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

V. B. Karpov*, V. V. Korobkin

Coherent and Nonlinear Optics Department, A.M.Prokhorov General Physics Institute, Russian Academy of Sciences, Vavilov Street 38, 119991 Moscow, Russia

Email address:
karpov@kapella.gpi.ru (V. B. Karpov)

To cite this article:

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-infrared spectral region (the pump wavelength is \( \lambda_p = 0.69 \div 1.06 \mu m \) ) such a transformation has been originally observed in [3, 4]. For the near-ultraviolet spectral region (\( \lambda_p = 193 \div 351 \text{nm} \) ) such a transformation has been originally observed in [5, Fig. 2]. Indeed, in the previous near-ultraviolet 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-ultraviolet, 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 \( 10^{-5} \text{ cm}^{-1} \)) [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]

\[
\alpha_c = \alpha + (I \gamma),
\]

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 processes of the absorbed electromagnetic energy. In our 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]

\[
\alpha_c = \alpha + (I \gamma),
\]

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 processes of the absorbed electromagnetic energy. In our first experimentally discovered in [5] was not studied theoretically and is also a subject of interest.

\[
\Delta \rho = \frac{\partial}{\partial t} (\rho \nu \text{div} (V)) + \rho \nu \text{div} (V) = 0 ,
\]

\[
\left( \rho c_v \frac{\partial}{\partial t} - \lambda \nabla \cdot \nabla (\Delta T) - \frac{c_v (\delta - 1)}{\beta} \frac{\partial}{\partial t} (\Delta \rho) =
\right.

\[
= \frac{nc \alpha_e}{4\pi} (E^2) - \frac{1}{8\pi} \left( \rho \frac{\partial}{\partial t} (\nabla \cdot E) \right) \left( \rho_0 \frac{\partial}{\partial t} (E^2) \right) .
\]

Here \( v = \frac{1}{\sqrt{\rho_0 \rho_s}} \) is the speed of sound in a medium with adiabatic compressibility \( \beta_s \), \( c \) is the speed of light in vacuum, \( \delta = \frac{c_v}{c_e} \) is a ratio of specific heats (a frequently used symbol \( \gamma \) in (1) provides the logical connection with [5]), \( \beta = -\frac{1}{\rho_s} \left( \frac{\partial \rho}{\partial T} \right)_p \) is a volumetric thermal expansion coefficient at constant pressure, \( \gamma = \frac{\rho_s (\partial \rho)}{\rho_s (\partial T)} \) is an electrostriction coefficient, \( \lambda \) is thermal conductivity, and \( n \) is a refractive index.

In the Navier-Stokes equation (2) the pressure deviation is expressed as [23, 24, 33 - 35]:

\[
\Delta P = \frac{v^2}{\delta} (\Delta \rho) + \frac{v^2}{\delta} \rho_0 \beta (\Delta T) ,
\]

and its gradient is moved into the left-hand side (\( \text{grad} P = 0 \)). The right-hand side of (2) represents the electrostrictive force [35]. The first and second right-hand side terms of the heat equation (4) represent heating due to the light absorption (both the linear and two-photon introduced by \( \alpha \)) and heating due to the electrocaloric effect [1, 36], respectively.

The Navier-Stokes equation (2) and the continuity equation (3) can be combined into one by eliminating the macroscopic velocity vector \( V \):

\[
\Delta \rho = \frac{\partial^2}{\partial t^2} (\rho \nu \text{div} (V)) + \frac{\partial}{\partial t} \left( \frac{\partial \rho}{\partial t} \right) \text{div} (\nabla \cdot E) + \left( \frac{\partial \rho}{\partial T} \right) \text{div} (\nabla \cdot E) (\Delta T) =
\]

\[
= \frac{T}{8\pi} \nabla \cdot (E^2) .
\]

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 \( \varepsilon_0 \):

\[
\Delta \varepsilon = \Delta \varepsilon^e + \Delta \varepsilon^{nl} = \frac{\partial \varepsilon}{\partial \rho} \Delta \rho + \frac{\partial \varepsilon}{\partial T} \Delta T =
\]

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

\[
\frac{\partial \epsilon}{\partial \rho} \Delta^{NL} \rho > > \left( \frac{\partial \epsilon}{\partial T} \right)_{\rho} \Delta^{NL} T,
\]

\[
\frac{\partial \epsilon}{\partial \rho} = -\beta \left( \rho \frac{\partial \epsilon}{\partial \rho} \right)_{\rho}.
\]

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 electrical field vectors \( E_p \) and \( E_s \):

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

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

Here, \( \epsilon \) is a common [21] unit vector (for definiteness sake \( \epsilon = \epsilon_X \)); \( E_1(z,t) \) 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]

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

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

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

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

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

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

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

\[
\equiv \left( \frac{\partial \epsilon}{\partial \rho} \right)_{\rho} \Delta^{NL} \rho(z,t) E(z,t). \quad (10)
\]

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

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

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

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

The waves are coupled by the scalar electrodynamical equations [1, 2, 17, 32, 37]

\[
\nabla^2 E - \epsilon(\omega) \frac{\partial^2}{\partial t^2} E_p = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} P^{NL}_{\omega}, \quad (11)
\]

\[
\nabla^2 E - \epsilon^{NL}(\omega, z, t) \frac{\partial^2}{\partial t^2} E_s = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} P^{NL}_{\omega}. \quad (12)
\]

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 ([\( \frac{\partial^2 E}{\partial x^2} \frac{\partial^2 E}{\partial y^2} E_{zy}(z,t) = 0 \).]

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 \( E^2 \)

\[
\langle E^2 \rangle = \langle (E_p + E_s)^2 \rangle = \langle 2 E_p E_s \rangle = \frac{1}{2} \left( \begin{array}{c} E_1(z) E_1^*(z) \exp \left[ -i(\omega_1 - \omega_2) t \right] + c.c. \end{array} \right), \quad (13)
\]

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

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

\[
\Delta^{NL} T(z,t) = \frac{1}{2} \left( \begin{array}{c} T(z) \exp \left[ i(\omega_1 + \omega_2) t \right] + c.c. \end{array} \right). \quad (15)
\]
On substitution of (6), (13) - (15) into (4) and (5), a linear system for the complex amplitudes \( \rho_\alpha(z) \), \( T_\alpha(z) \) and for the product \( E_i(z)E^*_\alpha(z) \) is found

\[
\begin{align*}
-\Omega^2 + \frac{v^2}{\delta}q^2 + i\frac{\eta}{\rho_0}q^2\Omega \rho_s \\
+ i^2 \frac{v^2}{\delta}q^2 T_s = \frac{1}{8\pi} \left( \rho_0 \frac{\partial \rho}{\partial \rho} \right)_T q^2 E_i E^*_\alpha
\end{align*}
\]

\( \text{(16)} \)

Rearrangement of

\[
\rho = \frac{1}{2} \left[ \frac{1}{\beta} \left( \rho_0 \frac{\partial \rho}{\partial \rho} \right)_T q^2 \right] E_i E^*_\alpha
\]

\( \text{(17)} \)

where \( q = k_1 + k_2 \). Solving (16) for \( T_s \)

\[
T_s = \frac{\delta}{q^2 v^2 \beta \rho_0} \left[ \left( \frac{1}{8\pi} \left( \rho_0 \frac{\partial \rho}{\partial \rho} \right)_T q^2 E_i E^*_\alpha \right) - i\frac{\eta}{\rho_0} q^2 \Omega \rho_s \right]
\]

and inserting the result into (17) we obtain

\[
\begin{align*}
i\frac{c_v}{\beta} \left[ \frac{1}{8\pi} \left( \rho_0 \frac{\partial \rho}{\partial \rho} \right)_T q^2 E_i E^*_\alpha \right] - i\frac{\eta}{\rho_0} q^2 \Omega \rho_s
\end{align*}
\]

Rearrangement of \( \rho_\alpha \) and \( E_i E^*_\alpha \) into the opposite sides gives

\[
\begin{align*}
\rho_\alpha = \frac{\delta}{q^2 v^2 \beta \rho_0} \left[ \left( \frac{1}{8\pi} \left( \rho_0 \frac{\partial \rho}{\partial \rho} \right)_T q^2 E_i E^*_\alpha \right) - i\frac{\eta}{\rho_0} q^2 \Omega \rho_s \right]
\end{align*}
\]

\( \text{(17)} \)

Upon multiplying the both sides by \( -\frac{8\pi q^2 v^2}{\delta} \rho_s \) we have

\[
\begin{align*}
-\Omega^2 + \frac{v^2}{\delta}q^2 \left( \lambda q^2 - i\rho_0 c_v \Omega \right) \rho_s \\
+ i\frac{v^2}{\delta}q^2 \rho_s T_s = \frac{1}{8\pi} \left( \rho_0 \frac{\partial \rho}{\partial \rho} \right)_T q^2 E_i E^*_\alpha
\end{align*}
\]

\( \text{(18)} \)

By the use of the expressions \( \beta_\delta (\delta - 1) = \frac{\beta_\delta}{\rho_0 c_v} \) and

\[
v^2 = \frac{1}{\rho_0 c_v} \text{ the other form of the last term in (18) is achieved}
\]

\[
i\frac{v^2}{\delta} \rho_0 \beta_\delta T_s \Omega = \frac{1}{\delta} \left( 1 - \frac{1}{\delta} \right) \rho_0 c_v \Omega
\]

From the standpoint of a cubic nonlinear susceptibility tensor, projections of the nonlinear polarization vectors appearing in (11), (12) into the Cartesian coordinates are [37]

\[
\left( \rho_\alpha \right) = \chi^{(3)}(\omega_1, \omega_2, \Omega) \left( E(z, t) \right)_1 \left( E(z, t) \right)_2 \left( E(z, t) \right)_3
\]

\( \text{(19)} \)

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

\[
\left( \rho_\alpha \right) = \chi^{(3)}(\omega_1, \omega_2, \Omega) \left( E(z, t) \right)_1 \left( E(z, t) \right)_2 \left( E(z, t) \right)_3
\]

\( \text{(20)} \)

\( \text{for the isotropic branch } \chi^{(3)} \text{ is a scalar:} \)

\[
\left( \rho_\alpha \right) = \chi^{(3)}(\omega_1, \omega_2, \Omega) \left( E(z, t) \right)_1 \left( E(z, t) \right)_2 \left( E(z, t) \right)_3
\]

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

$$
\chi^{(3)} = \frac{1}{4\pi^2} \left( \rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_t \cdot \frac{1}{16\pi\rho_0} \times 
$$

$$
\left\{ -2\beta_n c_{\omega} \rho_0 \frac{v^2 q^2}{\delta} + \left( \rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_t q^2 \left[ \lambda_1 q^2 - i\rho_0 c_{\omega} \Omega \right] + i\left( 1 - \frac{1}{\delta} \right) \rho_0 c_{\omega} \Omega v^2 q^2 \right\} \times 
$$

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

(21)

In our case $D\in\mathbb{R}$ [2]. The cubic nonlinear susceptibility (21) exhibits Rayleigh (labeled with R) resonance at $|\Omega|=0$ and Brillouin (labeled with B) resonance at $|\Omega|=\Omega_{\pm} = \pm (k_x + k_y)\Omega_x$.

Rayleigh resonances. For $\Omega \approx 0$, $|\Omega|<<\Omega_{\pm}$ (21) incorporates electrostrictive (labeled with R1) and absorptive (labeled with R2) terms:

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

where

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

(22)

$$
\chi^{(3)R2} = \frac{1}{64\pi^2 D} \left( \rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_t \frac{\alpha c\varepsilon_n \beta}{\rho_0 c_{\omega} \rho_0 \Omega + \Omega_{\pm}}
$$

(23)

$$
\Gamma_{\pm} = \frac{\lambda_1 q^2}{\rho_0 c_{\omega} \rho_0}
$$

The imaginary parts of (22) and (23) are (as to the origin of $\beta^{\pm}_n$ and $\beta^{\pm}_n$ see (34) below)

$$
\text{Im} \chi^{(3)R1} = -\frac{1}{32\pi^2 D} \left( \rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_t \left[ \frac{2 - \delta}{2(\delta - 1)} \right] \frac{1}{1 + \Omega / \Omega_{\pm}}
$$

(24)

$$
\text{Im} \chi^{(3)R2} = \frac{1}{64\pi^2 D} \left( \rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_t \frac{\alpha c\varepsilon_n \beta}{\rho_0 c_{\omega} \rho_0 \Omega + \Omega_{\pm}}
$$

(25)

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

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

where

$$
\chi^{(3)B1} = \frac{1}{64\pi^2 D} \left( \rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_t \beta_n (2 - \delta) \frac{1}{\eta_0 \Omega} \frac{\Omega_{\pm} / 2}{\Omega - \Omega_{\pm} \pm \Omega / \Omega_{\pm}}
$$

(26)

$$
\chi^{(3)B2} = \pm \frac{i}{32\pi^2 D} \left( \rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_t \frac{\alpha c\varepsilon_n \beta}{\rho_0 c_{\omega} \rho_0 \Omega - \Omega_{\pm} \pm \Omega / \Omega_{\pm}}
$$

(27)

$$
\Gamma_{\pm} = \frac{\eta_0^2}{\rho_0}
$$

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

$$
\text{Im} \chi^{(3)B1} = \pm \frac{1}{64\pi^2 D} \left( \rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_t \beta_n (2 - \delta) \frac{1}{\eta_0 \Omega} \frac{\Gamma_{\pm} / 4}{\Omega - \Omega_{\pm} \pm \Omega / \Omega_{\pm} + \Omega_{\pm} / \Omega_{\pm}}
$$

(28)

$$
\text{Im} \chi^{(3)B2} = \pm \frac{1}{32\pi^2 D} \left( \rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_t \beta_n (2 - \delta) \frac{1}{\eta_0 \Omega} \frac{1}{2[2\Omega / \Omega_{\pm} / \Omega_{\pm} + \Omega_{\pm} / \Omega_{\pm} + \Omega_{\pm} / \Omega_{\pm} + \Omega_{\pm} / \Omega_{\pm}] + 1}
$$

A couple of equal in magnitude to
Nonlinear amplification of the Brillouin-Rayleigh triplet caused by two-photon heating

V. B. Karpov et al.

\[ \Im \chi_{\text{MAX}}^{(3)B_{1}} \approx \frac{1}{64 \pi^{2}} \left( \rho_{0} \frac{\partial \varepsilon}{\partial \rho} \right)_{T}^{2} \beta_{1} (2 - \delta) \frac{\rho_{0} v}{\eta q}, \tag{30} \]

peaks of the electrostrictive (conventional) Brillouin term defined by (28) demonstrate positive \( G \approx -\Im \chi^{(3)} \) for \( \Omega \approx \Omega_{B} \), and negative \( G \approx -\Im \chi^{(3)} \) for \( \Omega \approx -\Omega_{B} \) (Fig. 1a).

Four equal in magnitude to

\[ \Im \chi_{\text{MAX}}^{(3)R_{1,2}} = \frac{\alpha \varepsilon \cos \beta}{64 \pi^{2} D_{c} \rho_{0}} \left( \rho_{0} \frac{\partial \varepsilon}{\partial \rho} \right)_{T}, \tag{31} \]

peaks of the absorptive (thermal) Brillouin term defined by (29) demonstrate positive \( G \approx -\Im \chi^{(3)} \) both 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):

\[ \frac{\Im \chi_{\text{MAX}}^{(3)R_{1,2}}}{\Im \chi_{\text{MAX}}^{(3)B_{1}}} = \frac{\alpha \varepsilon \cos \beta}{64 \pi^{2} D_{c} \rho_{0}} \left( \rho_{0} \frac{\partial \varepsilon}{\partial \rho} \right)_{T}; \tag{32} \]

where

\[ \frac{\beta}{\beta_{S}} = \frac{\rho_{0} C_{P} \frac{\sigma - 1}{\beta T_{0}}}{\beta_{S}}, \quad \lambda_{1} = \frac{2 \pi v}{\omega_{1}}, \quad k_{1} = \frac{2 \pi n}{\lambda_{1}}, \]

\[ \Omega_{B} = qv = 2 k_{1} v \sin(\theta / 2) = -\frac{4 \pi v n}{\lambda_{1}} \sin(\theta / 2) \]

\( \lambda_{1} \) is the pump wavelength and \( \theta \) is the angle of scattering.

On substitution of the resulting expressions for \( \rho_{0}^{\text{NL}} \) into (11), (12) we derive a system [17]:

\[ \left( \frac{\partial}{\partial z} + \alpha_{z} \right) |E_{1}(z)|^{2} = -G |E_{1}(z)|^{2} |E_{2}(z)|^{2}. \tag{33} \]

Following (33), when the pump intensity \( I_{p}(z) \propto |E_{1}(z)|^{2} \) is treated as constant over the length \( L \) of nonlinear interaction, the scattered wave intensity \( I_{s}(z) \propto |E_{2}(z)|^{2} \) exponentially increases along \( z \) with a gain coefficient

\[ g = G |E_{1}|^{2} - \alpha_{z}, \]

where \( G \approx -\Im \chi^{(3)} \) is a gain parameter. The general formula for \( G \) is [17] (See (24), (25), (28), and (29))

\[ G(\Omega) = \pm \beta_{B}^{2} \frac{1}{1 + (2 \Delta \Omega / \Gamma_{B})^{2}}, \tag{34} \]

\[ + \left( \beta_{R}^{2} - \beta_{B}^{2} \right) \frac{\Omega / \Gamma_{R}}{1 + (\Omega / \Gamma_{R})^{2}} \]

where

\[ \Omega = \omega_{1} - \omega_{2}, \quad \Delta \Omega = |\Omega - \Omega_{B} - \Omega_{B} = (k_{1} + k_{2})v, \]

\[ \Gamma_{B} = \frac{\eta (k_{1} + k_{2})^{2}}{\rho_{0}}, \quad \Gamma_{R} = \frac{\lambda_{1}^{2} (k_{1} + k_{2})^{2}}{\rho_{0} C_{P}} \tag{35} \]

In the first two expressions of (34) signs “+” and “-” correspond to the stokes \((\omega_{1} > \omega_{2}, \Omega > 0)\) and anti-stokes \((\omega_{1} < \omega_{2}, \Omega < 0)\) 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 \( \beta_{B}^{2} \) in (34) represents the conventional 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 \( \beta_{R}^{2} \approx \alpha \varepsilon \approx \alpha \) (see (29)) represents the thermal stokes 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 $\Gamma / 2$.

The term proportional to $\beta^\omega_\gamma \propto \alpha_\Sigma = \alpha$ (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 $\Gamma / R$.

The term proportional to $\beta^\omega_\gamma$ 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](image)

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 $\gamma_p$. In particular, $\beta^\omega_\gamma, \beta^\gamma_\gamma \propto \gamma_p$.

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 $\Gamma / R$, 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 ($\Omega_B >> \Gamma_B >> \Gamma_R$ = 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 $\gamma_p$ (see Section 7). In the anti-stokes region the positive two-photon thermal values, being proportional to $\beta^\gamma_\gamma \propto I_p$, are added to the negative conventional values, being proportional to $\beta^\gamma_\gamma = \text{const}$, 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 $\beta_B$ and dividing it by $\beta_B^\omega$, we obtain

$$\left(\frac{\beta_B}{\beta_B^\omega}\right) = \frac{1}{1 + (2\Delta \Omega / \Gamma_B)^2} \quad \text{(36)}$$

Setting

$$\left(\frac{\beta_B}{\beta_B^\omega}\right) = Z, \quad \left(\frac{\beta_B^\gamma}{\beta_B^\omega}\right) = Y, \quad \left(2\Delta \Omega / \Gamma_B\right) = X,$$

we rewrite (36) as

$$Z(X,Y) = \frac{1}{1 + X^2} + Y \frac{X}{1 + X^2} \quad \text{(37)}$$

The parameters $\beta_B^\omega, \beta_B^\gamma$, 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 $\Delta \Omega = \Gamma_B / 2$. 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))

$$Y = \frac{\beta_B^\omega}{\beta_B} = \frac{\delta - 1}{2 - \delta} \frac{\alpha_\lambda}{4\pi\beta} \frac{c/\nu}{\sin(\theta/2)\rho_0 \frac{\partial \theta}{\partial \rho}} \quad \text{(38)}$$

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

$$Y = (\text{const}) \times \alpha \quad \text{(39)}$$

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

$$Y = (\text{const}) \times (\gamma_p) \quad \text{(40)}$$

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 $\text{const} \times \beta_s(X)$ 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.


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,

$$\eta = \frac{2\eta_1}{3}, \quad k_1 + k_2 = \frac{4m}{\lambda_1},$$

where $\eta_1$ is a shear viscosity coefficient. From (35) we obtain

$$\Gamma_s = \frac{2\eta_1}{3\rho_0} \left( \frac{4\pi n^2\eta_1}{\lambda_1} \right)^2 = \frac{32\pi^2n^2\eta_1}{3\rho_0\lambda_1^2}. \quad (41)$$

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

- $\lambda_i = 3 \times 10^{-2}$ cm, $\theta = \pi$, $\rho_0 = 0.66 \frac{g}{cm^3}$, $n = 1.4$,
- $\eta_1 = 3.2 \times 10^{-3}$ $\rho = 3.2 \times 10^{-1} \frac{g}{cm^3}$,
- $\beta = 1.4 \times 10^{-10}$ K$^{-1}$, $T_s = 300$ K,
- $\nu = 10^7 \frac{cm}{s}$, $c = 3 \times 10^5 \frac{cm}{s}$, $\rho_0 \left( \frac{\partial \rho}{\partial \nu} \right) = 1$, $\delta = \frac{C_p}{C_v} = 1.3$.

Then (41) yields

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

Equation (38) gives

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

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

For two-photon absorption $\alpha$ is substituted by $\gamma_p$ in (42), and

$$Y = 0.73 \times (\gamma_p), \quad (43)$$

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

$$Y^\text{max} = 0.73 \times (\gamma_p^\text{max}) = 0.73.$$

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 $X \approx 0.5$, and the frequency shift is

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

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 \( \gamma_p = 0.1 cm^{-1} \) (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 \( \gamma_p \) to the total absorption coefficient at \( \lambda = 308nm \) in hexane for the three values of the pump intensity \( I_p \)

<table>
<thead>
<tr>
<th>( I_p, W / cm^2 )</th>
<th>( \gamma_p, cm^{-1} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \geq 10^9 )</td>
<td>( \geq 1.0 )</td>
</tr>
<tr>
<td>( 10^8 )</td>
<td>( \approx 0.1 )</td>
</tr>
<tr>
<td>( 2.5 \times 10^8 )</td>
<td>( \approx 0.025 )</td>
</tr>
</tbody>
</table>

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 gain being proportional to \( I_p \), two-photon absorptions are compared. 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 (sin-

References

[21] I.L.Fabelinskii, Molecular Scattering of Light. New York:


