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Effect of polariton propagation on spectra of SRS amplification and CARS from polaritons

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***Abstract*. The properties of *k* spectra of SRS amplification and CARS from polaritons caused by 'running out' of polaritons from the volume of their interaction with incident light beams are theoretically analysed. It is shown that the shape and width of the spectra depend on the relation between the size of the overlap region of exciting waves in a crystal along the direction of polariton propagation and the mean free path of polaritons. The conditions are found under which the widths of SRS amplification and CARS spectra give information on the polariton decay.**

Keywords: *coherent anti-Stokes scattering, amplification upon stimulated Raman scattering, polaritons, nonlinear-optical mixing, shape and width of spectra.*

1. Introduction

Coherent anti-Stokes Raman scattering (CARS) and spectroscopy based on the detection of amplification spectra of a Stokes wave due to stimulated Raman scattering (SRS) (the SRS amplification) are extensively used for solving many scientific and applied problems (see, for example, Ref. [1]). These methods were also employed for studying phonon polaritons, which are active in CARS and SRS amplification spectra in crystals without the centre of symmetry [1 – 12].

The spectra of SRS amplification and CARS from polaritons exhibit a number of specific features compared to the spectra of scattering by nonpolar excitations in a medium. Some of these features are related to the property of polaritons to propagate in a medium over macroscopic distances, which are typically from several tens to several hundreds of micrometers, while in the region of the upper and lower dispersion branches far from the dipole phonon resonances, the mean free path of polaritons can achieve several centimetres.

The property of polaritons to propagate over macroscopic distances was used, in particular, in CARS spectroscopy in the geometry when the regions of biharmonic (in the field of two waves) excitation of polaritons and probing of excited coherent polaritons by a probe wave are separated in a crystal [13 – 18]. Experiments performed using such a geometry open up new possibilities of CARS spectroscopy of polaritons and are of interest from the point of view of energy transfer in crystals.

This paper is devoted to the study of the influence of the propagation of polaritons over macroscopic distances on the shape and width of SRS amplification and CARS spectra. We also found the conditions under which the width of these spectra can give information on the decay of polaritons. Note that the broadening of the spectra caused by 'running out' of polaritons from the interaction region was found in recent experiments on SRS amplification [11] and CARS [18] from polaritons.

2. Dependence of spectra on the relation between the length of a crystal and the mean free path of polaritons (plane-wave approximation)

Let us assume that a scattering noncentrally symmetric medium is a layer of thickness L, and plane waves with frequencies ω_L and ω_s and wave vectors $\boldsymbol{k}_L$ and $\boldsymbol{k}_s$ are propagating along the normal to the layer boundary (along the z axis). CARS is a nonlinear-optical mixing, which is accompanied by the generation of a wave at frequency $\omega_a = 2\omega_L - \omega_s$ while polaritons are observed in CARS spectra if the condition

$$\omega_L - \omega_s \equiv \omega \approx \omega_p, \quad \boldsymbol{k}_L - \boldsymbol{k}_s = \boldsymbol{k} \approx \boldsymbol{k}_p, \tag{1}$$

is fulfilled, where ω_p and $\boldsymbol{k}_p$ are the frequency and the wave vector of polaritons. Upon SRS amplification, a weak Stokes wave with frequency ω_s is amplified in the intense pump field with frequency ω_L, provided the condition (1) is valid. We assume that the medium absorbs polaritons, being transparent for the rest of the waves.

During CARS, a coherent polariton wave is excited in the field of two waves (biharmonic pumping) $E_L(\omega_L, \boldsymbol{k}_L)$ and $E_s(\omega_s, \boldsymbol{k}_s)$ due to the quadratic nonlinear susceptibility $\chi^{(2)}(\omega_p = \omega_L - \omega_s)$ of the medium and then the probe wave $E_L(\omega_L, \boldsymbol{k}_L)$ is scattered by coherently excited polaritons. The field of the excited polariton wave calculated in the approximation of the specified amplitudes of incident waves can be represented as a sum of a particular solution of the inhomogeneous wave equation (the stimulated wave

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with the wave vector $\boldsymbol{k} \equiv \boldsymbol{k}_{\rm L} - \boldsymbol{k}_{\rm s}$) and the general solution of the homogeneous equation (the free wave with the wave vector $\boldsymbol{k}_{\rm p}$) [19]:

$$\tilde{E}_{\rm p}(\omega, k) = E_{\rm p}(\omega, k)\mathrm{e}^{-\mathrm{i}\omega t} = 4\pi\, \frac{\chi^{(2)}(\omega = \omega_{\rm L} - \omega_{\rm s})}{k^2c^2/\omega^2 - \varepsilon(\omega)}$$

$$\times E_{\rm L}E_{\rm s}^*\left[\mathrm{e}^{\,\mathrm{i}(k_{\rm L}-k_{\rm s})z} - \mathrm{e}^{\,\mathrm{i}k_{\rm p}z}\right]\mathrm{e}^{-\mathrm{i}\omega t}$$

$$\approx \frac{2\pi\omega^2}{c^2k'_{\rm p}}\frac{\chi^{(2)}(\omega_{\rm p} = \omega_{\rm L} - \omega_{\rm s})}{\Delta k_{\rm p} - \mathrm{i}k''_{\rm p}}$$

$$\times E_{\rm L}E_{\rm s}^*\left[\mathrm{e}^{\,\mathrm{i}(k_{\rm L}-k_{\rm s})z} - \mathrm{e}^{\,\mathrm{i}k'_{\rm p}z - k''_{\rm p}z}\right]\mathrm{e}^{-\mathrm{i}\omega t}, \tag{2}$$

where

$$k_{\rm p} = k'_{\rm p} + \mathrm{i}k''_{\rm p}; \quad \left(\frac{\boldsymbol{k}_{\rm p}c}{\omega}\right)^2 = \varepsilon(\omega);$$

$$\Delta k_{\rm p} = k - k'_{\rm p} \equiv k_{\rm L} - k_{\rm s} - k'_{\rm p};$$

$\varepsilon(\omega)$ is the complex permittivity of the crystal. The approximate equality in (2) becomes exact if $k'_{\rm p} \gg k''_{\rm p}, \Delta k_{\rm p}$.

By using a standard method of truncated equations [19], we obtain in the approximation of the specified amplitudes of wave at frequencies $\omega_{\rm L}$ and $\omega_{\rm s}$ the equation for the anti-Stokes-wave amplitude at the frequency $\omega_{\rm a}$

$$\frac{\partial E_{\rm a}}{\partial z} = \frac{\mathrm{i}2\pi\omega_{\rm a}^2}{c^2k_{\rm a}}\Big[\chi^{(3)}(\omega_{\rm a} = 2\omega_{\rm L} - \omega_{\rm s})E_{\rm L}^2E_{\rm s}^*\mathrm{e}^{\mathrm{i}\Delta k_{\rm a}z}$$

$$+ \chi^{(2)}(\omega_{\rm a} = \omega_{\rm L} + \omega)E_{\rm L}E_{\rm p}(\omega, k)\mathrm{e}^{\,\mathrm{i}(k_{\rm L}-k_{\rm a})z}\Big], \tag{3}$$

where $\chi^{(3)}$ is the cubic nonlinear susceptibility; $\Delta\boldsymbol{k}_{\rm a} = 2\boldsymbol{k}_{\rm L} - \boldsymbol{k}_{\rm s} - \boldsymbol{k}_{\rm a}$; $E_{\rm p}(\omega, k)$ is described by expression (2). As a result, we obtain after integration

$$\frac{|E_{\rm a}|^2}{|E_{\rm L}^2E_{\rm s}^*|^2} = |A_{\rm a}|^2L^2\left|\eta\frac{\mathrm{e}^{\mathrm{i}\Delta k_{\rm a}L} - 1}{\Delta k_{\rm a}L} + \frac{1}{(\Delta k_{\rm p} - \mathrm{i}k''_{\rm p})L}\right.$$

$$\left.\times\left[\frac{\mathrm{e}^{\mathrm{i}\Delta k_{\rm a}L} - 1}{\Delta k_{\rm a}} - \frac{\mathrm{e}^{\,\mathrm{i}(\Delta k_{\rm a} - \Delta k_{\rm p})L - k''_{\rm p}L} - 1}{\Delta k_{\rm a} - \Delta k_{\rm p} + \mathrm{i}k''_{\rm p}}\right]\right|^2, \tag{4}$$

where

$$A_{\rm a} = \frac{4\pi^2\omega_{\rm a}^2\omega^2}{c^4k_{\rm a}k'_{\rm p}}\chi^{(2)}(\omega_{\rm a} = \omega_{\rm L} + \omega)\chi^{(2)}(\omega = \omega_{\rm L} - \omega_{\rm s});$$

$$\eta = \frac{c^2k'_{\rm p}}{2\pi\omega^2}\frac{\chi^{(3)}(\omega_{\rm a} = 2\omega_{\rm L} - \omega_{\rm s})}{\chi^{(2)}(\omega_{\rm a} = \omega_{\rm L} + \omega)\chi^{(2)}(\omega = \omega_{\rm L} - \omega_{\rm s})}. \tag{5}$$

The parameter η determines the relative contributions of direct four-photon and cascade three-photon processes to the CARS signal.

The equation for the wave amplitude at the frequency $\omega_{\rm s}$ in the field of the specified more intense pump wave with frequency $\omega_{\rm L}$ can be represented in the form

$$\frac{\partial E_{\rm s}}{\partial z} = \frac{\mathrm{i}2\pi\omega_{\rm s}^2}{c^2k_{\rm s}}\Big[\chi^{(3)}(\omega_{\rm s} = \omega_{\rm L} - \omega_{\rm L} + \omega_{\rm s})|E_{\rm L}|^2E_{\rm s}$$

$$+ \chi^{(2)}(\omega_{\rm s} = \omega_{\rm L} - \omega_{\rm p})E_{\rm L}E_{\rm p}^*(\omega, k)\mathrm{e}^{\,\mathrm{i}(k_{\rm L}-k_{\rm s})z}\Big], \tag{6}$$

where $E_{\rm p}(\omega, k)$ is described by expression (2). The solution of equation (6) has the exponential form

$$|E_{\rm s}|^2 = |E_{\rm s0}|^2\exp(g_{\rm s}|E_{\rm L}|^2L) \approx |E_{\rm s0}|^2(1 + g_{\rm s}|E_{\rm L}|^2L),$$

where

$$g_{\rm s} = -\frac{4\pi\omega_{\rm s}^2}{c^2k_{\rm s}}\mathrm{Im}\Big\{\chi^{(3)}(\omega_{\rm s} = \omega_{\rm L} - \omega_{\rm L} + \omega_{\rm s})$$

$$+ \frac{2\pi\omega^2}{c^2k_{\rm p}}\chi^{(2)}(\omega_{\rm s} = \omega_{\rm L} - \omega_{\rm p})\chi^{(2)*}(\omega = \omega_{\rm L} - \omega_{\rm s})$$

$$\times\left[\frac{1}{\Delta k_{\rm p} + \mathrm{i}k''_{\rm p}} - \frac{\mathrm{e}^{\mathrm{i}\Delta k_{\rm p}L - k''_{\rm p}L} - 1}{L(\Delta k_{\rm p} + \mathrm{i}k''_{\rm p})(\mathrm{i}\Delta k_{\rm p} - k''_{\rm p})}\right]\Big\}. \tag{7}$$

Below, we will analyse the k spectra, i.e., the dependence of signals of interest to us on the phase mismatch $\Delta\boldsymbol{k}_{\rm p}$ for the specified frequency ω. For simplicity, we will consider only the spectral region of polaritons in which the imaginary part of nonlinear susceptibility can be neglected. This is valid when the difference between the frequencies of polaritons and the dipole phonon resonance is equal to several widths of phonon lines. We also assume that $\Delta\boldsymbol{k}_{\rm a} = 0$ because CARS from polaritons is usually detected under phase matching conditions for the four-photon process, which corresponds to the maximum efficiency. In this case, the shape of CARS and SRS amplification spectra is described, according to (4) and (7), by the expressions

$$S(\Delta k_{\rm p}, k''_{\rm p}L) = \left|\mathrm{i}\eta + \frac{1}{\Delta k_{\rm p} - \mathrm{i}k''_{\rm p}}\left[\mathrm{i} + \frac{\mathrm{e}^{-\mathrm{i}\Delta k_{\rm p}L - k''_{\rm p}L} - 1}{(\Delta k_{\rm p} - \mathrm{i}k''_{\rm p})L}\right]\right|^2, \tag{8}$$

$$G(\Delta k_{\rm p}, k''_{\rm p}L) = -\mathrm{Im}\left[\frac{1}{\Delta k_{\rm p} + \mathrm{i}k''_{\rm p}} - \frac{\mathrm{e}^{\mathrm{i}\Delta k_{\rm p}L - k''_{\rm p}L} - 1}{L(\Delta k_{\rm p} + \mathrm{i}k''_{\rm p})(\mathrm{i}\Delta k_{\rm p} - k''_{\rm p})}\right]$$

$$= \frac{k''_{\rm p}}{\Delta k_{\rm p}^2 + k''^2_{\rm p}} + \frac{\Delta k_{\rm p}^2 - k''^2_{\rm p}}{(\Delta k_{\rm p}^2 + k''^2_{\rm p})^2L} \tag{9}$$

$$- \frac{(\Delta k_{\rm p}^2 - k''^2_{\rm p})\cos(\Delta k_{\rm p}L) + 2k''_{\rm p}\Delta k_{\rm p}\sin(\Delta k_{\rm p}L)}{(\Delta k_{\rm p}^2 + k''^2_{\rm p})^2L}\mathrm{e}^{-k''_{\rm p}L},$$

respectively.

By analysing CARS from polaritons, we will first consider the case when the contribution of the direct four-photon process is negligible compared to the contribution caused by cascade transitions, i.e., we assume that $\eta = 0$. In this case, expression (8) has the form

$$S(\Delta k_{\rm p}, k''_{\rm p}L, \eta = 0) = \left[\frac{\mathrm{e}^{-k''_{\rm p}L}\cos(\Delta k_{\rm p}L) - 1 + k''_{\rm p}L}{(\Delta k_{\rm p}^2 + k''^2_{\rm p})L}\right]^2$$

$$+ \left[\frac{\mathrm{e}^{-k''_{\rm p}L}\sin(\Delta k_{\rm p}L) - \Delta k_{\rm p}L}{(\Delta k_{\rm p}^2 + k''^2_{\rm p})L}\right]^2 = \frac{1}{\Delta k_{\rm p}^2 + k''^2_{\rm p}} + \frac{\mathrm{e}^{-2k''_{\rm p}L} - 1 + 2(k''_{\rm p}L - 1)\left[\mathrm{e}^{-k''_{\rm p}L}\cos(\Delta k_{\rm p}L) - 1\right] - 2\Delta k_{\rm p}L\mathrm{e}^{-k''_{\rm p}L}\sin(\Delta k_{\rm p}L)}{(\Delta k_{\rm p}^2 + k''^2_{\rm p})^2L^2}. \tag{10}$$

It follows from (9) and (10)* that in the limit of strong absorption of polaritons ($k_p''L \gg 1$), the shape of SRS amplification and CARS spectra (the first terms in (9) and (10), respectively) is Lorentzian with the FWHM equal to $\alpha_p = 2k_p''$, which permits the measurement of absorption (spatial decay) α_p of coherent polaritons directly from the experimental spectra.

When the absorption is very weak ($k_p''L \ll 1$), i.e., when the mean free path $L_p = 1/\alpha_p$ of coherent polaritons is much greater than the crystal length, the shapes of CARS and SRS amplification spectra noticeably change and are described by the functions

$$S\left(\Delta k_p, k_p''L \ll 1, \eta = 0\right) = \frac{1 - 2\mathrm{sinc}(\Delta k_p L) + \mathrm{sinc}^2(\Delta k_p L/2)}{\Delta k_p^2}, \qquad (11)$$

$$G\left(\Delta k_p, k_p''L \ll 1\right) = (L/2)\mathrm{sinc}^2(\Delta k_p L/2) \qquad (12)$$

respectively, with the FWHM

$$\delta k_S(\alpha_p = 0) \approx 6.954/L, \qquad (13)$$

$$\delta k_G(\alpha_p = 0) \approx 2\pi \times 0.885/L \approx 5.56/L. \qquad (14)$$

respectively.

One can see that in this case, the widths of the spectra no longer contain the information on the decay of polaritons.

In the general case, the shape of the spectrum is intermediate between a Lorentzian and the shape described by expression (11) or (12), while the width of the spectrum depends on $k_p''L$, i.e., on the ratio L/L_p of the crystal length to the polariton mean free path. The dependence of the spectra on L/L_p is shown in Fig. 1, where the polariton k spectra calculated by expressions (9) and (10) for different L/L_p and normalised to their maximum are presented. One can see that both the shape and width of the spectra change with changing L/L_p.

Fig. 2 shows the dependences of the spectral widths $\delta k_{S,G}/\alpha_p$ normalised to the absorption of polaritons on the ratio L/L_p. One can see that the CARS and SRS amplification spectra broaden with decreasing L/L_p. In particular, when the crystal length is equal to the mean free path of coherent polaritons ($L/L_p = 1$), the widths of the SRS amplification and CARS spectra become greater than the width of the spectrum determined by the polariton decay approximately by factors of 6 and 7.3, respectively. In this case, it is in fact impossible to determine the decay of polaritons from the SRS amplification and CARS spectra. It follows from our calculations presented in Fig. 2 that the broadening of CARS and SRS spectra caused by the propagation of polaritons does not exceed 10 % if $L > 22L_p$ and is no greater than 2 % if $L > 100L_p$.

Because the decay of polaritons, i.e., the ratio L/L_p is unknown preliminary in experiments, the question arises of to what extent the decay of polaritons is determined by the measured spectral widths. In this connection, the dependence of the ratio $\delta k_{S,G}/\alpha_p$ of the observed width of the spectrum to the decay of polaritons on the ratio $\delta k_{S,G}/\delta k_{S,G}(\alpha_p = 0)$ of the observed width of the spectrum to that for the zero decay of polaritons can be useful from the practical point of view. Such dependences calculated for the CARS and SRS amplification spectra by expressions (9), (10) and (13), (14) are shown in Fig. 3. In practice, one should first compare the widths $\delta k_{S,G}(\alpha_p = 0)$, calculated by expressions (13) and (14), with the observed widths $\delta k_{S,G}$ of the corresponding spectra and then, using the curves presented in Fig. 3, to measure the ratio $\delta k_{S,G}/\alpha_p$ and, hence, the required decay α_p of polaritons.

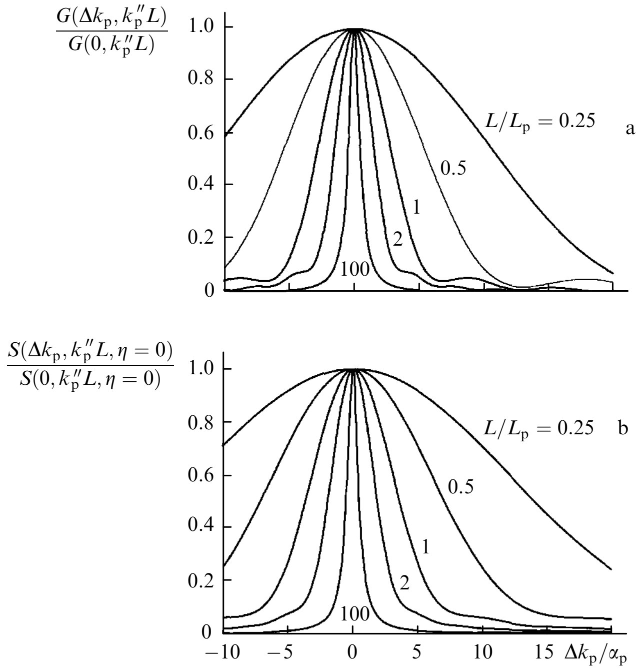

Figure 1. The k spectra of SRS amplification (a) and CARS (b) from polaritons calculated for the ratio L/L_p of the crystal length to the mean free path equal to 100, 2, 1, 0.5, and 0.25.

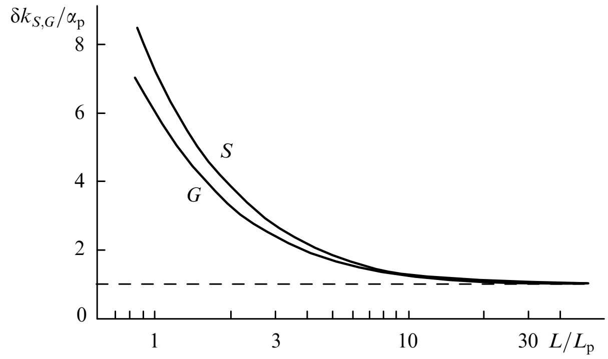

Figure 2. Ratio $\delta k_{S,G}$ of the widths of CARS (S) and SRS amplification (G) spectra to the decay α_p of polaritons as a function of the ratio of the crystal length L to the mean free path L_p of polaritons.

The contribution caused by four-photon processes ($\eta \neq 0$) leads to the interference of direct and cascade processes, resulting in the distortion of the spectrum [4,

*The expression analogous to (9), was obtained in paper [20] where spontaneous Raman scattering from polaritons was studied.

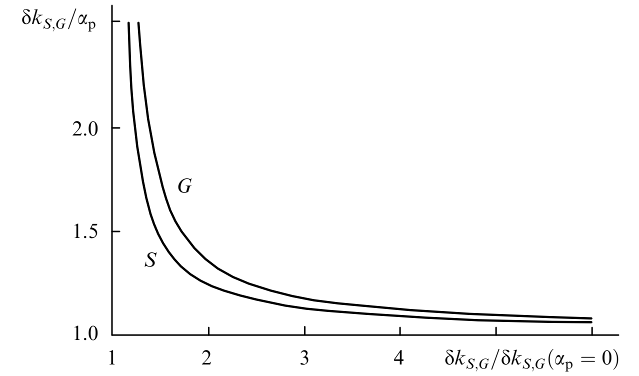

Figure 3. Ratio $\delta k_{S,G}/\alpha_p$ of the widths of CARS (S) and SRS amplification (G) spectra to the decay of polaritons as a function of the ratio of the widths of these spectra to the widths expected for the zero decay of polaritons $\delta k_{S,G}/\delta k_{S,G}(\alpha_p = 0)$.

7], which acquires the 'dispersion' form. Fig. 4 shows the CARS spectra normalised to the maximum, which were calculated by expression (8) taking into account direct four-photon processes with the relative contribution $\eta = 1$. Fig. 4 illustrates in fact the degree of transformation of the CARS spectrum when only the crystal length is changed. Note that in the case of strong decay of polaritons ($k''_p L \gg 1$), to which curve 100 corresponds in Fig. 4, expression (8) takes the simple form (see also Ref. [4])

$$S\big(\Delta k_p, k''_p L \gg 1\big) = \eta^2 + 2\eta \frac{\Delta k_p}{\Delta k_p^2 + k''^2_p} + \frac{1}{\Delta k_p^2 + k''^2_p}. \qquad (15)$$

In this limiting case, the CARS spectrum is no longer dependent on the crystal length.

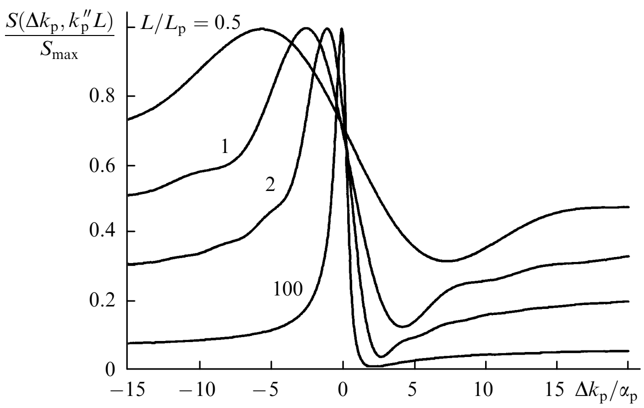

Figure 4. Spectra of CARS from polaritons calculated taking into account the contribution of four-photon processes ($\eta = 1$) for $L/L_p =$ 100, 2, 1, and 0.5.

The broadening of CARS and SRS amplification spectra caused by the propagation of polaritons within the framework of the model adopted can be experimentally observed using, for example, the collinear excitation of polaritons, which was applied in anisotropic crystals in the case of mutually orthogonal polarisations of exciting waves [6]. The studies of samples with a different thickness will allow us to observe the features of polariton spectra discussed above.

3. Effect of 'running out' of polaritons from the excited region (limited beams)

The transverse size of interacting waves in real experiments is always limited, and polaritons are, as a rule, excited when the wave vectors $\boldsymbol{k}_L$ and $\boldsymbol{k}_s$ are noncollinear. In this case, aperture effects, i.e., the 'running out' of polaritons from the region of excitation and probing, can play a significant role in the broadening of the spectra under study. Indeed, the synchronism condition $\Delta k_p = 0$ for polariton excitation is known [21] to be fulfilled for small angles φ between $\boldsymbol{k}_L$ and $\boldsymbol{k}_s$, which change from zero to several degrees, but because $k_L, k_s \gg k'_p$, the angle θ_p between $\boldsymbol{k}_L$ and $\boldsymbol{k}_p$ greatly exceeds φ (for $\varphi \neq 0$).

Therefore, the angle θ_p will be several tens of degrees even for small values of φ, i.e., the excited polaritons will propagate at a rather large angle to the direction of propagation of exciting beams. For this reason, polaritons can run out from the region of overlap of transversely limited exciting beams** at the distance L_a from the input face of the crystal, which is smaller than the crystal length L. As a result, for $L_a < L$, the broadening of the spectra should depend not on the ratio L/L_p (as in the case of plane waves) but on the ratio L_θ/L_p, where L_θ is the size of the excited region along the direction of propagation of polaritons. (For $L_a > L$, the expressions obtained above remain valid if α_p is replaced by $\alpha_p/\cos\theta_p$.)

We will describe the spectra of SRS amplification and CARS from polaritons taking into account aperture effects assuming that incident waves with the Gaussian transverse distribution propagate in the xz plane at small angle to the z axis, which is directed normally to the input face of the crystal. The truncated equation for the field amplitude of excited polaritons in the approximation of specified incident fields, mild focusing ($L \ll b$), and neglecting diffraction effects can be written in the form

$$\frac{\partial E_p(x,y,z)}{\partial z} + \rho \frac{\partial E_p(x,y,z)}{\partial x} + \frac{\alpha}{2} E_p(x,y,z)$$
$$= \mathrm{i} \frac{2\pi\omega^2}{c^2 k'_p \cos\theta_p} \chi^{(2)} E_L(x,y) E_s^*(x,y) \mathrm{e}^{\mathrm{i}\Delta k_{pz} z}, \qquad (16)$$

where $\rho = \tan\theta_p$; $\alpha = 2k''_p/\cos\theta_p = \alpha_p/\cos\theta_p$; Δk_{pz} is the projection of $\boldsymbol{k}_p = \boldsymbol{k}_L - \boldsymbol{k}_s - \boldsymbol{k}'_p$ on the z axis.

The solution of this equation for the boundary conditions

$$E_p(x,y,0) = 0, \quad E_{L,s}(x,y,0) = E_{L0,s0}\, \mathrm{e}^{-(x^2+y^2)/w^2} \qquad (17)$$

has the form

$$E_p(x,y,z) = \mathrm{i} E_{L0} E_{s0} \frac{2\pi\omega^2}{c^2 k'_p \cos\theta_p} \chi^{(2)} \exp\left(-\frac{y^2}{w^2}\right)$$
$$\times \int_0^z \exp\left[-\frac{\alpha}{2}(z - z') + \mathrm{i}\Delta k'_{pz} z'\right]$$
$$\times \exp\left\{-2\frac{\left[x - \rho(z - z')\right]^2}{w^2}\right\} \mathrm{d}z'.$$

**In experiments, the regime of 'mild focusing' is normally used, i.e., $L \ll b$ (b is the confocal parameter of the focusing optics) to exclude the effect of the divergence on the broadening of polariton spectra, and the exciting-beam diameter is usually from several hundreds of micrometers to approximately a millimetre.

By substituting (16) in (3) and (6), we obtain the required expressions describing the SRS amplification and CARS spectra

$$G = -\mathrm{Im}\left[\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\exp\left(-3\frac{y^2}{w^2}-\frac{x^2}{w^2}\right)\mathrm{d}x\mathrm{d}y \times \int_0^L\int_0^z \exp\left[\left(\mathrm{i}\Delta k_{\mathrm{p}z}-\frac{\alpha}{2}\right)(z-z')\right] \times \exp\left\{-2\frac{\left[x-\rho(z-z')\right]^2}{w^2}\right\}\mathrm{d}z\mathrm{d}z'\right], \tag{18}$$

$$S = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\mathrm{d}x\mathrm{d}y\left|\mathrm{i}L\eta\exp\left(-3\frac{x^2+y^2}{2}\right) - \exp\left(-3\frac{y^2}{w^2}-\frac{x^2}{w^2}\right) \times \int_0^L\mathrm{d}z\int_0^z\exp\left[-\left(\mathrm{i}\Delta k_{\mathrm{p}z}+\frac{\alpha}{2}\right)(z-z')\right] \times \exp\left\{-2\frac{\left[x-\rho(z-z')\right]^2}{w^2}\right\}\mathrm{d}z'\right|^2. \tag{19}$$

The results of numerical calculations of the ratio $\delta k_{S,G}/\alpha_{\mathrm{p}}$ of the width $\delta k_{S,G}$ of the spectra to the polariton decay α_{p} as a function of the ratio L_θ/L_{p} of the size L_θ of the excited region along the direction of polariton propagation to the mean free path L_{p} of polaritons performed by expressions (18) and (19) are presented in Fig. 5. The calculations were performed assuming that the contribution of direct four-photon processes is negligible compared to the contribution of cascade processes, i.e., $\eta = 0$. We also assumed that $L \gg L_{\mathrm{a}}$ and the size L_θ of the excited region is determined by the relation $L_\theta = w/\sin\theta_{\mathrm{p}}$, where w is the radius of the interacting beams from (17).

It follows from our calculations, in particular, (Fig. 5) that the broadening of CARS and SRS amplification spectra caused by running out of polaritons from the the region of interaction with incident beams will not exceed 10% only when $L_\theta > 14L_{\mathrm{p}}$. For example, for $w = 0.5$ mm and $\theta_{\mathrm{p}} = 45°$, this condition is satisfied for $L_{\mathrm{p}} < 50$ μm or for $\alpha_{\mathrm{p}} > 200$ cm^{-1}.

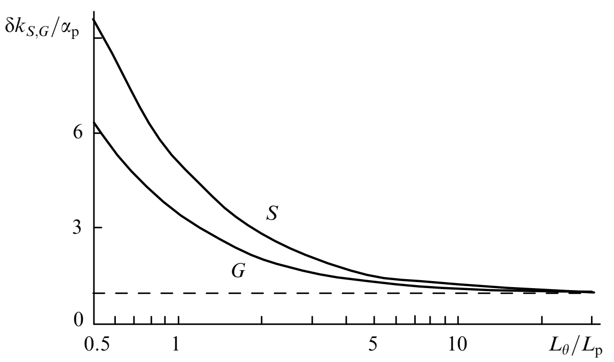

Figure 5. Ratio $\delta k_{S,G}$ of the widths of CARS (S) and SRS amplification (G) spectra to the decay α_{p} of polaritons as a function of the ratio of the size $L_\theta = w/\sin\theta_{\mathrm{p}}$ of the overlap region for exciting beams along the direction of propagation of polaritons to the mean free path L_{p} of polaritons.

Note also that the results of calculations presented in Fig. 5 well agree with the experimental broadening of the ω spectra of CARS from polaritons in a BeO crystal [18]. Indeed, the width $\Delta\nu_{\mathrm{obs}}$ of the CARS ω spectrum at the frequency $\nu_{\mathrm{p}} = 372$ cm^{-1} is ~ 11.5 cm^{-1}. However, according to the direct measurement, the mean free path of coherent polaritons is $L_{\mathrm{p}} = 0.3$ mm [18], which should correspond to the width of the CARS spectrum, neglecting the running out of polaritons,

$$\Delta\nu \approx \frac{1}{2\pi c L_{\mathrm{p}}|V_{\mathrm{p}}^{-1} - V_{\mathrm{s}}^{-1}\cos\theta|} \approx 1.7\ \mathrm{cm}^{-1},$$

where V_{p} and V_{s} are the group velocities of polaritons and the scattered wave, respectively. Therefore, the broadening is $\Delta\nu_{\mathrm{obs}}/\Delta\nu \approx 6.8$. Under the conditions of the experiment described, $L_\theta/L_{\mathrm{p}} \approx 0.65$ and the running out of polaritons should result, according to curve S in Fig. 5, to the sevenfold broadening of the CARS spectrum, in agreement with the experimental broadening equal to 6.8.

4. Conclusions

The analysis performed above showed that the propagation of polaritons affects in the general case both the shape and width of the spectra of SRS amplifications and CARS from polaritons. The shape and width of the spectra depend on the ratio L_θ/L_{p} of the size L_θ of the region of overlap of incident light waves in a crystal along the direction of propagation of polaritons to the mean free path L_{p} of coherent polaritons. (In the case of the transversely infinite beams or for $L_{\mathrm{a}} > L$, their size L_θ is determined by the crystal length.) When polaritons are strongly absorbed ($L_{\mathrm{p}} \ll L_\theta$), they do not 'run out' from the interaction region, and the spectrum is described by a Lorentzian with the width determined by the decay of polaritons. In the case of weal absorption ($L_{\mathrm{p}} \gg L_\theta$), polaritons 'run out' from the interaction region, and the widths of the spectra become much greater than the spatial decay of polaritons and, hence, they contain no information on the decay.

The calculations performed for Gaussian incident beams showed, in particular, that the broadening of the spectra caused by 'running out' of polaritons will not exceed 10 % only when the size of the excited region along the direction of propagation of polaritons is at least 14 times greater than the mean free path of coherent polaritons ($L_\theta > 14L_{\mathrm{p}}$). This is rather stringent condition, which should be taken into account in real measurements of the decay from the widths of SRS amplification and CARS spectra in studies of not only polaritons of the lower and upper dispersion branches, for which the mean free path can exceed several millimetres, but also polaritons related to intermediate dispersion branches. Indeed, for $w = 0.5$ mm and $\theta_{\mathrm{p}} = 45°$, which approximately corresponds to typical experimental conditions, the broadening of the spectra caused by 'running out' of polaritons will not exceed 10 % only when the mean free path of polaritons is less than 50 μm or the decay is $\alpha_{\mathrm{p}} > 200$ cm^{-1}.

The broadening of CARS spectra observed in paper [18] is in good quantitative agreement with the results of calculations presented above.

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