

Nonstationary Stimulated Raman Scattering by Polaritons in Cubic Crystals

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Abstract

The purpose of this article is to consider two aspects of the nonstationary stimulated Raman scattering by polaritons in cubic crystals. The first feature is related to the pump field, which, by deforming the permittivity of the medium, changes its symmetry. As a result, for example, the cubic crystal becomes anisotropic. The second one results from the possibility of exciting anomalous longitudinal waves at the frequency of the mechanical phonons which is the fundamental difference between scattering by dipole-active (polar) phonons and that of by dipole-inactive (nonpolar) ones. When the phonon frequency is approached, the amplitude of the transverse polariton wave decreases due to increased absorption and the wave mismatch. The polariton wave becomes practically longitudinal. Such a wave is maintained by the pump field and exists only in a pumped medium. The system of four shortened nonstationary equations (two for the Stokes waves with perpendicular polarizations and two for both transverse and longitudinal polariton waves) is obtained. The analysis is carried out for a given stationary pump field which is assumed to be a linearly polarized plane electromagnetic wave. Principal attention was paid to the calculation and analysis of the gain factor which defines the intensities of both stimulated (SRS) and spontaneous Raman scattering. The expressions for two proper gain factors g_{μ} are obtained for Stokes waves in nonstationary case. It was shown that the pumped cubic crystal becomes anisotropic. It is also shown that the values of intensities calculated by using the expression for g_{μ} are consistent with the experimental results for spectra of ZnS.

Keywords

Nonstationary Stimulated Raman Scattering, Polaritons, Phonons

1. Introduction

Study of polaritons in different structures has been attracting increasing attention of late [1] [2]. This phenomenon was investigated both theoretically and experimentally, in particular [3] [4] [5] [6]. One of the effective methods of investigations polariton characteristics is SRS [7]. One of the shortcomings of the preceding studies was the assumption that only waves with fixed transverse polarizations interact. In [8] was shown that this assumption was not held for a polariton wave in the vicinity of the phonon frequency. When the polariton frequency approaches that of phonon, the amplitude of transverse part of the polariton wave decreases rapidly and as a result is much lower than the amplitude of the longitudinal polariton wave, which is less sensitive to the absorption and does not depend on the wave mismatch. The investigation of such case was introduced in [8] in which principal attention was paid to a calculation and analysis of the gain of both stationary SRS and spontaneous Raman scattering. In this paper, we considered the case of nonstationary SRS in cubic crystals and showed that the theory developed is consistent with experimental results.

2. Basic Principles and Equations

In this paper, we carry out our analysis in the approximation of a given stationary pump field, which is a linearly polarized plane electromagnetic wave. It is also assumed that the nonlinear medium takes the form of a layer bounded by the planes $z = 0$ and $z = L$. The pump wave

$$\vec{E}_l(\vec{r}, t) = \hat{e}_l A_l \exp\left[i(k_l^z z - \omega_l t)\right] + c.c. \quad (1)$$

propagates along the z -axis. The subscripts l , s , and p henceforth denote the pump (laser), Stokes and polariton wave fields; ω is the frequencies, n and \vec{k} are the refractive indices and the wave vectors in the unpumped medium, and \hat{e} are the real unit vectors of electromagnetic fields. The medium is assumed to be nonmagnetic and transparent at the frequencies $\omega_{l,s}$. We use the Stokes and polariton fields in the form

$$\vec{E}_s(\vec{r}, t) = \sum_{\mu=1,2} \hat{e}_s^{(\mu)} A_s^{(\mu)} \exp\left[i(\vec{k}_s \cdot \vec{r} - \omega_s t)\right] + c.c., \quad (2)$$

$$\vec{E}_p(\vec{r}, t) = \sum_{\sigma=1,2,3} \hat{e}_p^{(\sigma)} A_p^{(\sigma)} \exp\left[i(\vec{W} \cdot \vec{r} - \omega_p t)\right] + c.c., \quad (3)$$

where: $\hat{e}_s^{(\mu)} \perp \vec{k}_s$, $\hat{e}_s^{(1)} \perp \hat{e}_s^{(2)}$, $k_s = q_s n_s$, $q_s = \omega_s / c$, $\vec{W} = \vec{k}_l - \vec{k}_s$, $\hat{e}_p^{(1,2)} \perp \vec{W}$, $\hat{e}_p^{(1)} \perp \hat{e}_p^{(2)}$, $\hat{e}_p^{(3)} = \frac{\vec{W}}{W}$, $\omega_p = \omega_l - \omega_s$.

The longitudinal component of the Stokes wave can obviously be neglected, but this cannot be done for the polariton wave in the phonon region. It has been shown in [8] that with a further advance into this region all three amplitudes $A_p^{(\sigma)}$ first become comparable, after which $A_p^{(3)}$ becomes dominant, provided,

of course, the excitation of the longitudinal waves is allowed by the selection rules. The phase shift of the polariton wave is determined by the vector \vec{W} and not by \vec{k}_p ($k_p = q_p \sqrt{\varepsilon_p}$, $q_p = \omega_p/c$, $\varepsilon_p = \varepsilon'_p + i\varepsilon''_p$ is the dielectric constant at the frequency ω_p).

The fields $\vec{E}_{s,p}$ are interrelated via the nonlinear part of the polarization $\vec{P}(\vec{r}, t)$.

The latter quantity has at the frequencies $\omega_{s,p}$ the following forms

$$P_s = \chi_s^{\mu\sigma} A_l A_p^{(\sigma)*} \exp\left[i\left(k_l^z - k_p^{(\sigma)z}\right)z - \omega_s t\right] + \gamma_s^{\mu\mu'} |A_l|^2 A_s^{(\mu')} \exp\left[i\left(k_s^z z - \omega_s t\right)\right] + c.c. \tag{4}$$

$$P_p = \chi_p^{\mu\sigma} A_l A_s^{(\mu)*} \exp\left[i\left(W^z z - \omega_p t\right)\right] + c.c. \tag{5}$$

where

$$\chi_s^{\mu\sigma} = e_s^{(\mu)i} e_l^j e_p^{(\sigma)k} \chi_{s,ijk}(\omega_l, -\omega_p), \quad \gamma_s^{\mu\mu'} = e_s^{(\mu)i} e_s^{(\mu')j} e_l^k e_l^m \gamma_{s,ijkm}(\omega_p, \omega_l, -\omega_l). \tag{6}$$

The shortened equations for the amplitudes $A_{s,p}^{(\mu,\sigma)}$ are obtained from Maxwell's equations by the standard procedure [9] and take the form

$$\frac{\partial A_s^{(\mu)}}{\partial z} + \frac{1}{v_s^{z(\mu)}} \frac{\partial A_s^{(\mu)}}{\partial t} = i \frac{2\pi\omega_s}{cn_s^{(\mu)} \cos(\theta_s^{z(\mu)})} \left(\chi_s^{\mu\sigma} A_l A_p^{(\sigma)*} + \gamma_s^{\mu\mu'} |A_l|^2 A_s^{(\mu')} \right), \quad \sigma = 1, 2, 3 \tag{7}$$

$$2iW^z \frac{\partial A_p^{(\sigma)*}}{\partial z} - iW e_p^{(\sigma)z} \frac{\partial A_p^{(3)*}}{\partial z} + i \frac{2\omega_p \varepsilon_p^{(\sigma)*}}{c^2} \frac{\partial A_p^{(\sigma)*}}{\partial t} + (W^2 - k_p^{2*}) A_p^{(\sigma)*} = 4\pi q_p^2 \chi_p^{\mu\sigma} A_s^{(\mu)} A_l^* \tag{8}$$

$$-iW \left(e_p^{(1)z} \frac{\partial A_p^{(1)*}}{\partial z} + e_p^{(2)z} \frac{\partial A_p^{(2)*}}{\partial z} \right) + i \frac{2\omega_p \varepsilon_p^{(3)*}}{c^2} \frac{\partial A_p^{(3)*}}{\partial t} - k_p^{2*} A_p^{(3)*} = 4\pi q_p^2 \chi_p^{\mu 3} A_s^{(\mu)} A_l^* \tag{9}$$

Note, that in (8) and (9) $\sigma = 1, 2$.

In view of the strong absorption we have

$$\left| W \left(A_p^{(\sigma)} \right)^{-1} \frac{\partial A_p^{(\sigma)}}{\partial z} \right| \approx \left| \frac{\omega_p}{c^2} \left(A_p^{(\sigma)} \right)^{-1} \frac{\partial A_p^{(\sigma)}}{\partial t} \right| \ll \left| W^2 - k_p^{2*} \right|, \tag{10}$$

and we can, therefore, neglect in (8) and (9) the terms with the derivatives after which these equations yield

$$A_p^{(\sigma)*} = 4\pi \chi_p^{\mu\sigma} A_s^{(\mu)} A_l^* / (s^2 - \varepsilon_p^*),$$

$$A_p^{(3)*} = -4\pi \chi_p^{\mu 3} A_s^{(\mu)} A_l^* / \varepsilon_p^*, \quad s = W/q_p, \quad \sigma = 1, 2. \tag{11}$$

Substituting the obtained expressions in (4) and (5), we arrive at a system of two differential equations with respect to $A_s^{(\mu)}$,

$$\frac{\partial A_s^{(\mu)}}{\partial z} + \frac{1}{v_s^{z(\mu)}} \frac{\partial A_s^{(\mu)}}{\partial t} = i \frac{2\pi\omega_s}{cn_s^{(\mu)} \cos(\theta_s^{z(\mu)})} \left\{ \bar{\gamma}_s^{\mu\sigma} |A_l|^2 A_s^{(\mu)} + \gamma_s^{\mu\mu'} |A_l|^2 A_s^{(\mu')} \right\}, \tag{12}$$

where

$$\bar{\gamma}_s^{\mu\sigma} = 4\pi\chi_s^{\mu\sigma} \left(\frac{\chi_p^{\mu\sigma}}{s^2 - \varepsilon_p^*} - \frac{\chi_p^{\mu 3}}{\varepsilon_p^*} \right), \quad \varepsilon_p = \varepsilon_p' + i\varepsilon_p'', \quad (13)$$

$$\varepsilon_p' = \varepsilon_p^0 + \sum_f s_f v_f^2 (v_f^2 - v_p^2) / \left[(v_f^2 - v_p^2)^2 + \gamma_f^2 v_p^2 \right],$$

s_f is the oscillator strength of the o-f transition.

3. Gain Factor g_μ

Now we show that the system of Equation (12) is consistent with the experimental results presented, for example, in [10]. In order to do that we first bring the system (12) to unitless form and change the variables z, t to variables $\tilde{z}, \tilde{t} = \tilde{t} - \tilde{z}/\tilde{v}_s^z$ (we assume that $\tilde{v}_s^{z(1)} \approx \tilde{v}_s^{z(2)} \approx \tilde{v}_s^z$):

$$\frac{\partial \tilde{A}_s^{(\mu)}}{\partial \tilde{z}} = i\beta_s^{(\mu)} \left\{ \tilde{\gamma}_s^{\mu\sigma} |\tilde{A}_l|^2 \tilde{A}_s^{(\mu)} + \tilde{\gamma}_s^{\mu\mu'} |\tilde{A}_l|^2 \tilde{A}_s^{(\mu')} \right\}, \quad (14)$$

where

$$\tilde{A}_s^{(\mu)} = A_s^{(\mu)} / A_0, \tilde{A}_l = A_l / A_0, \tilde{z} = z / z_0, \tilde{t} = t / \tau_0, z_0 = c\tau_0, \quad (15)$$

$$\beta_s^{(\mu)} = 2\pi\omega_s z_0 / \left(cn_s^{(\mu)} \cos(\theta_s^{(\mu)}) \right),$$

$$\tilde{\gamma}_s^{\mu\sigma} = \bar{\gamma}_s^{\mu\sigma} A_0^2, \tilde{\gamma}_s^{\mu\mu'} = \gamma_s^{\mu\mu'} A_0^2, \tilde{v}_s^{z(\mu)} = v_s^{z(\mu)} / c,$$

τ_0 is the characteristic time related to the laser field (pump).

The theoretical consideration of the gain factor for SRS by polaritons is based on the modeling of the quasi-stationary solutions of the coupled wave equations for the different polarizations of the Stokes. Therefore, we seek the solutions of (8) in the form $\tilde{A}_s^{(\mu)} = B_\mu \exp(\kappa\tilde{z})$, assuming B_μ and κ to be independent of z . We then obtain the system of algebraic equations with respect to B_μ . Choosing in a plane perpendicular to a two-dimensional coordinate system with axes along the unit vectors, we represent the equations for B_μ in the form of a tensor relation

$$\Delta_{\mu\mu'} B_{\mu'} = -i\kappa B_\mu, \quad \mu = 1, 2 \quad (16)$$

where

$$\Delta_{\mu\mu'} = \beta_s^{(\mu)} |\tilde{A}_l|^2 \left(\tilde{\gamma}_s^{\mu\mu'} + \tilde{\gamma}_s^{\mu\sigma} \right). \quad (17)$$

Equating the determinant of the system (9) to zero, we obtain the solutions for κ

$$\kappa = i \left[(\Delta_{11} + \Delta_{22}) \pm \sqrt{(\Delta_{11} - \Delta_{22})^2 + 4\Delta_{12}\Delta_{21}} \right] / 2. \quad (18)$$

We will need the explicit expressions for the tensors χ and Y . They can be found within the framework of the microscopic theory in the dipole approximation based on the perturbation theory states [8]. The resultant expressions are

$$\chi_{ijk}(\omega_l, -\omega_p) = \chi_{ijk}^o(\omega_l, -\omega_p) + \frac{\sqrt{N}}{\hbar} \sum_{fv} \alpha_{ij}^{(fv)} P_{fv}^k F_f(\omega_p) \quad (19)$$

$$\gamma_{ijkm} = \frac{1}{\hbar v_0} \sum_{fv} \alpha_{ik}^{(fv)} \alpha_{jm}^{(fv)} F_f(\omega_p) + \gamma_{ijkm}^\circ, \tag{20}$$

where

$$F_f(\omega) \approx 2\omega_i / (\omega_f^2 - \omega_p^2 + i\tilde{\gamma}\omega_p). \tag{21}$$

The summation in (19) and (20) is over all dipole-active phonons, the frequencies of which are considered to be equal $\omega_f - i\tilde{\gamma}_f/2$, where $\tilde{\gamma}_f$ are the attenuation constants. For example in a cubic crystal, the dipole-active phonons are triply degenerate [8] so that the number the mutually degenerated oscillations we introduce the index ν (\hat{e}_ν is a triad of real unit vectors denoting the vibrations along the edges of the unit cube. Furthermore, $\vec{P}_{fv} = P_f \hat{e}_\nu$ is the dipole moment of the transition 0-fv for the unit cell with its volume v_0 ; $\alpha_{ij}^{(fv)}$ is the tensor of the phonon spontaneous scattering per cell [11]; $N = V/v_0$ is the number of cells in the crystal. The tensor χ_{ijk}° represents the contribution to χ_{ijk} by the remote electronic states. The tensor γ_{ijkm}° determines the contribution due to the electronic states as well. It is convenient to represent the tensors χ_{ijk} (19) and γ_{ijkm} (20) in the simplified form as follows

$$\chi_{ijk} = \chi'_{ijk} + i\chi''_{ijk}, \chi'_{ijk} = \chi_{ijk}^0 + \sum_f \chi_f v_f^2 (v_f^2 - v_p^2) / [(v_f^2 - v_p^2)^2 + \gamma_f^2 v_p^2], \tag{22}$$

$$\chi''_{ijk} = -\sum_f \chi_f v_f^2 \gamma_f v_p / [(v_f^2 - v_p^2)^2 + \gamma_f^2 v_p^2], \tag{23}$$

$$\gamma_{ijk} = \gamma'_{ijk} + i\gamma''_{ijk}, \gamma'_{ijk} = \gamma_{ijk}^0 + \sum_f \gamma_f v_f^2 (v_f^2 - v_p^2) / [(v_f^2 - v_p^2)^2 + \gamma_f^2 v_p^2], \tag{24}$$

$$\gamma''_{ijkm} = -\sum_f \gamma_f v_f^2 \gamma_f v_p / [(v_f^2 - v_p^2)^2 + \gamma_f^2 v_p^2], \tag{25}$$

where

$$\chi_f = (hc/2\pi)^{-1/2} v_i^{-2} (s_f \sigma_f / v_f)^{1/2}, \gamma_f = (8\pi^2 \sigma_f) / (hcv_i^4 v_f), \tag{26}$$

σ_f is the Raman differential cross-section per unit cell v_0 (cm^{-1}/sr).

We introduce the principal axes of the tensor $\Delta_{\mu\mu}$ as a whole. If we denote its principal values as Δ_μ we obtain from (18) $\kappa_\mu = i\Delta_\mu$. Finally, we introduce the gain $g_\mu = 2 \text{Re } \kappa_\mu$ which can be expressed as

$$g_\mu = \frac{8\pi^2 \omega_l \omega_s z_0 I_l}{c^2 n_l n_s \cos(\theta)} \left(4\pi \left[\frac{K'_\mu \epsilon_p'' - K_\mu'' (s^2 - \epsilon'_p)}{(s^2 - \epsilon'_p)^2 + \epsilon_p''^2} + \frac{L'_\mu \epsilon_p'' + L_\mu'' \epsilon_p'}{\epsilon_p'^2 + \epsilon_p''^2} \right] - M_\mu \right), \mu = 1, 2 \tag{27}$$

where

$$K_\mu = \sum_{\sigma=1,2} (\chi^{\mu\sigma})^2 = K'_\mu + iK''_\mu, L_\mu = (\chi^{\mu 3})^2 = L'_\mu + iL''_\mu, I_l = cn_l |A_l|^2 / 2\pi \tag{28}$$

is the pump intensity, M_μ are the principal values of the tensor $\gamma_s^{(\mu\mu)'}$, θ is the scattering angle (the angle between \vec{k}_l and \vec{k}_s ($n_s^{(\mu)} \approx n_s, \cos(\theta_s^{(\mu)}) = \cos(\theta)$). Formula (27) denotes two gain coefficients for Stokes waves polarized along $\hat{e}_s^{(\mu)}$. To verify (27), we were using the parameters of crystals widely used in

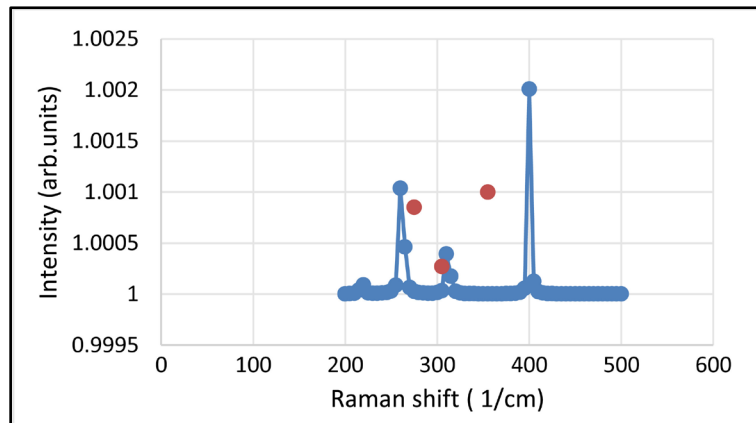


Figure 1. Gain factor versus polariton frequency in zinc blende ZnS. The red dots correspond to the experimental points ([10]); blue solid lines are the result of a calculation based on (27).

optical display and storage, optical communication network, optical detection, etc. such as ZnO [12]-[19] and ZnS [10] [20] [21] [22] [23]. In calculations for the gain, we used the following: pulse width of the pulsed Ar⁺ laser $\approx 5 \mu\text{s}$, the peak output power $\approx 150 \text{ mW}$, the average output power $\approx 7.5 \mu\text{W}$, the wavelength was $514.5 \mu\text{m}$ [10], the cross-section $\approx 10^{-18} \text{ cm}^{-2}$, $\gamma_f \approx 10 \text{ cm}^{-1}$, the polarizability $\approx 10^{-3}$ [18], and $\chi \approx 10^{-8} \text{ esu}$. In **Figure 1**, it is shown the intensity as a function of the polariton frequency in zinc blende ZnS in the range 200 - 400 cm^{-1} . The red dots represent the experimental points [10].

4. Conclusion

In this paper, we showed that the expression (27) for the gain factor of Stokes radiation in cubic crystals based on taking into account the contributions of both transverse and longitudinal polariton waves in the vicinity of the phonon resonance is consistent with the experimental results (the SRS spectra of ZnS).

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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