crystalline laser Raman shifters [4].

In this lecture I would like to summarize data on new laser potentialities, which were discovered recently [5–7]* in the $LaBGeO_5$ and $Gd_2(MoO_4)_3$ ferroelectrics, efficient multiple Stokes and anti-Stokes stimulated Raman scattering (SRS) and second harmonic generation (SHG) due to Cerenkov-type matching.

2. Efficient multiple Stokes and anti-Stokes SRS (in trigonal $LaBGeO_5$ and orthorhombic $Gd_2(MoO_4)_3$ ferroelectrics

In our SRS experiments we used a high-power picosecond $Y_3Al_5O_{12}:Nd^{3+}$ laser system (with two-stage amplifier) designed in Optical Institute of the Berlin Technical University (Fig. 1) [8]. It can generate *ca.* 120 ps single pulses at 1.06415 μm wavelength up to 10 mJ at 0.53027 μm wavelength of SHG (with KD*P frequency doubler) with a pulse duration of about 80 ps up to 3 mJ. The crystalline elements as a rectangular oriented parallelepipeds are placed in the focus of a lens with $F = 500$ mm with waist diameter of about 75 μm. Spectral composition of Stokes and anti-Stokes generation is measured with a Si-CCD Spectrometric Multichannel Analyzer (SCMA) consisting of a modified McPherson Model 218 Scanning grating monochromator. The main results are listed in Table (for $LaBGeO_5$ crystals) and illustrated in Figs. 1–5.

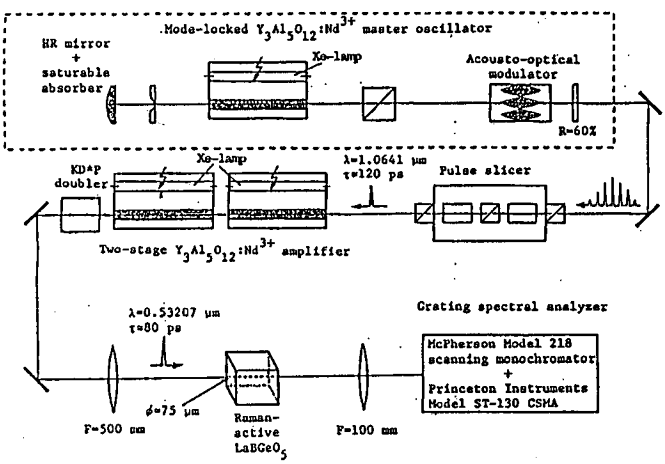

Fig. 1. Schematic experimental set-up for SRS excitation in acentric $LaBGeO_5$ and $Gd_2(MoO_4)_3$ single crystals. For explanations see the text.

One can see that spectral composition of Stokes and anti-Stokes emission of the $LaBGeO_5$ and $Gd_2(MoO_4)_3$ is related to their several SRS-active vibration modes — 385 and 803 cm^{-1} for $LaBGeO_5$ and about 100, 857, 943, and 960 cm^{-1} for the $Gd_2(MoO_4)_3$ crystals, respectively. High efficiency (more than 60%) of the SRS-processes in these two acentric compounds is confirmed by the results of the

*Within a cooperation of the Joint Open Laboratory for Laser Crystals and Precise Laser Systems with Professors S.N. Bagaev, H.-J. Eichler, and K. Ueda, as well as with Doctors A.V. Butashin, D. Grebe, R. Macdonald, H. Nishioka, A.A. Pavlyuk et al.

TABLE

Spectral composition of the Stokes and anti-Stokes generation of Raman lasers based on acentric $LaBGeO_5$ crystals at room temperature under pumping with picosecond $Y_3Al_5O_{12}:Nd^{3+}$ laser at 1.06415 and 0.53207 μm (SHG) wavelengths.

Pumping conditions		Anti-Stokes and Stokes emission		SRS- and RFWM-line attribution	SRS-active vibration mode [cm^{-1}]	
λ_p [μm]	Geometry of excitation[a]	Component	Wavelength[b] [μm]		ω_{R1}	ω_{R2}
1.06415	$\parallel a$, $\parallel b$	ASt_{6-1}	0.7035*	$\omega_p + 6\omega_{R1}$	803	
		ASt_{5-1}	0.7456*	$\omega_p + 5\omega_{R1}$	803	
		ASt_{4-1}	0.7931*	$\omega_p + 4\omega_{R1}$	803	
		ASt_{3-1}	0.8470*	$\omega_p + 3\omega_{R1}$	803	
		ASt_{2-1}	0.9088	$\omega_p + 2\omega_{R1}$	803	
		ASt_{1-1}	0.9804	$\omega_p + \omega_{R1}$	803	
		St_{1-1}	1.1636	$\omega_p - \omega_{R1}$	803	
1.06415	$\parallel b$, $\parallel a$	ASt_{3-2}	0.9477	$\omega_p + 3\omega_{R2}$		385
		ASt_{2-2}	0.9834	$\omega_p + 2\omega_{R2}$		385
		ASt_{1-2}	1.0223	$\omega_p + \omega_{R2}$		385
		St_{1-2}	1.1096*	$\omega_p - \omega_{R2}$		385
0.53207	$\parallel a$, $\approx\parallel b$	ASt_{2-1}	0.4902	$\omega_p + 2\omega_{R1}$	803	
		ASt_{1-1}	0.5103	$\omega_p + \omega_{R1}$	803	
		St_{1-1}	0.5558	$\omega_p - \omega_{R1}$	803	
		St_{2-1}	0.5818*	$\omega_p - 2\omega_{R1}$	803	
		St_{3-1}	0.6103*	$\omega_p - 3\omega_{R1}$	803	
		St_{4-1}	0.6418*	$\omega_p - 4\omega_{R1}$	803	
		St_{5-1}	0.6766*	$\omega_p - 5\omega_{R1}$	803	
0.53207	$\parallel b$, $\parallel a$	ASt_{1-2}	0.5214	$\omega_p + \omega_{R2}$		385
		St_{2-1}	0.5432	$\omega_p - \omega_{R2}$		385
		St_{2-2}	0.5548*	$\omega_p - 2\omega_{R2}$		385
		St_{1-1}	0.5558	$\omega_p - \omega_{R1}$	803	
		St_{3-2}	0.5669*	$\omega_p - 3\omega_{R2}$		385
0.53207	$\parallel c$, $\parallel a$	ASt_{1-2}	0.5214	$\omega_p + \omega_{R2}$		385
		St_{2-1}	0.5432	$\omega_p - \omega_{R2}$		385
		St_{2-2}	0.5548*	$\omega_p - 2\omega_{R2}$		385
		St_{3-2}	0.5669*	$\omega_p - 3\omega_{R2}$		385
		St_{4-2}	0.5796*	$\omega_p - 4\omega_{R2}$		385

[a]Direction and polarization of pumping laser emission.
[b] Measurement accuracy was ± 0.0005 μm. We have also observed the co-axial ring anti-Stokes emission by Raman induced four-wave mixing (RFWM, indicated by asterisk).

excitation of their multiple anti-Stokes lines which transform IR-pump emission to a generation in the visible (6-th anti-Stokes component).

Except the above measurements, the special experiment was performed in which one oriented $Gd_2(MoO_4)_3$ crystalline element carried out two functional roles of nonlinear laser optics, as a frequency doubler of the picosecond pump emission at 1.06415 μm wavelength and as Raman shifter. In the latter one anti-Stokes and three Stokes components were excited in the visible by the high intensity

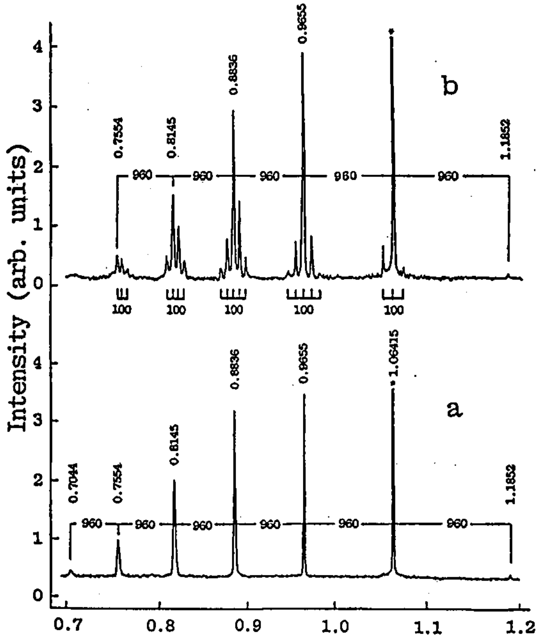

Fig.2

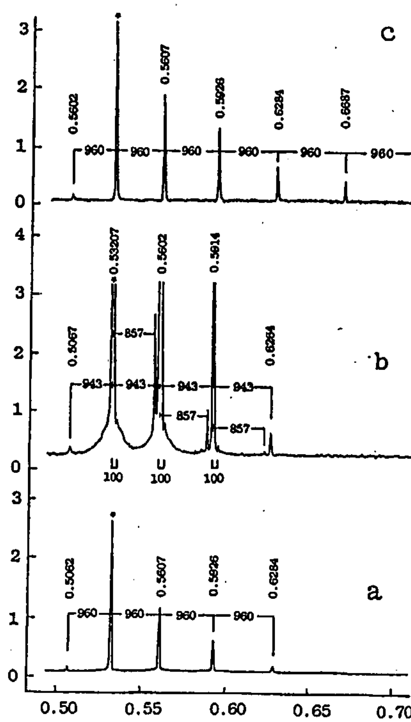

Fig.3

Fig. 2. Orientational SRS spectra of orthorhombic $Gd_2(MoO_4)_3$ crystal at 300 K and under IR pumping ($\lambda_p = 1.06415$ μm): (a) — excitation along the c-axis with polarization vector along the a-axis; (b) — excitation along the b-axis with the polarization vector along the c-axis. The pumped line is marked with asterisk. Intensities of SRS components and pumping lines are shown without a correction on spectral sensitivity of Si-CCD photodiode array. Wavelengths of all lines are given in μm. The connection of Stokes and anti-Stokes generation lines with SRS-active vibration modes of a crystal with frequencies of about 100 and 960 cm^{-1} are indicated by lines and brackets.

Fig. 3. Orientational SRS spectra of orthorhombic $Gd_2(MoO_4)_3$ crystal at 300 K and under "green" pumping ($\lambda_p = 0.53207$ μm): (a) — fundamental IR laser emission and its SHG (pumping) under of the angle $\angle bc \approx 70°$ with polarization along the a-axis; (b) — excitation along the a-axis with polarization vector along the b-axis; and (c) — excitation along c-axis with polarization vector along the a-axis. The connection of Stokes and anti-Stokes generation lines with SRS-active vibration modes of a crystal with frequencies about 100, 857, 943 and 960 cm^{-1} are indicated by lines and brackets. Other notations as in Fig. 2.

SHG at 0.53207 μm wavelength (see Fig. 3a) arised in the $Gd_2(MoO_4)_3$. To our knowledge, such a combined SRS experiment with insulating crystals has not been performed before.

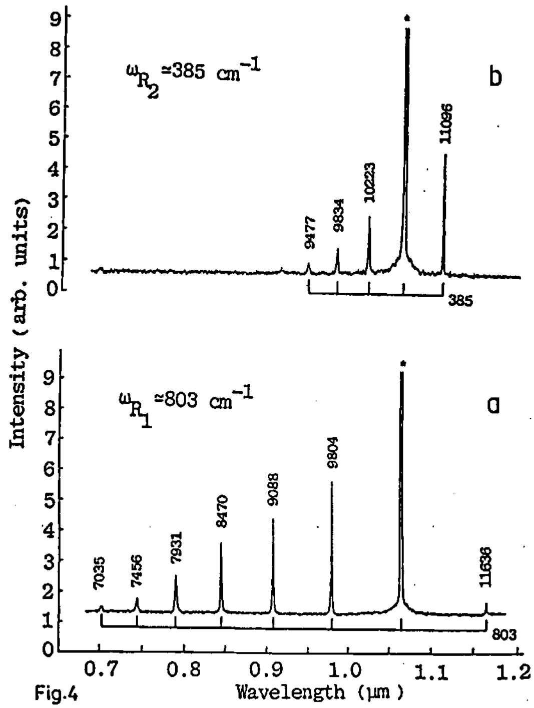

Fig.4

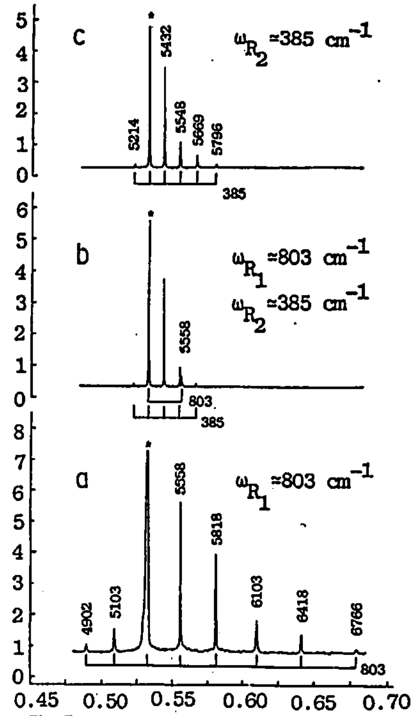

Fig.5

Fig. 4. Orientational SRS spectra of trigonal $LaBGeO_5$ crystal at 300 K and under IR pumping ($\lambda_p = 1.06415$ μm): (a) — excitation along the *a*-axis with polarization vector along the *b*-axis; (b) — excitation along the *b*-axis with polarization vector along the *a*-axis. The connection of Stokes and anti-Stokes generation lines with SRS-active vibration modes of a crystal with frequencies 385 and 803 cm^{-1} are indicated by brackets. Other notations as in Fig. 2.

Fig. 5. Orientational SRS spectra of trigonal $LaBGeO_5$ crystal at 300 K and under "green" pumping ($\lambda_p = 1.06415$ μm): (a) —excitation along the *a*-axis with polarization vector along the *b*-axis; (b) — excitation along the *b*-axis with polarization vector along the *a*-axis; (c) — excitation along the *c*-axis with polarization vector along the *a*-axis. Other notations as in Figs. 2. and 4.

3. SHG due to Cerenkov-type phase matching in trigonal $LaBGeO_5$ crystal

For SHG and SHG with Cerenkov phase matching (CSHG) in $LaBGeO_5$ single crystals we used three types of high-intensive pumping sources. Among them there were the commercial pico- and nano-second crystalline lasers based on $Al_2O_3:Ti^{3+}$ (Continuum TR-TW, USA) and $Y_3Al_5O_{12}:Nd^{3+}$ (LTIPCH-7 with IZ-25 head, Russia) emitted at fixed wavelengths of about 0.79 μm and 1.06415 μm, respectively (Fig. 6), as well as the hyper-continuum femto-second coherent-radiation laser station designed in the Institute for Laser Science of the

Tokyo University of Electro-Communications (Fig. 7) [9]. As is seen from the latter figure, a super-intense 2 TW $Al_2O_3:Ti^{3+}$ laser was utilized as a short-pulse generator in the hyper-continuum station. It produced pulses of about 125 fs with 250 mJ energy at *ca.* 0.79 μm wavelength. Multiple self-focusing spots appear 3 m behind the pulse-compressor of grating pair due to nonlinear refractive index of the air. The multi-spot beam was directed with an $r = 10$ m Au-mirror into 8-m traveling tube filled with atmospheric pressure Kr gas. This ultra-broad band coherent light was generated by a self-phase modulation owing to the optical Kerr effect in the Kr gas. Typically the continuum has a spectral intensity of about 0.1 GW/Å and a pulse width of *ca.* 150 fs for the visible and near-IR ranges. A laser wavelength of the continuum was selected by a pair of BK-7 dispersion prism and moving 2-mm optical slit. The spectral characteristics of the collinear SHG and conical ring CSHG of the $LaBGeO_5$ single crystals were measured by a fiber-coupled grating S-10 monochromator and multi channel spectro-analyzer Hamamatsu PMA-11 which was provided with a cooled Si-CCD array for the spectral range 0.3–0.9 μm.

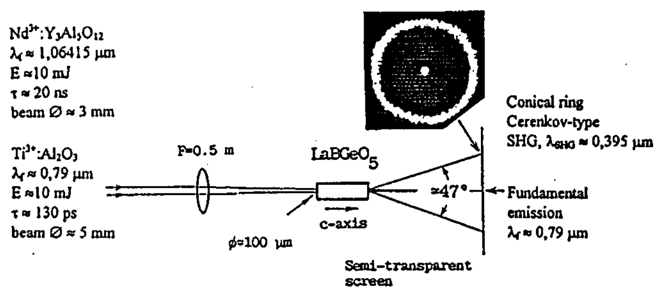

Fig. 6. Schematic experimental set-up for CSHG excitation with acentric $LaBGeO_5$ crystal with fixed wavelength nano- ($\lambda_p = 1.06415$ μm) and picosecond ($\lambda_p \approx 0.79$ μm) pumping. For explanation see the text.

In the case of pumping at fixed wavelength of about 0.79 or 1.06415 μm the $LaBGeO_5$ crystal oriented along the *c*-axis with high reliability generated one conical CSHG ring at about 0.395 or 0.53207 μm, respectively (Fig. 6). The phase matching condition for such Cerenkov-type SHG is

$$\cos\theta_C = \frac{v_{SHG}}{v_p} = \frac{n_p}{n_{SHG}},$$

where v_p, v_{SHG}, and n_p, n_{SHG} are the phase velocities and refractive indexes for the pumping (fundamental) and SHG waves, respectively, and $2\theta_C$ is the Cerenkov cone angle. For the deep-red pumping this $2\theta_C^{ex}$ (external) angle for the $LaBGeO_5$ was measured of about 47°, and for one-micron pumping $2\theta_C^{ex} \approx 35°$. Under the near-IR continuum (broad-band centered at about 0.92 μm) pumping the $LaBGeO_5$ crystal has emitted multicolor Cerenkov ring ("Cerenkov rainbow"). In this case our nonlinear boro-germanate besides their power CSHG can generate also weak one-color conventional collinear SHG (SHG$\approx$ 0.424 μm, this is a critical wavelength for SHG with the *ee–o* type phase matching [10]. Of course, with the

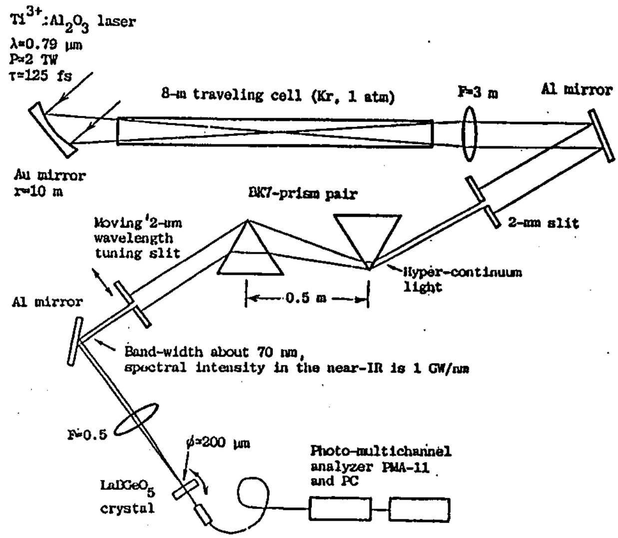

Fig. 7. Schematic experimental set-up for CSHG excitation in acentric $LaBGeO_5$ crystal with broad-band continuum femtosecond pumping. For explanation see the text.

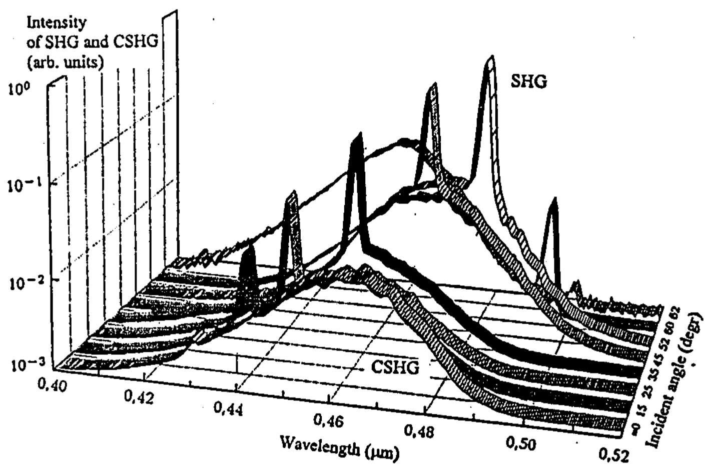

Fig. 8. Several SHG (narrow peaks) and CSHG spectrogramms for trigonal nonlinear $LaBGeO_5$ crystal at 300 K under femtoseconds broad-band continuum pumping.

change of the incident angle for the pumping beam (by rotations of the sample) the collinear SHG wavelength will be also tuned (Fig. 8).

In conclusion, we have demonstrated that two ferroelectrics $LaBGeO_5$ and $Gd_2(MoO_4)_3$ are very attractive as high-gain Raman and promising nonlinear media for quantum electronics. The observed multiple Stokes and anti-Stokes SRS and efficient SHG due to Cerenkov-type phase matching are new manifestation of nonlinear interaction in insulating crystal hosts. Our study also indicated that activated (by Ln^{3+} ions) crystals having considerable quadratic and cubic susceptibilities open up a broad range of opportunities for creation new types of crystalline lasers.

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