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Discrete and continuous tuning in the visible range by intracavity stimulated Raman scattering in nonlinear crystals

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Abstract. An analysis is reported of the effectiveness of the method of expanding the range of frequencies generated in a laser system by stimulated Raman scattering and subsequent frequency summation. It is shown that the most promising are noncentrosymmetric crystals with a strong electronic nonlinearity. Experimental results are reported on stimulated Raman scattering and nonlinear summation in LiIO$_3$, LiNbO$_3$, KDP (potassium dihydrogen phosphate), and KTP (potassium titanyl phosphate) crystals.

Stimulated Raman scattering (STRS) and nonlinear summation are used widely, the former mainly to reduce the frequency and the latter to increase the frequency, in broadening the range of frequencies generated in laser systems. Combining these two methods makes it possible to expand even further the range of frequencies of coherent monochromatic radiation.

We reported earlier [1] the use of a nonlinear LiIO$_3$ crystal in the generation of coherent radiation of frequencies $2\nu_L + m\nu_{ph}$ ($m = 0, \pm 1, \ldots$; ν_L is the laser radiation frequency; ν_{ph} is the frequency of phonon vibrations of a crystal). Both processes (STRS and nonlinear summation) occur simultaneously in this crystal when it is placed in front of the laser cavity. The present paper reports an investigation of the feasibility of extending this frequency-widening method to other crystals†.

It is known that the gain G_S of the Stokes wave in the case of STRS by a vibrational mode m of a crystal can be written in the form [2]

$$G_S = \frac{32\pi^2 c^3}{\hbar\omega_S^3} \frac{S_m/d\Omega}{n_L n_S (n_0^{(m)} + 1)\Gamma} I_L \,, \qquad (1)$$

where I_L is the intensity of the laser (scattered) radiation; $n_{L,S}$ are the refractive indices at the laser (ω_L) and Stokes (ω_S) frequencies; $n_0^{(m)} = [\exp(\hbar\omega_m/kT) - 1]^{-1}$ is the Bose factor; Γ is the width of a spontaneous Raman scattering (SPRS) line for the same vibrational mode;

$$S_m = e_i^{(S)} l_j^{(L)} S_{ijk}^{(m)} l_k^{(m)} \;; \qquad (2)$$

$S_{ijk}^{(m)}$ is the efficiency of the SPRS by the vibrational mode m representing phonons with the $\boldsymbol{e}^{(m)}$ polarisation [summation over repeated indices that are not enclosed in parentheses is implied here]. The SPRS efficiency is understood to be the ratio of the total power of the radiation scattered with the polarisation $\boldsymbol{e}^{(S)}$ into a solid angle $d\Omega$ in a crystal of length l, to the power of the incident radiation with the polarisation $\boldsymbol{e}^{(L)}$.

The SPRS efficiency is in its turn governed by the polarisation $\boldsymbol{P}_{NL}(\omega_S)$ at the Stokes frequency, which appears as a result of a nonlinear interaction with the electron and nonlinear subsystems in a unit cell of a crystal. The main contribution to the nonlinear polarisation of centrosymmetric crystals comes from scattering by what is known as the deformation potential of the vibrations of ions in the mode m, which gives rise to a periodic deformation of the electron shells of the ions and, as a result, to a periodic modulation of the polarisability (and permittivity) of the unit cell. Moreover, parametric scattering is possible in crystals with a significant electronic nonlinearity and such scattering can also contribute to the polarisation at the Stokes frequency.

The nonlinear polarisation at the Stokes frequency can therefore be described phenomenologically by an expression consisting of two terms [3]:

$$\begin{aligned} p_i^{NL}(\omega_S) = {} & r_{ijk}^{(m)} E_j(\omega_L) Q_k^{(m)*} \\ & + \chi_{ijk}(\omega_S; \omega_L, \omega_S - \omega_L) E_j(\omega_L) E_k^*(\omega_L - \omega_S) \,, \end{aligned} \qquad (3)$$

where $Q_k^{(m)}$ are the normal coordinates of the vibrational mode m of the investigated crystal; $\chi_{ijk}(\omega_1 + \omega_2; \omega_1, \omega_2)$ is the nonlinear susceptibility tensor, which at $\omega_1 = \omega_2$ is identical with the tensor describing the generation of the second harmonic. The tensor $r_{ijk}^{(m)}$ is determined by the characteristics of the phonons in the crystal (SPRS phonon scattering, oscillator strengths, etc.).

In noncentrosymmetric (piezoelectric) crystals some of the vibrational modes are polariton in nature, i.e. the vibrations of the lattice ions give rise to a dipole moment vibrating at the same frequency and, consequently, to a low-frequency (infrared) electromagnetic wave in the crystal. Such modes are active both in the infrared absorption and in the Raman scattering. The macroscopic infrared field generated in a noncentrosymmetric crystal results (because of the electrooptic effect which occurs in these crystals) in additional modulation of the permittivity and an additional contribution to the nonlinear polarisability at the Stokes frequency. Phenomenological theory shows that this additional contribution to the polarisation can be allowed for on the assumption that

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the displacements $Q_k^{(m)}$ of the ions are proportional to the intensity of the low-frequency infrared field:

$$Q_k^{(m)} = \beta^{(m)} E_k \ . \tag{4}$$

Then, substituting Eqn (4) into Eqn (3), we find that the nonlinear polarisation for the scattering by the polariton mode m is described by

$$\begin{aligned} P_i^{\mathrm{NL}}(\omega_{\mathrm{S}}) &= [r_{ijk}^{(m)} \beta^{(m)*} + \chi_{ijk}] E_j(\omega_{\mathrm{L}}) E_k^*(\omega_{\mathrm{L}} - \omega_{\mathrm{S}}) \\ &= \alpha_{ijk}^{(m)} E_j(\omega_{\mathrm{L}}) E_k^*(\omega_{\mathrm{L}} - \omega_{\mathrm{S}}) \ . \end{aligned} \tag{5}$$

The efficiency of the SPRS on a polariton mode is then described by the nonlinear polarisability tensor $\alpha_{ijk}^{(m)}$ [4]:

$$S_{ijk}^{(m)} = \frac{\hbar\omega_m \omega_{\mathrm{S}}^4 [n_0^{(m)} + 1] \upsilon l \, \mathrm{d}\Omega}{64\pi^2 \varepsilon_0^2 c^4 K_k^{(m)}} |\alpha_{ijk}^{(m)}|^2 \ , \tag{6}$$

where υ is the volume of a unit cell of the crystal in question and $K_k^{(m)}$ is the elastic constant of the crystal along the k axis for the mode m.

It follows that the most promising media for the proposed method to convert coherent radiation frequencies are piezoelectric crystals with a strong electronic quadratic nonlinearity. The electrooptic effect enhances the STRS on some vibrational modes. The electronic nonlinearity may additionally increase (when constructive interference takes place) the gain of Eqn (1) and, moreover, it provides an opportunity for summing in the same crystal the resultant Stokes and anti-Stokes components. A strong dispersion of the polariton modes makes it possible also to tune the frequency continuously by rotating the crystal.

We investigated experimentally the intracavity collinear STRS with nonlinear summation in a number of noncentrosymmetric crystals used widely in frequency doubling. We used the mode-locking YAG:Nd^{3+} laser ($\lambda_{\mathrm{L}} = 1.06$ μm) earlier [5] and the method employed in our experiments was the same as in the previous investigation [1]. Table 1 lists the coherent radiation wavelengths obtained as a result of STRS followed by phase-matched summation in an LiIO$_3$ crystal.

The results listed for LiIO$_3$ were obtained partly for one intracavity crystal and partly for two crystals, where an intracavity crystal was used for STRS and the summation was carried out in an identical crystal outside the cavity (see Ref. [1]). In the case of KTP (potassium titanyl phosphate), KDP (potassium dihydrogen phosphate), and LiNbO$_3$ crystals the process of STRS occurred inside the cavity and the summation took place outside the cavity in the LiIO$_3$ crystal.

We were able to interpret fully the results listed in Table 1 for LiIO$_3$. The 820 and 342 cm^{-1} modes were found to have the E_2 symmetry and were infrared-inactive, whereas the 650 cm^{-1} mode had a mixed $(A + E_1)$ symmetry and, consequently, was polariton in nature. Rotation of the intracavity crystal enabled us to ensure continuous frequency tuning in the vicinity of the wavelengths 552, 573, and 596 nm, corresponding to the frequencies $2\nu_{\mathrm{L}} - \nu_{\mathrm{ph}}$, $2\nu_{\mathrm{L}} - 2\nu_{\mathrm{ph}}$, and $2\nu_{\mathrm{L}} - 3\nu_{\mathrm{ph}}$. The tuning ranges were 1.5, 3.5, and 4.4 nm, in good agreement with the tuning curve of the polariton frequency deduced theoretically.

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Table 1.

Crystal	ν_{ph}/cm^{-1}	$1/(2\nu_{\mathrm{L}} + m\nu_{\mathrm{ph}}) = \lambda$ /nm									
		$m = 4$	$m = 3$	$m = 2$	$m = 1$	$m = 0$	$m = -1$	$m = -2$	$m = -3$	$m = -4$	$m = -5$
LiIO$_3$	820	–	–	489.2	509.6	532	556.3	583.0	612.4	664.3	680.5
	650	–	–	–	–	532	551.3 – 552.8	571.0 – 574.5	593.3 – 597.7	–	–
	342	–	–	–	–	532	541.8	552.0	–	–	–
	492	–	–	–	518.5	532	546.3	561.4	577.7	–	–
LiNbO$_3$	115	–	–	–	529	532	535.3	538.5	541.7	–	–
KTP	562	–	–	–	516.5	532	548.6	565.8	584.7	–	–
KDP	23	529.4	530	530.7	531.4	532	532.7	533.5	–	–	–

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