Nonlinear amplification of the brillouin-rayleigh triplet caused by two-photon heating

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Abstract: The thin structures of stimulated Brillouin scattering (SBS) and stimulated temperature scattering (STS) spectral components caused by two-photon heating are analyzed theoretically. In contrast to the linear (single-photon) case for two-photon heating a stokes SBS component exhibits the spectral shift depending on the pump intensity. Emergence of an anti-stokes SBS component is possible when the pump intensity is sufficiently high so that the positive two-photon thermal gain may compensate the negative electrostrictive gain. The spectral components of STS caused by linear or two-photon absorption (essentially different linear or two-photon STS-2) possess the same thin structures.

Keywords: Nonlinear Optics; Stimulated Brillouin Scattering (SBS); Stimulated Temperature Scattering (STS); Brillouin-Rayleigh Triplet; Two-Photon Heating; Stokes and Anti-Stokes Components; Near Ultraviolet Radiation; Excimer Lasers

1. Introduction

For high enough light intensity and coherence the well-known weak spontaneous Brillouin-Rayleigh triplet [1, 2] transforms into the powerful doublet of a “slightly” anti-stokes shifted line of stimulated temperature scattering (STS) caused by linear (single-photon) or two-photon absorption (linear or two-photon STS-2) and a “strongly” stokes shifted line of stimulated Brillouin scattering (SBS). Various experiments display the doublet’s lines singly or grouped. For the near-ir spectral region (the pump wavelength is \( \lambda_0 = 0.69 \div 1.06 \mu m \)) such a transformation has been originally observed in [3, 4]. For the near-uv spectral region (\( \lambda_0 = 193 \div 351 \text{nm} \)) such a transformation has been originally observed in [5, Fig. 2]. Indeed, in the previous near-uv studies [6 - 14] two-photon STS-2 lines have been associated mistakenly with SBS and linear STS-2 lines [5].

SBS is the unique high-efficiency converter of a coherent light wave (hereinafter called the pump wave that carries the pump intensity \( I_p \)) into a coherent hyperacoustical wave. Also SBS is the nonlinear-optical phenomenon providing phase conjugation (PC) of the best quality [15, 16], [5, Fig. 3]. There are two physical mechanisms responsible for nonlinear amplification of the scattered and hyperacoustical waves during an SBS process [1, 2, 4, 17, 18]. The first one (hereinafter called the conventional SBS) is due to a local variation of pressure caused by the electrostrictive force [19 - 22]. The second one (hereinafter called the thermal SBS) is attributed to a local variation of pressure caused by the thermal expansion. For the linear light absorption the thermal SBS (hereinafter called the linear thermal SBS) has been discussed in [23 - 25].

The purely conventional SBS was considered in [5], the thermal SBS was ignored. This is a quite typical approximation, used for instance in the study of the PC provided by SBS [15], when information gained from roughly measured spectral shifts is enough. It should be noted that too rough measurements can lead to a loss of new physics. Such a loss of the genuine SBS for the near-uv, the two-photon STS-2, and other effects [5] has happened in [6 - 14].

Following [5], the unshifted lines in the left sides of [5, Fig. 2] (relative to the pump ones in the right sides) correspond to the linear and two-photon STS-2; the shifted lines correspond to the genuine conventional SBS. The observation of the thin structures of these lines including the pump ones is restricted by a Fabry-Perot etalon based spectrum analyzer [5]. The spectral resolution of a Fabry-Perot etalon is limited by several MHz (or \( 10^{-3} \text{cm}^{-1} \)) [26]. To reach the higher spectral resolution methods of
heterodyning and intensity fluctuations correlation should be used. An experimental high-resolution spectral profile of a Brillouin line exhibiting an antisymmetrical behavior is given in [27].

In this paper, a contribution of the two-photon heating to the thermal SBS (hereinafter called the two-photon thermal SBS) modifying the stokes and anti-stokes branches is considered. The thin structure of a two-photon STS-2 line first experimentally discovered in [5] was not studied theoretically and is also a subject of interest.

An effective linear absorption coefficient \( \alpha_{\omega} \) (\( \omega \) ) has been introduced [24] for gases to describe the thermalization processes of the absorbed electromagnetic energy. In our analysis a total absorption coefficient [5]

\[
\alpha_{\gamma} = \alpha + (I, \gamma),
\]

should be used to describe the two-photon effect (\( \alpha \) is a linear absorption coefficient and \( \gamma \) is a two-photon absorption coefficient).

2. Mass (Bulk) and Surface Forces

The theory of the coupling of light and elastic waves is based on the Lagrange equation [19, 20]. In [19] a nonlinear system was developed and a linearized system for small perturbations has been solved. The photoelastic coupling of a longitudinal acoustic wave in an isotropic medium was studied in [20], and the nonlinear system from [19] including the saturation effect has been solved.

The Lagrange equation [28] describes the mechanics of discrete bodies. It takes into account the mass (bulk) forces and ignores the surface ones. For continuous media both mass and surface forces should be incorporated [29, 30] and ignores the surface ones. For continuous media both mass and surface forces should be incorporated [29, 30] and for the thin structure of a two-photon STS-2 line the thermal SBS (hereinafter called the two-photon thermal SBS) modifying the stokes and anti-stokes branches is considered. The thin structure of a two-photon STS-2 line was first experimentally discovered in [5] was not studied theoretically and is also a subject of interest.

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An ideal dielectric medium with uniform optical properties cannot scatter light. Both spontaneous (SP) and stimulated (NL) manners of the scattering arise from permittivity variation about the equilibrium value \( \varepsilon_{0} \):

\[
\Delta \varepsilon = \Delta^{SP} \varepsilon + \Delta^{NL} \varepsilon = \left( \frac{\partial \varepsilon}{\partial \rho} \right) T \Delta \rho + \left( \frac{\partial \varepsilon}{\partial T} \right) \rho \Delta T = \left( \frac{\partial \varepsilon}{\partial \rho} \right) T \Delta \rho + \left( \frac{\partial \varepsilon}{\partial T} \right) \rho \Delta T + \left( \frac{\partial \varepsilon}{\partial \rho} \right) \Delta \rho \Delta T + \left( \frac{\partial \varepsilon}{\partial T} \right) \rho \Delta T + \left( \frac{\partial \varepsilon}{\partial \rho} \right) \Delta \rho \Delta T + \left( \frac{\partial \varepsilon}{\partial T} \right) \rho \Delta T.
\]
Usually \([1, 2, 17, 32]\)

\[
\left( \frac{\partial \hat{e}}{\partial \rho} \right)_T \Delta^{NL} \rho = \left( \frac{\partial \hat{e}}{\partial T} \right)_T \Delta^{NL} T,
\]

\[
\left( \frac{\partial \hat{e}}{\partial T} \right)_T = -\beta \left( \frac{\partial \hat{e}}{\partial \rho} \right)_T.
\]

(6)

The task has been provided by the material equations (4) and (5) involving independent variables \(\Delta \rho\) and \(\Delta T\), and as a new element a total absorption coefficient \(\alpha_z\).

### 4. SBS and STS Gain

Physically speaking SBS and STS are nonresonant parametric phenomena \([22, 37]\). (Simulated Raman scattering is a resonant parametric phenomenon.)

Consider two counterpropagating linearly polarized plane electromagnetic waves, a pump wave and a backscattered wave, characterized by electric field vectors \(E_p\) and \(E_s\):

\[
E_p = \frac{1}{2} e \left\{ E_1(z,t) \exp \left( ik_1 z - i \omega_1 t \right) + c.c. \right\}, \quad (7)
\]

\[
E_s = \frac{1}{2} e \left\{ E_2(z,t) \exp \left( -ik_2 z - i \omega_2 t \right) + c.c. \right\}. \quad (8)
\]

Here, \(e\) is a common \([21]\) unit vector (for definiteness sake \(e = e_X\)); \(E_1(z,t)\) and \(E_2(z,t)\) are complex amplitudes; \(\omega_1\), \(\omega_2\) and \(k_1, k_2\) are temporal frequencies and wave numbers, respectively. A total electric field vector is

\[
E = E_p + E_s. \quad (9)
\]

The linear (L) and nonlinear (NL) electrical induction vectors are \([21, 22, 37]\)

\[
D^L(z,t) = \hat{\varepsilon}(\omega) E(z,t),
\]

\[
D^{NL}(z,t) = \hat{\varepsilon}^{NL}(\omega, z, t) E(z,t),
\]

where \(\hat{\varepsilon}(\omega)\) and \(\hat{\varepsilon}^{NL}(\omega, z, t)\) are the Fourier transforms of the linear and nonlinear permittivity tensors \(\hat{\varepsilon}(t)\) and \(\hat{\varepsilon}^{NL}(t_1, t_2, t_3)\) \([22, 37]\). For the isotropic medium the tensors are replaced by the scalars \([22, 37]\):

\[
D^L(z,t) = \varepsilon(\omega) E(z,t),
\]

\[
\Delta^{NL}(z,t) = \varepsilon^{NL}(\omega, z, t) E(z,t),
\]

\[
D^{NL}(z,t) = \varepsilon^{NL}(\omega, z, t) E(z,t),
\]

\[
D^{NL}(z,t) = \varepsilon^{NL}(\omega, z, t) E(z,t) \equiv
\]

\[
\left( \frac{\partial \varepsilon}{\partial \rho} \right)_T \Delta^{NL} \rho(z,t) + \left( \frac{\partial \varepsilon}{\partial T} \right)_T \Delta^{NL} T(z,t) \right) E(z,t) \equiv
\]

\[
\left( \frac{\partial \varepsilon}{\partial \rho} \right)_T \Delta^{NL} \rho(z,t) E(z,t) \right) .
\]

Accordingly, the linear and nonlinear polarization vectors are \([21]\)

\[
P^{L}(z,t) = \frac{\varepsilon(\omega) - 1}{4\pi} E(z,t),
\]

\[
P^{NL}(z,t) = \frac{1}{4\pi} \varepsilon^{NL}(\omega, z, t) E(z,t)
\]

\[
\Delta^{NL}(z,t) \equiv \frac{1}{4\pi} \left( \frac{\partial \varepsilon}{\partial \rho} \right)_T \Delta^{NL} \rho(z,t) E(z,t) . (10)
\]

The waves are coupled by the scalar electrodynamical equations \([1, 2, 17, 32]\)

\[
\begin{align*}
\nabla^2 - \frac{\varepsilon(\omega_1)}{c^2} \frac{\partial^2}{\partial t^2} E_1 & = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} P^{NL}_{\omega_1}, \\
\nabla^2 - \frac{\varepsilon(\omega_2)}{c^2} \frac{\partial^2}{\partial t^2} E_2 & = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} P^{NL}_{\omega_2}.
\end{align*}
\]

(11)

(12)

with the right-hand sides representing the nonlinear polarizations oscillating with the frequencies \(\omega_1\) and \(\omega_2\). For the plane waves diffraction is absent (\(\left[ \frac{\partial \varepsilon}{\partial \rho} \frac{\partial \varepsilon}{\partial \rho} \right]_{E_1(z,t) = 0} \)).

Following (4) and (5) the nonlinearity of (10) is due to the dependence of \(\Delta \rho \equiv \Delta^{NL} \rho\) and \(\Delta T \equiv \Delta^{NL} T\) on the scalar product \(E^2\). We seek the steady-state solution (the complex amplitudes do not depend on \(t\)) based on the slowly oscillating part of \(E^2\)

\[
\left\langle E^2 \right\rangle = \left\langle (E_p + E_s)^2 \right\rangle = \left\langle 2E_p E_s \right\rangle = \frac{1}{2} \left\{ E_1(z) E_1^*(z) \exp \left( -i(\omega_1 - \omega_2) t + i(k_1 + k_2) z \right) + c.c. \right\},
\]

(13)

and the appropriate approximations for \(\Delta^{NL} \rho\) and \(\Delta^{NL} T\)

\[
\Delta^{NL} \rho(z,t) = \frac{1}{2} \left\{ \rho_1(z) \exp \left( -i(\omega_1 - \omega_2) t + i(k_1 + k_2) z \right) + c.c. \right\}. \quad (14)
\]

\[
\Delta^{NL} T(z,t) = \frac{1}{2} \left\{ T_1(z) \exp \left( -i(\omega_1 - \omega_2) t + i(k_1 + k_2) z \right) + c.c. \right\}. \quad (15)
\]
On substitution of (6), (13) - (15) into (4) and (5), a linear system for the complex amplitudes $\rho_s(z)$, $T_s(z)$ and for the product $E_{1}(z)E_{2}\ast(z)$ is found

$$
\begin{align*}
&\left(-\Omega^2 + \frac{v^2}{\delta}q^2 + i \frac{\eta}{\rho_0}q^2\Omega\right)\rho_s + \\
&\frac{v^2}{\delta}\beta \rho_s - v^2 T_s = \frac{1}{8\pi}\left(\frac{\partial\varepsilon}{\partial \rho}\right)_{T_q} q^2 E_1E_2^\ast,
\end{align*}
$$

(16)

where $q = q_1 + q_2$, $\Omega = \omega_1 - \omega_2$. Solving (16) for $T_s$

$$
T_s = \frac{\delta}{q^2 v^2 \beta \rho_0} \left[\frac{1}{8\pi}\left(\frac{\partial\varepsilon}{\partial \rho}\right)_{T_q} q^2 E_1E_2^\ast - \left(-\Omega^2 + \frac{v^2}{\delta}q^2 + i \frac{\eta}{\rho_0}q^2\Omega\right)\rho_s\right],
$$

and inserting the result into (17) we obtain

$$
\begin{align*}
&i\Omega \frac{c_v}{\beta} (\delta - 1) \rho_s + \\
&\frac{\delta}{q^2 v^2 \beta \rho_0} \left[\frac{1}{8\pi}\left(\frac{\partial\varepsilon}{\partial \rho}\right)_{T_q} q^2 E_1E_2^\ast - \left(-\Omega^2 + \frac{v^2}{\delta}q^2 + i \frac{\eta}{\rho_0}q^2\Omega\right)\rho_s\right] = \\
&= \frac{1}{4\pi} \left[nc\alpha_s - \frac{i}{2} \frac{\partial\varepsilon}{\partial \rho}\right]_{T_q} E_1E_2^\ast,
\end{align*}
$$

(17)

Rearrangement of $\rho_s$ and $E_{1}E_{2}\ast$ into the opposite sides gives

$$
\begin{bmatrix}
& i\Omega \frac{c_v}{\beta} (\delta - 1) - \\
& \frac{\delta}{q^2 v^2 \beta \rho_0} \left[\frac{1}{8\pi}\left(\frac{\partial\varepsilon}{\partial \rho}\right)_{T_q} q^2 E_1E_2^\ast - \left(-\Omega^2 + \frac{v^2}{\delta}q^2 + i \frac{\eta}{\rho_0}q^2\Omega\right)\rho_s\right] = \\
& = E_{1}E_{2}\ast
\end{bmatrix}
$$

Upon multiplying the both sides by $\left[-\frac{8\pi v^2}{\delta} \rho_s\right]$ we have

$$
\begin{align*}
&-\Omega^2 + \frac{v^2}{\delta}q^2 + i \frac{\eta}{\rho_0}q^2\Omega = \\
&= E_{1}E_{2}\ast
\end{align*}
$$

(18)

By the use of the expressions $\beta_s(\delta - 1) = \beta \rho \rho_s$ and $v^2 = \frac{1}{\rho_0 \beta s}$ the other form of the last term in (18) is achieved

$$
\begin{align*}
&= \frac{i}{\delta} v^2 \rho_s \beta^2 T_{s0} \Omega = \\
&= E_{1}E_{2}\ast
\end{align*}
$$

(19)

From the standpoint of a cubic nonlinear susceptibility tensor, projections of the nonlinear polarization vectors appearing in (11), (12) into the Cartesian coordinates are

$$
\begin{align*}
&= \chi^{(3)}(\omega_1, \omega_2, \Omega)(E(z, t))_x (E(z, t))_y (E(z, t))_z
\end{align*}
$$

For our case defined by (7) - (9):

$$
\begin{align*}
&\left[P_{\omega_1, \omega_2}(z, t)\right]_x = \chi^{(3)}(\omega_1, \omega_2, \Omega)(E(z, t))_x (E(z, t))_y (E(z, t))_z
\end{align*}
$$

(20)

For the isotropic branch $\chi^{(3)}$ is a scalar:

$$
\begin{align*}
&\left[P_{\omega_1, \omega_2}(z, t)\right]_x = \chi^{(3)}(E(z, t))_x (E(z, t))_y (E(z, t))_z
\end{align*}
$$

(20)
On substitution of $\Delta^{NL}_\rho$ from (14) into (10) in accordance with (18), (19), (20) we derive [2]:

$$\chi^{(3)} = \frac{1}{4\pi D} \rho_0 \frac{\partial \varepsilon}{\partial \rho} \frac{1}{16\pi \rho_0} \times$$

$$-2\beta n c_\varepsilon \rho_0 \frac{\partial \varepsilon}{\partial \rho} \delta^2 + \left( \rho_0 \frac{\partial \varepsilon}{\partial \rho} \right) \left[ \frac{\lambda_1 q^2}{i \rho_0 c_\varepsilon \Omega} \right]$$

$$+ i \left( 1 - \frac{1}{\delta} \right) \rho_0 c_\varepsilon \Omega \left[ \frac{\lambda_1 q^2}{i \rho_0 c_\varepsilon \Omega} \right]$$

$$\times \left[ \frac{\Omega^2 + \frac{\Omega^2}{\delta}}{\Omega + \frac{i \Omega}{\delta}} \right]$$

$$\left[ \frac{i \frac{\eta q^2}{\rho_0} \Omega}{\rho_0} + i \left( 1 - \frac{1}{\delta} \right) \rho_0 c_\varepsilon \Omega \delta^2 \right].$$

(21)

In our case $D = 3$ [2]. The cubic nonlinear susceptibility (21) exhibits Rayleigh (labeled with R) resonance at $|q|=0$ and Brillouin (labeled with B) resonance at $|q| = \Omega_n = q v = (k + k_v) v$.

Rayleigh resonances. For $\Omega \approx 0, |q| << \Omega_n$ (21) incorporates electrostatic (labeled with R1) and absorptive (labeled with R2) terms:

$$\chi^{(3)R} \approx \chi^{(3)R1} + \chi^{(3)R2}$$

where

$$\chi^{(3)R1} = -\frac{1}{32\pi D} \rho_0 \frac{\partial \varepsilon}{\partial \rho} \beta_s (\delta - 1) \left[ \frac{2 - \delta}{2(\delta - 1)} + \frac{2}{\Omega + i \Omega_n} \right]$$

$$\chi^{(3)R2} = \frac{1}{64\pi^2 D} \rho_0 \frac{\partial \varepsilon}{\partial \rho} \alpha c_n \beta \frac{\Omega_n}{\Gamma_n}$$

$$\Gamma_n = \frac{\lambda_1 q^2}{\rho_0 c_\varepsilon \rho_0}$$

(22)

(23)

The imaginary parts of (22) and (23) are (as to the origin of $\beta^E_n$ and $\beta^R_n$ see (34) below)

$$\text{Im}\chi^{(3)R1} = -\frac{1}{32\pi D} \rho_0 \frac{\partial \varepsilon}{\partial \rho} \beta_s (\delta - 1) \frac{\Omega_n}{\Gamma_n}$$

$$\text{Im}\chi^{(3)R2} = \frac{1}{64\pi^2 D} \rho_0 \frac{\partial \varepsilon}{\partial \rho} \alpha c_n \beta \frac{\Omega_n}{\Gamma_n}$$

$$\frac{1}{\Gamma_n}$$

(24)

(25)

Brillouin resonances. For $\Omega \approx \pm \Omega_n$ (21) incorporates electrostatic (or conventional labeled with B1) and absorptive (or thermal labeled with B2) terms:

$$\chi^{(3)B} = \chi^{(3)B1} + \chi^{(3)B2}$$

where

$$\chi^{(3)B1} = \frac{1}{64\pi^2 D} \rho_0 \frac{\partial \varepsilon}{\partial \rho} \beta_s (2 - \delta) \frac{\rho_0 c_\varepsilon \Omega}{\eta q |\Omega - \Omega_n| \pm i \Omega_n/2}$$

$$\chi^{(3)B2} = \pm \frac{i}{32\pi^2 D} \rho_0 \frac{\partial \varepsilon}{\partial \rho} \alpha c_n \beta \frac{\Omega_n}{\Gamma_n}$$

$$|\Omega - \Omega_n| \pm i \Omega_n/2$$

$$\Gamma_n = \frac{\eta q^2}{\rho_0}$$

(26)

(27)

The bottom signs in (26), (27), (28), and (29) correspond to the stokes ($\Omega_1 > \Omega_2, \Omega > 0, |q| = \Omega$) and the top signs - to the anti-stokes ($\Omega_1 < \Omega_2, \Omega < 0, |q| = -\Omega$) spectral regions, respectively. The imaginary parts of (26) and (27) are (as to the origin of $\beta^E_n$ and $\beta^R_n$ see (34) below)

$$\text{Im}\chi^{(3)B1} = \pm \frac{1}{64\pi^2 D} \rho_0 \frac{\partial \varepsilon}{\partial \rho} \beta_s (2 - \delta) \frac{\rho_0 c_\varepsilon \Omega}{\eta q |\Omega - \Omega_n| \pm i \Omega_n/2}$$

$$\pm \frac{1}{64\pi^2 D} \rho_0 \frac{\partial \varepsilon}{\partial \rho} \alpha c_n \beta \frac{\Omega_n}{\Gamma_n}$$

$$|\Omega - \Omega_n| \pm i \Omega_n/2$$

$$\Gamma_n$$

(28)

(29)

A couple of equal in magnitude to
peaks of the electrostrictive (conventional) Brillouin term defined by (28) demonstrate positive $G \propto -\text{Im} \chi^{(3)}$ for \( \Omega \approx \Omega_B \), and negative $G \propto -\text{Im} \chi^{(3)}$ for \( \Omega \approx -\Omega_B \) (Fig. 1a).

Four equal in magnitude to

\[
\text{Im} \chi^{(3)B}_{\text{MAX}} = \frac{1}{64 \pi^2 D} \left( \frac{\partial \varepsilon}{\partial \rho} \right)^2 \frac{\left( \frac{\partial \varepsilon}{\partial \rho} \right)^2}{\beta_4 (2 - \delta) \Omega_B} \frac{\beta_4 (2 - \delta) \Omega_B}{64 \pi^2 D \Gamma_n} (30)
\]

be found in [2, 18]. A couple of the SBS curves in the vicinity of the stokes resonance can be found in [4, 17]. Ex- ventional SBS. The spectral profile (Fig. 1a) possesses positive stokes and negative anti-stokes values. The widths (FWHM) of resonance peaks of the electrostrictive (conventional) Brillouin term defined by (28) demonstrate positive $G \propto -\text{Im} \chi^{(3)}$ for \( \Omega \approx \Omega_B \) and for \( \Omega \approx -\Omega_B \) (Fig. 1b). The ratio of (31) and (30) provides the relative contribution of the absorptive (thermal) and electrostrictive (conventional) mechanisms into SBS gain (see Section 7):

\[
\frac{\text{Im} \chi^{(3)B}_{\text{MAX}}}{\text{Im} \chi^{(3)E}_{\text{MAX}}} = \frac{64 \pi^2 D c_p \rho_0}{\beta_4 (2 - \delta) \Omega_B} \frac{\beta_4 (2 - \delta) \Omega_B}{64 \pi^2 D \Gamma_n} \frac{\left( \frac{\partial \varepsilon}{\partial \rho} \right)^2}{\left( \frac{\partial \varepsilon}{\partial \rho} \right)^2 + (2 - \delta) \Omega_B (32)}
\]

\[
\Omega = \omega_i - \omega_2 - \Delta \Omega = \Omega_B - \Delta \Omega = (k_1 + k_2) \omega\nu, \quad \gamma_B = \frac{\eta (k_1 + k_2)^2}{\rho_0 \lambda_1}, \quad \Gamma_B = \frac{\lambda_1 (k_1 + k_2)^2}{\rho_0 C_p}. (35)
\]

In the first two expressions of (34) signs "+" and "-" correspond to the stokes (\( \omega_i > \omega_2, \Omega > 0 \)) and anti-stokes (\( \omega_i < \omega_2, \Omega > 0 \)) spectral regions, respectively.

5. SBS and ST Spectral Components for Linear Absorption

Fig. 1 shows the spectral profiles of $G(\Omega)$ for the SBS and STS mechanisms. For linear absorption such curves can be found in [2, 18]. A couple of the SBS curves in the vicinity of the stokes resonance can be found in [4, 17]. Excitation of an anti-stokes SBS component was not considered there.

The term proportional to $\beta'_B$ in (34) represents the con- ventional SBS. The spectral profile (Fig. 1a) possesses positive stokes and negative anti-stokes values. The widths (FWHM) of resonance peaks are approximately equal to $\Gamma_B$.

The term proportional to $\beta'_B \propto \alpha_k = \alpha$ (see (29)) represents the linear thermal SBS. The spectral profile (Fig. 1b) possesses positive and negative values in both stokes and anti-stokes regions. The widths (FWHM) of resonance
peaks are approximately equal to $\frac{\Gamma_B}{2}$.

The term proportional to $\beta^c_B \propto \alpha$ (see (25)) represents the STS due to linear absorption (the linear STS-2). The spectral profile (Fig. 1b) possesses positive anti-stokes and negative stokes values. The widths (FWHM) of resonance peaks are approximately equal to $\frac{\Gamma_B}{R}$.

The term proportional to $\beta^c_B \propto \alpha \beta = \alpha \sum a \Gamma (\text{see (25)})$ represents the STS due to an electrocaloric effect (the STS-1). The spectral profile (Fig. 1a) is mirror symmetric to that of the linear STS-2.

Figure 1. The gain parameter $G(\Omega)$ defined by (34) for the SBS and STS mechanisms: (a) conventional SBS and STS-1; (b) thermal SBS and STS-2.

6. SBS and STS Spectral Components for Two-Photon Absorption

Single-photon and two-photon transitions provide complementary spectroscopic data [17]. In analysis [5] linear (single-photon) absorption switches to two-photon one by replacing $\alpha$ with $\gamma \rho$. In particular, $\beta^e_B \propto \gamma \rho$.

Being essentially different [5], the linear STS-2 and two-photon STS-2 are characterized by the common gain curve (Fig. 1b). The shifts and widths of its resonance peaks are approximately equal to $\frac{\Gamma_B}{R}$, which is close to the spectral resolution of the typical experimental setup [5]. In this respect, the linear and two-photon STS-2 spectral components are experimentally indistinguishable not only from one another, but also from the STS-1 one, characterized by the mirror symmetric gain curve (Fig. 1a).

SBS contains the experimentally distinguishable [17] conventional and thermal components ($\Omega_B << \Gamma_B >> \Gamma_R = \text{spectral resolution}$). In the stokes region the conventional SBS must be shifted by the thermal SBS. For linear absorption such a shift depends on $\alpha$. For two-photon absorption such a shift depends on $\gamma \rho$ (see Section 7). In the anti-stokes region the positive two-photon thermal values, being proportional to $\beta^c_B \propto I_p$, are added to the negative conventional values, being proportional to $\beta^c_B = \text{const}$, and the positive overall SBS gain can be achieved when $I_p$ is sufficiently high.

7. Overall Stokes SBS Gain

Denoting the stokes SBS part of (34) by $\beta^c_B$ and dividing it by $\beta^c_B$, we obtain

$$\left(\frac{\beta^c_B}{\beta^c_B}\right) = \frac{1}{1 + \left(\frac{2\Delta \Omega}{\Gamma_B}\right)^2}$$

Setting

$$\left(\frac{\beta^c_B}{\beta^c_B}\right) \equiv Z, \quad \left(\frac{\beta^e_B}{\beta^c_B}\right) \equiv Y, \quad \left(2\Delta \Omega / \Gamma_B\right) \equiv X,$$

we rewrite (36) as

$$Z(X,Y) = \frac{1}{1 + X^2} + YX \frac{1}{1 + X^2}.$$  \hspace{1cm} (37)

The parameters $\beta^c_B$, $\beta^e_B$, and $\Gamma_B$ are independent of $\Delta \Omega$. A variable $X$ in (37) describes the frequency shift $\Delta \Omega$. When $X = 1$, the shift is $\Delta \Omega = \frac{\Gamma_B}{2}$. A variable $Y \geq 0$ expresses a relative contribution of the thermal and conventional SBS. $Y < 1$ is the realistic case of strong conventional and weak thermal mechanisms; and $Y > 1$ is the unrealistic case because of the self-action and phase mismatch due to heating [5]. For linear absorption [2, 4] (see (32))

$$Y = \frac{\beta^e_B}{\beta^c_B} = \frac{\delta - 1}{\alpha \lambda} \frac{c}{v} \sin(\theta/2)$$

where $\theta = \pi$ for the backscattering. Thus,

$$Y = (\text{const}) \times \alpha.$$ \hspace{1cm} (39)

For two-photon absorption $\alpha$ is substituted by $\gamma \rho$ in (38), and

$$Y = (\text{const}) \times (\gamma \rho).$$ \hspace{1cm} (40)

According to (39) and (40), when the material properties are held constant ($\alpha, \gamma = \text{const}$), a change in $I_p$ can cause a change in $Y$ for the two-photon thermal SBS, only.
A function $Z(X,Y)$ can be treated as a dependence $\text{const} \times \beta_B(X)$ at different values of $Y$. Fig. 2 shows a three-dimensional plot of $Z = Z(X,Y)$ for $Y$ varying from 0 to 1.5. The intersection of $Z(X,Y)$ with the plane $Y = 0$ is the even function of $X$ corresponding to the conventional SBS. The contour plot in Fig. 3 demonstrates the shift more clearly.

The experimentally observed stimulated scattering spectral components are 5-times narrower [15] than the gain curves in Figs. 2 and 3. Hence, in an experiment the extra shift must be more pronounced.

Figure 2. A three-dimensional plot of the overall stokes SBS gain parameter for the conventional and thermal mechanisms.

8. Numerical Estimates for Spectral Shift

According to the relaxation theory developed by Mandelstam and Leontovich, attenuation of a hypersonic wave in a liquid is dominated by shear viscosity [21, 34, 35, 38]. Therefore,

$$\eta = \frac{2\eta_t}{3} , \quad k_1 + k_2 = 2k_1 = \frac{4m}{\lambda_1} ,$$

where $\eta_t$ is a shear viscosity coefficient. From (35) we obtain

$$\Gamma_B = \frac{2\eta_t}{3\rho_0} \left( \frac{4\pi n^2 \eta_t}{\lambda_1} \right)^2 \approx \frac{32}{3} \pi^3 n^3 \eta_t \lambda_1 . \quad (41)$$

Following [5], we perform estimates for liquid hexane (C6H14), $\lambda_i = 308 \text{ nm}$, and the material parameters [1, 21, 38, 39]:

$$\lambda_i = 3 \times 10^{-1} \text{ cm} , \quad \theta = \pi , \quad \rho_0 = 0.66 \times 10^{-1} \text{ g cm}^{-3} , \quad n = 1.4 ,$$

$$\eta_t = 3.2 \times 10^{-3} P = 3.2 \times 10^{-1} \times 6 \times 10^{-3} \text{ cm s}^{-1} ,$$

$$\beta = 1.4 \times 10^{-1} K^{-1} , \quad T_0 = 300 K ,$$

$$\nu = 10^{-1} \text{ cm s}^{-1} , \quad c = 3 \times 10^{10} \text{ cm s}^{-1} , \quad \rho_0 \left( \frac{\partial \epsilon}{\partial \rho} \right) = 1 , \quad \delta = \frac{C_v}{C_T} = 1.3 .$$

Then (41) yields

$$\Gamma_B = 1.1 \times 10^9 \text{ Hz} , \quad 0.03 \text{ cm}^{-1} .$$

Equation (38) gives

$$Y = \frac{\beta_B^a}{\beta_B^c} \approx 0.73 \times \alpha , \quad (42)$$

where $\alpha$ is measured in $\text{ cm}^{-1}$. $Y = 1$ corresponds to $\alpha = 1.37 \text{ cm}^{-1}$.

For two-photon absorption $\alpha$ is substituted by $\mathcal{P}_p$ in (42), and

$$Y = 0.73 \times (\mathcal{P}_p) . \quad (43)$$

Table 1 from [5] lists the two-photon contribution $\mathcal{P}_p$ obtained for the three experimental values of $I_p$. The maximum value is $Y_{p\text{max}} = 1 \text{ cm}^{-1}$ and

$$Y_{p\text{max}} = 0.73 \times (\mathcal{P}_p)_{\text{max}} = 0.73 .$$

Due to losses and saturation the stimulated scattering is generated near the top of the curve in Fig. 2. Hence, from Fig. 3 $Y = 0.73$ corresponds to $X = 0.5$, and the frequency shift is

$$\Delta \Omega = \frac{\Gamma_B}{4} \approx 0.007 \text{ cm}^{-1} .$$

It should be noted that the SBS component was suppressed at $I_{p\text{max}}$ because of the phase mismatch associated.
with the two-photon heating [5]. For \( I_p = 10^{9} W / cm^2 \) we have \( \gamma(\lambda) = 0.1 \ cm^{-1} \) (see Table 1) and \( Y \approx 0.07 \) (see (43)). The appropriate shift (see Fig. 3) is too small, to be detected under the experimental conditions of [5]. This is not surprising because the analysis presented in [5] was focused on other issues.

**Table 1.** Two-photon contribution \( \gamma(\lambda) \) to the total absorption coefficient at \( \lambda = 308 \text{ nm} \) in hexane for the three values of the pump intensity \( I_p \)

| \( I_p, \ W / cm^2 \) | \( \gamma(\lambda), \ cm^{-1} \) |
|---------------------|------------------|
| \( \geq 10^9 \)     | \( \geq 1.0 \)    |
| \( 10^7 \)          | \( \approx 0.1 \) |
| \( 2.5 \times 10^8 \)| \( \approx 0.025 \) |

**9. Conclusions**

The basic equations describing SBS and STS are used to determine the spectral profiles of the gain. The linear (single-photon) and two-photon absorptions are compared.

In the stokes region the conventional SBS is shifted by the thermal SBS. In contrast to linear absorption, for two-photon one this shift depends on the pump intensity \( I_p \). In the anti-stokes region the positive two-photon thermal gain being proportional to \( I_p \) is added to the negative conventional gain, and the positive overall SBS gain can be achieved when \( I_p \) is sufficiently high.

Estimates made for liquid hexane and the pump wavelength 308 nm show that the typical extra shift of the stokes SBS component is 0.007 cm\(^{-1}\). The spectral resolution of a Fabry-Perot etalon is limited by several MHz (or \( 10^{-5} \) cm\(^{-1}\)). To reach the higher spectral resolution the methods of heterodyning and intensity fluctuations correlation should be used.

For a Fabry-Perot etalon based spectrum analyzer the linear STS-2 and two-photon STS-2 components are experimentally indistinguishable not only from one another, but also from the STS-1 component.

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