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Intracavity stimulated Raman scattering by polaritons in an $LiNbO_3$ crystal

A M Bel'skiĭ, I M Gulis, K A Saechnikov

Abstract. An experimental investigation was made of stimulated Raman scattering by polaritons in a crystal of $LiNbO_3$ inside the cavity of an Nd : YAG system. The scattering was followed by frequency summation in an $LiIO_3$ crystal. Coherent radiation was generated at 16 wavelengths in the visible range. A scattering mechanism accounting for these results was proposed.

We recently reported [1] preliminary results aimed to widen the range of coherent emission from an Nd^{3+} : YAG laser in the visible range by intracavity stimulated Raman scattering (SRS) in nonlinear piezoelectric crystals, involving the summation of the SRS components with one another and with the fundamental frequency. The present paper reports a more detailed investigation of this mechanism for extending the range of coherent emission with the aid of an $LiNbO_3$ crystal.

The apparatus was similar to that described by us in Ref. [2]. Inside the cavity of a passively mode-locked Nd^{3+} : YAG laser we placed an $LiNbO_3$ crystal. This crystal was cut in the direction of the optimal phase matching for generation of the second harmonic of the fundamental-frequency radiation ($\lambda_0 = 1.064$ μm). The crystal was oriented so that the direction of the electric vector of the laser radiation made an angle of $\sim 45°$ with the principal plane. The radiation emerging from the cavity was passed through an $LiIO_3$ crystal, which was cut along the direction of phase matching for generation of the second harmonic of the fundamental-frequency radiation. Rotation of the lithium iodate crystal made it possible to align it, so as to ensure phase matching for the summation of the Stokes and anti-Stokes components of the output radiation if intracavity SRS occurred in $LiNbO_3$.

Our experimental results are listed in Table 1. The first column gives the radiation wavelengths generated when the angle θ between the direction of propagation of the fundamental-frequency radiation in $LiNbO_3$ and the optic axis was close to the phase-matching angle for second-harmonic generation. Rotation of the lithium niobate crystal in such a way that the angle θ was close to 90° resulted in additional generation of coherent radiation at the wavelengths listed in the third column of Table 1.

An analysis of the frequency shifts at these wavelengths led us to the conclusion that propagation of laser radiation in a direction close to that corresponding to phase matching in $LiNbO_3$ resulted in multistage ('cascade') SRS at the vibrational (polariton) mode $\nu_{p1} \sim 492$ cm^{-1}, giving rise to the first and second Stokes components and to the first anti-Stokes component. The subsequent summation of these components in $LiIO_3$ generated the radiation at the listed wavelengths, in accordance with the mechanism given in the second column of Table 1. When the angle θ was close to 90°, SRS in $LiNbO_3$ occurred simultaneously at two vibrational modes: at the $\nu_{p1} \approx 492$ cm^{-1} mode, giving rise to the Stokes and anti-Stokes components listed above, and at the mode $\nu_{p2} \approx 115$ cm^{-1}, giving rise to the first, second, and third Stokes component, in accordance with the mechanism given in the fourth column of Table 1.

An investigation of the polarisation of the fundamental-frequency radiation and of the radiation at the wavelengths listed in Table 1 demonstrated that excitation of the vibrations with $\nu_{p1} \approx 492$ cm^{-1} resulted in the o – o scattering (the fundamental-frequency and scattered radiations behaved as ordinary waves in $LiNbO_3$), but when vibrations with $\nu_{p2} \approx 115$ cm^{-1} were excited, the scattering was of the e – o type (the laser radiation propagated as an extraordinary wave).

All the vibrational modes of the crystal lattice of $LiNbO_3$ active in the Raman scattering were of the polariton nature and had been attributed to vibrations of the A_1 and E types [3]. The scattering tensors for a crystal of the C_{3v}

Table 1. Parameters of intracavity SRS in $LiNbO_3$ and extracavity frequency summation in $LiIO_3$.

λ/nm	Summation mechanism ($\nu_{v1} = 492$ cm^{-1})	λ/nm	Summation mechanism ($\nu_{v2} = 115$ cm^{-1})
505.7	$(\nu_L + \nu_{v1}) + (\nu_L + \nu_{v1})$	526.1	$(\nu_L + \nu_{v2}) + (\nu_L + \nu_{v2})$
		529.0	$\nu_L + (\nu_L + \nu_{v2})$
518.5	$\nu_L + (\nu_L + \nu_{v1})$	532.0	$\nu_L + \nu_L$
		535.3	$\nu_L + (\nu_L - \nu_{v2})$
532.0	$\nu_L + \nu_L$	538.5	$\nu_L + (\nu_L - 2\nu_{v2})$
			$(\nu_L - \nu_{v2}) + (\nu_L - \nu_{v2})$
546.3	$\nu_L + (\nu_L - \nu_{v1})$	541.7	$(\nu_L + (\nu_L - 3\nu_{v2})$
			$(\nu_L - \nu_{v2}) + (\nu_L - 2\nu_{v2})$
		545.2	$(\nu_L - \nu_{v2}) + (\nu_L - 3\nu_{v2})$
561.4	$(\nu_L - \nu_{v1}) + (\nu_L - \nu_{v1})$		$(\nu_L - 2\nu_{v2}) + (\nu_L - 2\nu_{v2})$
		548.7	$(\nu_L - 2\nu_{v2}) + (\nu_L - 3\nu_{v2})$
	$\nu_L + (\nu_L - 2\nu_{v1})$		$(\nu_L - \nu_{v1}) + (\nu_L - \nu_{v2})$
		552.5	$(\nu_L - 3\nu_{v2}) + (\nu_L - 3\nu_{v2})$
577.7	$(\nu_L - \nu_{v1}) + (\nu_L - 2\nu_{v1})$		$(\nu_L - \nu_{v1}) + (\nu_L - 2\nu_{v2})$
		557.1	$(\nu_L - \nu_{v1}) + (\nu_L - 3\nu_{v2})$
595.0	$(\nu_L - 2\nu_{v1}) + (\nu_L - 2\nu_{v1})$	564.5	$(\nu_L - 2\nu_{v1}) + (\nu_L - \nu_{v2})$

A M Bel'skiĭ, I M Gulis, K A Saechnikov Belarus State University, pr. F Skoriny 4, 200050 Minsk, Belarus

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symmetry [4] enabled us to determine readily the dependence of the efficiency of the scattering by ordinary and extraordinary polaritons for various polarisations of the incident and scattered radiations.

Our analysis showed that the scattering mechanism resulting in the emission at the wavelengths listed in Table 1 is as follows. When the laser radiation propagates in the $LiNbO_3$ crystal at an angle θ close to the phase matching angle for second-harmonic generation, the intensity of the ordinary wave is higher than the threshold value for SRS by an ordinary polariton (E symmetry) with $\nu_{p1} \approx 492$ cm^{-1}. When the angle θ is increased, the intensity of the o–o scattering by an ordinary polariton does not change, but the intensity of the e–o scattering increases and at some angle close to $\theta \sim 90°$ the intensity of the e component of the fundamental-frequency radiation is sufficient for the excitation of SRS by an ordinary polariton with $\nu_{p2} \approx 115$ cm^{-1}.

An additional confirmation of this scattering mechanism is provided by a comparison of the experimentally observed frequency shifts in the scattered radiation with the theoretically found frequencies of the polariton vibrations which may be excited in a crystal of $LiNbO_3$ by collinear scattering of the $\lambda_0 = 1.064$ μm radiation.

Fig. 1 gives the dispersion curves of the ordinary polaritons in $LiNbO_3$. The fundamental frequencies of the transverse and E vibrational modes are taken from Ref. [5] and the theory of dispersion of the polariton vibrations is based on Ref. [6]. The wave vector of the excited polariton vibrations is governed by the law of conservation of momentum in stimulated Raman scattering. If we ignore the difference between the refractive indices for the laser radiation and the Stokes component, this collinear scattering law can be written in the form

$$K_p = 2\pi n_o(\lambda_0)\nu_p \tag{1}$$

for the o–o mechanism and

$$K_p = \pm 2\pi\left[\frac{n_o(\lambda_0) - n_e(\lambda_0)}{\lambda_0} - n_o(\lambda_0)\nu_p\right] \tag{2}$$

for the e–o mechanism. Fig. 1 shows the straight lines plotted on the basis of formulas (1) and (2); they are labelled *1* and *2*. The polariton frequency $\nu_{p1} \approx 492$ cm^{-1} corresponds to the o–o scattering by the polariton branch characterised by $\nu_{LO} \approx 454$ cm^{-1} and $\nu_{TO} \approx 582$ cm^{-1}. In the case of the polariton with $\nu_{p2} \approx 115$ cm^{-1}, the e–o scattering by the first polariton branch yields a frequency which is too low ($\nu_{p1} \approx 80$ cm^{-1}) and the scattering by the second branch yields a frequency which is too high (lines labelled *2* in Fig. 1).

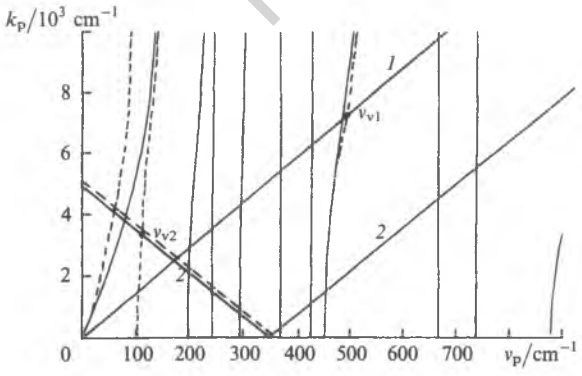

Figure 1. Dispersion of ordinary polaritons in an $LiNbO_3$ crystal, calculated on the basis of the results reported in Ref. [5] (continuous curves) and using the results of Ref. [7] (dashed curves). Line *1* corresponds to the law of conservation of momentum in the o–o scattering and lines labelled *2* correspond to the e–o scattering (dashed line *2* represents the law of conservation of momentum when the photorefraction is taken into account).

Better results are obtained if it is assumed that the fundamental frequency of the transverse E vibrational mode is less than $\nu_{TO} \approx 155$ cm^{-1}. The existence of a mode with this frequency is supported by some of the investigations of the spectra of spontaneous Raman scattering in a crystal of $LiNbO_3$ [7, 8], although its existence is rejected in Ref. [5]. The dashed curves in Fig. 1 identify the two lower branches of the polariton vibrations and the branch corresponding to the $\nu_{p1} \approx 492$ cm^{-1} polariton on the assumption that the lowest fundamental frequencies are $\nu_E^{TO} \approx 90$ cm^{-1} and $\nu_E^{LO} \approx 105$ cm^{-1}. The other fundamental frequencies were taken from Ref. [5], subject to the correction for the results given in Ref. [7]: $\nu_E^{TO} \approx 155$, 238, 262, 322, 371, 431, 585, and 630 cm^{-1}; $\nu_E^{LO} \approx 198$, 243, 298, 371, 428, 454, 621, and 880 cm^{-1}. The point of intersection of line *2* with the second polariton branch gives $\nu_{p2} \approx 110$ cm^{-1}, which is much closer to the experimental value 115 cm^{-1}.

The $LiNbO_3$ crystal is a photorefractive medium [9, 10], so that a more accurate comparison of the theoretical and experimental results has to take account of changes in the refractive indices under the influence of high-intensity optical radiation. According to Ref. [9], the main result of the photorefraction in lithium niobate is a reduction in the extraordinary refractive index n_e by $\sim 10^{-3}$ with hardly any change in the ordinary refractive index n_o. For this reason the photorefraction has almost no influence on the polariton frequency in the case of the o–o scattering [see formula (1)], but there is some increase in the polariton frequency in the e–o scattering (see Fig. 1, where dashed line *2* is plotted on the assumption that the photorefraction reduces n_e by 0.001). It should be pointed out also that the photorefraction increases most (by 5–10 cm^{-1}) the frequencies of the polaritons excited by collinear scattering on the lower polariton branch.

The dispersion of the refractive indices gives rise to a dependence of the frequency of the excited polariton vibrations on the wavelength λ of the exciting radiation. It follows from the law of conservation of momentum that in the normal dispersion case the polariton frequency decreases as λ is increased. For the ν_{p2} polariton, the wavelengths listed in Table 1, we obtained ~116, 111, and 110 cm^{-1} for the excitation by the laser radiation, and by the first and second Stokes components, respectively. In the case of the o–o scattering the influence of dispersion is negligible.

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