

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 $\lambda_1 = 0.69 \div 1.06 \mu\text{m}$) such a transformation has been originally observed in [3, 4]. For the near-uv spectral region ($\lambda_1 = 193 \div 351 \text{ nm}$) 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} 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 $\alpha_{\text{eff}}(\omega)$ has been introduced [24] for gases to describe the thermalization processes of the absorbed electromagnetic energy. In our analysis a total absorption coefficient [5]

$$\alpha_{\Sigma} = \alpha + (I_p \gamma), \quad (1)$$

should be used to describe the two-photon effect (α is a linear absorption coefficient and γ 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 the Navier-Stokes equation is used instead of the Lagrange one. Indeed, any plane acoustic wave propagating in a continuous medium provides shear motion leading to the attenuation due to viscosity η [31] (see (2) below).

3. Material Equations

Interaction of a light wave, characterized by a total electrical field vector \mathbf{E} , with an isotropic dielectric medium is described by the hydrodynamic equations linearized with respect to the small deviations of density $\Delta\rho$, temperature ΔT , pressure ΔP , and a macroscopic velocity vector \mathbf{V} from the equilibrium values ρ_0, T_0, P_0 , and $\mathbf{V}_0 = 0$ [1, 2, 4, 17, 32]:

$$\begin{aligned} & \rho_0 \frac{\partial \mathbf{V}}{\partial t} + \frac{v^2}{\delta} \text{grad}(\Delta\rho) \\ & + \frac{v^2 \beta \rho_0}{\delta} \text{grad}(\Delta T) - \eta \nabla^2 \mathbf{V} = \\ & = \frac{\gamma^e}{8\pi} \text{grad}(\mathbf{E}^2), \end{aligned} \quad (2)$$

$$\frac{\partial}{\partial t}(\Delta\rho) + \rho_0 \text{div}(\mathbf{V}) = 0, \quad (3)$$

$$\begin{aligned} & \left(\rho_0 c_V \frac{\partial}{\partial t} - \lambda_T \nabla^2 \right) (\Delta T) - \frac{c_V (\delta - 1)}{\beta} \frac{\partial}{\partial t} (\Delta\rho) = \\ & = \frac{nc\alpha_{\Sigma}}{4\pi} (\mathbf{E}^2) - \frac{1}{8\pi} \left(\frac{\partial \varepsilon}{\partial T} \right)_p \left(T_0 \frac{\partial}{\partial t} (\mathbf{E}^2) \right). \end{aligned} \quad (4)$$

Here $v = \sqrt{\frac{1}{\rho_0 \beta_s}}$ is the speed of sound in a medium with adiabatic compressibility β_s , c is the speed of light in vacuum, $\delta = \frac{c_p}{c_v}$ is a ratio of specific heats (a frequently used symbol γ in (1) provides the logical connection with [5]), $\beta = -\frac{1}{\rho_0} \left(\frac{\partial \rho}{\partial T} \right)_p$ is a volumetric thermal expansion coefficient at constant pressure, $\gamma^e = \rho_0 \left(\frac{\partial \varepsilon}{\partial \rho} \right)_T$ is an electrostriction coefficient, λ_T 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 ($\text{grad } P_0 = 0$). 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 α_{Σ}) 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 \mathbf{V} :

$$\begin{aligned} & \left(-\frac{\partial^2}{\partial t^2} + \frac{v^2}{\delta} \nabla^2 \right) (\Delta\rho) + \frac{v^2 \beta \rho_0}{\delta} \nabla^2 (\Delta T) = \\ & = \frac{\gamma^e}{8\pi} \nabla^2 (\mathbf{E}^2). \end{aligned} \quad (5)$$

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 ε_0 :

$$\begin{aligned} \Delta \varepsilon &= \Delta^{SP} \varepsilon + \Delta^{NL} \varepsilon = \left(\frac{\partial \varepsilon}{\partial \rho} \right)_T \Delta \rho + \left(\frac{\partial \varepsilon}{\partial T} \right)_\rho \Delta T = \\ &= \left(\frac{\partial \varepsilon}{\partial \rho} \right)_T (\Delta^{SP} \rho + \Delta^{NL} \rho) + \left(\frac{\partial \varepsilon}{\partial T} \right)_\rho (\Delta^{SP} T + \Delta^{NL} T). \end{aligned}$$

Usually [1, 2, 17, 32]

$$\left| \left(\frac{\partial \varepsilon}{\partial \rho} \right)_T \Delta^{NL} \rho \right| \gg \left| \left(\frac{\partial \varepsilon}{\partial T} \right)_\rho \Delta^{NL} T \right| ,$$

$$\left(\frac{\partial \varepsilon}{\partial T} \right)_\rho \approx -\beta \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T . \quad (6)$$

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

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 \mathbf{E}_p and

\mathbf{E}_s :

$$\mathbf{E}_p = \frac{1}{2} \mathbf{e} \{ E_1(z, t) \exp(i k_1 z - i \omega_1 t) + c.c. \} , \quad (7)$$

$$\mathbf{E}_s = \frac{1}{2} \mathbf{e} \{ E_2(z, t) \exp(-i k_2 z - i \omega_2 t) + c.c. \} . \quad (8)$$

Here, \mathbf{e} is a common [21] unit vector (for definiteness sake $\mathbf{e} = \mathbf{e}_x$); $E_1(z, t)$ and $E_2(z, t)$ are complex amplitudes; ω_1, ω_2 and k_1, k_2 are temporal frequencies and wave numbers, respectively. A total electric field vector is

$$\mathbf{E} = \mathbf{E}_p + \mathbf{E}_s . \quad (9)$$

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

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

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

where $\hat{\varepsilon}(\omega)$ and $\hat{\varepsilon}^{NL}(\omega, z, t)$ are the Fourier transforms of the linear and nonlinear permittivity tensors $\hat{\varepsilon}(t_1)$ 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]:

$$\mathbf{D}^L(z, t) = \varepsilon(\omega) \mathbf{E}(z, t) ,$$

$$\begin{aligned} \mathbf{D}^{NL}(z, t) &= \varepsilon^{NL}(\omega, z, t) \mathbf{E}(z, t) \cong \\ &\cong \left(\left(\frac{\partial \varepsilon}{\partial \rho} \right)_T \Delta^{NL} \rho(z, t) + \left(\frac{\partial \varepsilon}{\partial T} \right)_\rho \Delta^{NL} T(z, t) \right) \mathbf{E}(z, t) \cong \\ &\cong \left(\frac{\partial \varepsilon}{\partial \rho} \right)_T \Delta^{NL} \rho(z, t) \mathbf{E}(z, t) . \end{aligned}$$

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

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

$$\begin{aligned} \mathbf{P}^{NL}(z, t) &= \frac{1}{4\pi} \varepsilon^{NL}(\omega, z, t) \mathbf{E}(z, t) \\ &\cong \frac{1}{4\pi} \left(\frac{\partial \varepsilon}{\partial \rho} \right)_T \Delta^{NL} \rho(z, t) \mathbf{E}(z, t) . \end{aligned} \quad (10)$$

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

$$\left[\nabla^2 - \frac{\varepsilon(\omega_1)}{c^2} \frac{\partial^2}{\partial t^2} \right] \mathbf{E}_p = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{P}_{\omega_1}^{NL} , \quad (11)$$

$$\left[\nabla^2 - \frac{\varepsilon(\omega_2)}{c^2} \frac{\partial^2}{\partial t^2} \right] \mathbf{E}_s = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{P}_{\omega_2}^{NL} , \quad (12)$$

with the right-hand sides representing the nonlinear polarizations oscillating with the frequencies ω_1 and ω_2 . For the plane waves diffraction is absent ($\left[\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right] E_{p,s}(z, t) = 0$).

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

$$\begin{aligned} \langle \mathbf{E}^2 \rangle &= \langle (\mathbf{E}_p + \mathbf{E}_s)^2 \rangle = \langle 2 \mathbf{E}_p \mathbf{E}_s \rangle = \\ &= \frac{1}{2} \left\{ E_1(z) E_2^*(z) \exp \left[\begin{array}{l} -i(\omega_1 - \omega_2) t \\ +i(k_1 + k_2) z \end{array} \right] + c.c. \right\} , \end{aligned} \quad (13)$$

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

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

$$\Delta^{NL} T(z, t) = \frac{1}{2} \left\{ T_a(z) \exp \left[\begin{array}{l} -i(\omega_1 - \omega_2) t \\ +i(k_1 + k_2) z \end{array} \right] + c.c. \right\} . \quad (15)$$

On substitution of (6), (13) - (15) into (4) and (5), a linear system for the complex amplitudes $\rho_a(z)$, $T_a(z)$ and for the product $E_1(z)E_2^*(z)$ is found

$$\left(-\Omega^2 + \frac{v^2}{\delta} q^2 + i \frac{\eta}{\rho_0} q^2 \Omega \right) \rho_a + \frac{v^2 \beta \rho_0}{\delta} q^2 T_a = \frac{1}{8\pi} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T q^2 E_1 E_2^* \quad (16)$$

$$i\Omega \frac{c_v(\delta-1)}{\beta} \rho_a + (-i\rho_0 c_v \Omega + \lambda_T q^2) T_a = \frac{1}{4\pi} n c \alpha_\Sigma E_1 E_2^* - \frac{i}{8\pi} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T \beta T_0 \Omega E_1 E_2^*, \quad (17)$$

where $q = k_1 + k_2$, $\Omega = \omega_1 - \omega_2$. Solving (16) for T_a

$$T_a = \frac{\delta}{q^2 v^2 \beta \rho_0} \left[\frac{1}{8\pi} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T q^2 E_1 E_2^* - \left(-\Omega^2 + \frac{v^2}{\delta} q^2 + i \frac{\eta}{\rho_0} q^2 \Omega \right) \rho_a \right]$$

and inserting the result into (17) we obtain

$$i\Omega \frac{c_v(\delta-1)}{\beta} \rho_a + \frac{\delta(-i\rho_0 c_v \Omega + \lambda_T q^2)}{q^2 v^2 \beta \rho_0} \left[\frac{1}{8\pi} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T q^2 E_1 E_2^* - \left(-\Omega^2 + \frac{v^2}{\delta} q^2 + i \frac{\eta}{\rho_0} q^2 \Omega \right) \rho_a \right] = \frac{1}{4\pi} \left[n c \alpha_\Sigma - \frac{i}{2} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T \beta T_0 \Omega \right] E_1 E_2^*.$$

Rearrangement of ρ_a and $E_1 E_2^*$ into the opposite sides gives

$$\rho_a \left[i\Omega \frac{c_v(\delta-1)}{\beta} - \frac{\delta(-i\rho_0 c_v \Omega + \lambda_T q^2)}{q^2 v^2 \beta \rho_0} \left(-\Omega^2 + \frac{v^2}{\delta} q^2 + i \frac{\eta}{\rho_0} q^2 \Omega \right) \right] = E_1 E_2^* \left[\frac{\delta(i\rho_0 c_v \Omega - \lambda_T q^2)}{8\pi q^2 v^2 \beta \rho_0} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T q^2 + \frac{1}{4\pi} \left[n c \alpha_\Sigma - \frac{i}{2} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T \beta T_0 \Omega \right] \right].$$

Upon multiplying the both sides by $\left(-\frac{8\pi q^2 v^2 \beta \rho_0}{\delta} \right)$ we have

$$8\pi \rho_a \left[\begin{aligned} & \left(-\Omega^2 + \frac{v^2 q^2}{\delta} \right) (\lambda_T q^2 - i\rho_0 c_v \Omega) \\ & + i \frac{\eta q^2}{\rho_0} \Omega \\ & - i \left(1 - \frac{1}{\delta} \right) \rho_0 c_v \Omega v^2 q^2 \end{aligned} \right] = E_1 E_2^* \left[\begin{aligned} & - (i\rho_0 c_v \Omega - \lambda_T q^2) \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T q^2 \\ & - \frac{2q^2 v^2 \beta \rho_0}{\delta} \left[n c \alpha_\Sigma - \frac{i}{2} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T \beta T_0 \Omega \right] \\ & - 2\beta n c \alpha_\Sigma \rho_0 \frac{v^2 q^2}{\delta} \\ & + \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T q^2 \left[\lambda_T q^2 - i\rho_0 c_v \Omega + \frac{i}{\delta} v^2 \rho_0 \beta^2 T_0 \Omega \right] \end{aligned} \right]. \quad (18)$$

By the use of the expressions $\beta_s(\delta-1) = \frac{\beta^2 T_0}{\rho_0 c_p}$ and

$v^2 = \frac{1}{\rho_0 \beta_s}$ the other form of the last term in (18) is achieved

$$\begin{aligned} & \frac{i}{\delta} v^2 \rho_0 \beta^2 T_0 \Omega = \\ & \frac{i}{\delta} v^2 \Omega \beta_s (\delta-1) \rho_0^2 c_p = \\ & = i \left(1 - \frac{1}{\delta} \right) \rho_0 c_p \Omega \end{aligned} \quad (19)$$

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

$$\begin{aligned} & (P_{\omega_1, \omega_2}^{NL}(z, t))_i \\ & = \chi_{ijkl}^{(3)}(\omega_1, \omega_2, \Omega) (E(z, t))_j (E(z, t))_k (E(z, t))_l \end{aligned}$$

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

$$\begin{aligned} (P_{\omega_1, \omega_2}^{NL})_x &= \chi_{xxxx}^{(3)} (E)_x^3 = \chi_{xxxx}^{(3)} (E_P + E_S)_x^3 = \\ &= \chi_{xxxx}^{(3)} [(E_P)_x^3 + 3(E_P)_x^2 (E_S)_x + 3(E_P)_x (E_S)_x^2 + (E_S)_x^3] \end{aligned}$$

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

$$\begin{aligned} (P_{\omega_1, \omega_2}^{NL})_x &= \chi^{(3)} (E)_x^3 = \chi^{(3)} (E_P + E_S)_x^3 = \\ &= \chi^{(3)} [(E_P)_x^3 + 3(E_P)_x^2 (E_S)_x + 3(E_P)_x (E_S)_x^2 + (E_S)_x^3] \end{aligned} \quad (20)$$

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

$$\chi^{(3)} = \frac{1}{4\pi D} \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T \frac{1}{16\pi \rho_0} \times \left\{ -2\beta_{nc} \alpha_{\Sigma} \rho_0 \frac{v^2 q^2}{\delta} + \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T q^2 \begin{bmatrix} \lambda_T q^2 \\ -i\rho_0 c_v \Omega \\ +i \left(1 - \frac{1}{\delta} \right) \rho_0 c_p \Omega \end{bmatrix} \right\} \times \left\{ \begin{bmatrix} -\Omega^2 + \frac{v^2 q^2}{\delta} \\ +i \frac{\eta q^2}{\rho_0} \Omega \end{bmatrix} \begin{bmatrix} \lambda_T q^2 \\ -i\rho_0 c_v \Omega \end{bmatrix} - i \left(1 - \frac{1}{\delta} \right) \rho_0 c_v \Omega v^2 q^2 \right\}^{-1} \quad (21)$$

In our case $D=3$ [2]. The cubic nonlinear susceptibility (21) exhibits Rayleigh (labeled with R) resonance at $|\Omega| \approx 0$ and Brillouin (labeled with B) resonance at $|\Omega| \approx \Omega_B = qv = (k_1 + k_2)v$.

Rayleigh resonances. For $\Omega \approx 0, |\Omega| \ll \Omega_B$ (21) incorporates electrocaloric (labeled with R1) and absorptive (labeled with R2) terms:

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

where

$$\chi^{(3)R1} = -\frac{1}{32\pi^2 D} \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T^2 \beta_s (\delta - 1) \begin{bmatrix} \frac{2 - \delta}{2(\delta - 1)} \\ + \frac{i\Gamma_R}{\Omega + i\Gamma_R} \end{bmatrix} \quad (22)$$

$$\chi^{(3)R2} = \frac{1}{64\pi^2 D} \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T \frac{\alpha_{\Sigma} c n \beta}{\Gamma_R c_p \rho_0} \frac{i\Gamma_R}{\Omega + i\Gamma_R} \quad (23)$$

$$\Gamma_R = \frac{\lambda_T q^2}{\rho_0 c_p}$$

The imaginary parts of (22) and (23) are (as to the origin of β_R^e and β_R^a see (34) below)

$$\begin{aligned} \text{Im} \chi^{(3)R1} &= -\frac{1}{32\pi^2 D} \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T^2 \beta_s (\delta - 1) \frac{\Gamma_R \Omega}{\Omega^2 + \Gamma_R^2} \\ &= -\frac{1}{32\pi^2 D} \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T^2 \beta_s (\delta - 1) \frac{\Omega / \Gamma_R}{(\Omega / \Gamma_R)^2 + 1} \\ &= -\beta_R^e \frac{\Omega / \Gamma_R}{(\Omega / \Gamma_R)^2 + 1}, \end{aligned} \quad (24)$$

$$\begin{aligned} \text{Im} \chi^{(3)R2} &= \frac{1}{64\pi^2 D} \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T \frac{\alpha_{\Sigma} c n \beta}{\Gamma_R c_p \rho_0} \frac{\Gamma_R \Omega}{\Omega^2 + \Gamma_R^2} \\ &= \frac{1}{64\pi^2 D} \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T \frac{\alpha_{\Sigma} c n \beta}{\Gamma_R c_p \rho_0} \frac{\Omega / \Gamma_R}{(\Omega / \Gamma_R)^2 + 1} \\ &= \beta_R^a \frac{\Omega / \Gamma_R}{(\Omega / \Gamma_R)^2 + 1}. \end{aligned} \quad (25)$$

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

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

where

$$\chi^{(3)B1} = \frac{1}{64\pi^2 D} \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T^2 \beta_s (2 - \delta) \frac{\rho_0 v}{\eta q} \frac{\Gamma_B / 2}{|\Omega| - \Omega_B \pm i\Gamma_B / 2} \quad (26)$$

$$\chi^{(3)B2} = \pm \frac{i}{32\pi^2 D} \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T \frac{\alpha_{\Sigma} c n \beta}{\Gamma_B c_p \rho_0} \frac{\Gamma_B / 2}{|\Omega| - \Omega_B \pm i\Gamma_B / 2} \quad (27)$$

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

The bottom signs in (26), (27), (28), and (29) correspond to the Stokes ($\omega_1 > \omega_2, \Omega > 0, |\Omega| = \Omega$) 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 β_B^e and β_B^a see (34) below)

$$\begin{aligned} \text{Im} \chi^{(3)B1} &= \pm \frac{1}{64\pi^2 D} \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T^2 \beta_s (2 - \delta) \frac{\rho_0 v}{\eta q} \frac{\Gamma_B^2 / 4}{(|\Omega| - \Omega_B)^2 + \Gamma_B^2 / 4} \\ &= \pm \frac{1}{64\pi^2 D} \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T^2 \beta_s (2 - \delta) \frac{\rho_0 v}{\eta q} \frac{1}{[2(|\Omega| - \Omega_B) / \Gamma_B]^2 + 1} \\ &= \mp \beta_B^e \frac{1}{[2(|\Omega| - \Omega_B) / \Gamma_B]^2 + 1}. \end{aligned} \quad (28)$$

$$\begin{aligned} \text{Im} \chi^{(3)B2} &= \pm \frac{1}{32\pi^2 D} \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T \frac{\alpha_{\Sigma} c n \beta}{\Gamma_B c_p \rho_0} \frac{(|\Omega| - \Omega_B) \Gamma_B / 2}{(|\Omega| - \Omega_B)^2 + \Gamma_B^2 / 4} \\ &= \pm \frac{1}{32\pi^2 D} \left(\rho_0 \frac{\partial \epsilon}{\partial \rho} \right)_T \frac{\alpha_{\Sigma} c n \beta}{\Gamma_B c_p \rho_0} \frac{2(|\Omega| - \Omega_B) / \Gamma_B}{[2(|\Omega| - \Omega_B) / \Gamma_B]^2 + 1} \\ &= \mp \beta_B^a \frac{2(|\Omega| - \Omega_B) / \Gamma_B}{[2(|\Omega| - \Omega_B) / \Gamma_B]^2 + 1}. \end{aligned} \quad (29)$$

A couple of equal in magnitude to

$$\begin{aligned} \text{Im}\chi_{\text{MAX}}^{(3)\text{B1}} &= \frac{1}{64\pi^2 D} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T \beta_s (2 - \delta) \frac{\rho_0 v}{\eta q} \\ &= \frac{\beta_s (2 - \delta) \Omega_B}{64\pi^2 D \Gamma_B} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T \end{aligned} \quad (30)$$

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

$$\begin{aligned} \text{Im}\chi_{\text{MAX}}^{(3)\text{B2}} &= \frac{1}{32\pi^2 D} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T \frac{\alpha_z \text{cn}\beta}{\Gamma_B c_p \rho_0} \frac{\Gamma_B^2 / 4}{\Gamma_B^2 / 4 + \Gamma_B^2 / 4} \\ &= \frac{\alpha_z \text{cn}\beta}{64\pi^2 D \Gamma_B c_p \rho_0} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T \end{aligned} \quad (31)$$

peaks of the absorptive (thermal) Brillouin term defined by (29) demonstrate positive and negative $G \propto -\text{Im}\chi^{(3)}$ 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):

$$\begin{aligned} \frac{\text{Im}\chi_{\text{MAX}}^{(3)\text{B2}}}{\text{Im}\chi_{\text{MAX}}^{(3)\text{B1}}} &= \left(\frac{\alpha_z \text{cn}\beta}{64\pi^2 D \Gamma_B c_p \rho_0} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T \right) \left(\frac{64\pi^2 D \Gamma_B}{\beta_s (2 - \delta) \Omega_B} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T^{-2} \right) \\ &= \frac{\alpha_z \text{cn}\beta}{c_p \rho_0 \beta_s (2 - \delta) \Omega_B} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T^{-1} \\ &= \frac{(\delta - 1)}{(2 - \delta)} \frac{\alpha_z \text{cn}}{\beta T_0 \Omega_B} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T^{-1} \\ &= \frac{(\delta - 1)}{(2 - \delta)} \frac{\alpha_z \lambda_1}{4\pi \beta T_0} \frac{c/v}{\sin(\theta/2)} \left(\rho_0 \frac{\partial \varepsilon}{\partial \rho} \right)_T^{-1}, \end{aligned} \quad (32)$$

where

$$\frac{\beta}{\beta_s} = \frac{\rho_0 c_p (\delta - 1)}{\beta T_0}, \quad \lambda_1 = \frac{2\pi c}{\omega_1}, \quad k_1 = \frac{2\pi n}{\lambda_1},$$

$$\Omega_B = qv = 2k_1 v \sin(\theta/2) = \frac{4\pi v n}{\lambda_1} \sin(\theta/2)$$

λ_1 is the pump wavelength and θ is the angle of scattering.

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

$$\left(\frac{\partial}{\partial z} + \alpha_z \right) |E_1(z)|^2 = -G |E_1(z)|^2 |E_2(z)|^2,$$

$$\left(\frac{\partial}{\partial z} - \alpha_z \right) |E_2(z)|^2 = -G |E_1(z)|^2 |E_2(z)|^2. \quad (33)$$

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

$$\begin{aligned} G(\Omega) &= \pm \beta_B^e \frac{1}{1 + (2\Delta\Omega / \Gamma_B)^2} \\ &\pm \beta_B^a \frac{2\Delta\Omega / \Gamma_B}{1 + (2\Delta\Omega / \Gamma_B)^2} \\ &+ (\beta_R^e - \beta_R^a) \frac{\Omega / \Gamma_R}{1 + (\Omega / \Gamma_R)^2} \end{aligned} \quad (34)$$

where

$$\Omega = \omega_1 - \omega_2, \quad \Delta\Omega = |\Omega| - \Omega_B, \quad \Omega_B = (k_1 + k_2)v,$$

$$\Gamma_B = \frac{\eta(k_1 + k_2)^2}{\rho_0}, \quad \Gamma_R = \frac{\lambda_T(k_1 + k_2)^2}{\rho_0 C_p}. \quad (35)$$

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 β_B^e 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 Γ_B .

The term proportional to $\beta_B^a \propto \alpha_z = \alpha$ (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 $\Gamma_B / 2$.

The term proportional to $\beta_R^a \propto \alpha_\Sigma = \alpha$ (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 Γ_R .

The term proportional to β_R^e 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.

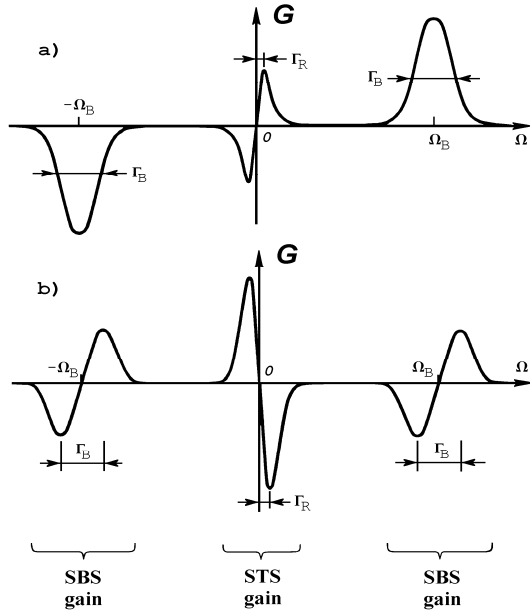


Figure 1. The gain parameter $G(\Omega)$ defined by (34) for the SBS and STS mechanisms: (a) conventional SBS and STS-1; (b) thermal SBS and STS-2.

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 α with \mathcal{A}_p . In particular, $\beta_R^a, \beta_B^a \propto \mathcal{A}_p$.

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 Γ_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 ($\Omega_B \gg \Gamma_B \gg \Gamma_R \approx \text{spectral resolution}$). In the Stokes region the conventional SBS must be shifted by the thermal

SBS. For linear absorption such a shift depends on α . For two-photon absorption such a shift depends on \mathcal{A}_p (see Section 7). In the anti-stokes region the positive two-photon thermal values, being proportional to $\beta_B^a \propto I_p$, are added to the negative conventional values, being proportional to $\beta_B^e = \text{const}$, 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 β_B and dividing it by β_B^e , we obtain

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

Setting

$$\left(\beta_B / \beta_B^e\right) \equiv Z, \quad \left(\beta_B^a / \beta_B^e\right) \equiv Y, \quad (2\Delta\Omega / \Gamma_B) \equiv X,$$

we rewrite (36) as

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

The parameters β_B^a , β_B^e , and Γ_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))

$$Y = \frac{\beta_B^a}{\beta_B^e} = \frac{\delta - 1}{2 - \delta} \frac{\alpha \lambda_1}{4\pi\beta T_0} \frac{c/v}{\sin(\theta/2)\rho_0(\partial\epsilon/\partial\rho)_T}, \quad (38)$$

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

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

For two-photon absorption α is substituted by \mathcal{A}_p in (38), and

$$Y = (\text{const}) \times (\mathcal{A}_p). \quad (40)$$

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.

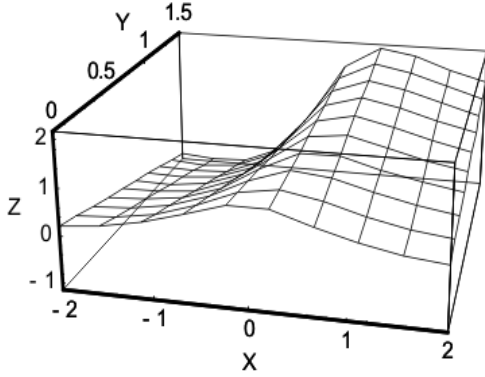


Figure 2. A three-dimensional plot of the overall Stokes SBS gain parameter for the conventional and thermal mechanisms.

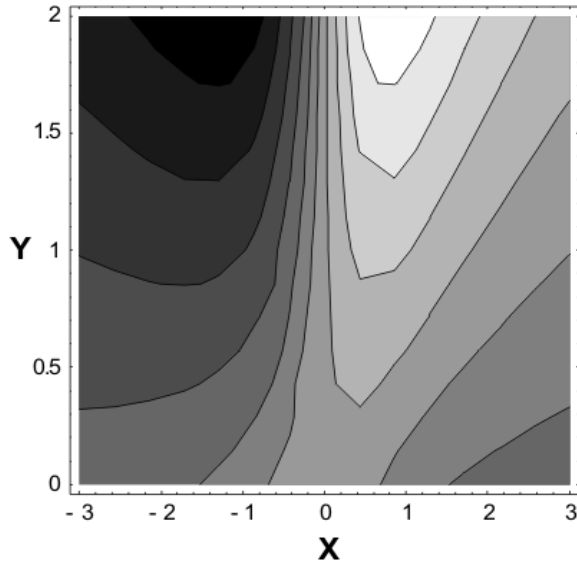


Figure 3. The contour plot corresponding to Fig. 2. The gray-scale legend is evident from Fig. 2.

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,

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

where η_1 is a shear viscosity coefficient. From (35) we obtain

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

Following [5], we perform estimates for liquid hexane (C6H14), $\lambda_1 = 308 \text{ nm}$, and the material parameters [1, 21, 38, 39]:

$$\begin{aligned} \lambda_1 &\approx 3 \times 10^{-5} \text{ cm}, \quad \theta = \pi, \quad \rho_0 \approx 0.66 \frac{\text{g}}{\text{cm}^3}, \quad n \approx 1.4, \\ \eta_1 &\approx 3.2 \times 10^{-3} \text{ P} \approx 3.2 \times 10^{-3} \frac{\text{g}}{\text{cm s}}, \\ \beta &\approx 1.4 \times 10^{-3} \text{ K}^{-1}, \quad T_0 \approx 300 \text{ K}, \\ v &\approx 10^5 \frac{\text{cm}}{\text{s}}, \quad c \approx 3 \times 10^{10} \frac{\text{cm}}{\text{s}}, \quad \rho_0 \left(\frac{\partial \epsilon}{\partial \rho} \right)_T \approx 1, \quad \delta = \frac{C_p}{C_v} \approx 1.3. \end{aligned}$$

Then (41) yields

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

Equation (38) gives

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

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

For two-photon absorption α is substituted by \mathcal{A}_p in (42), and

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

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

$$Y^{\max} = 0.73 \times (\mathcal{A}_p^{\max}) \approx 0.73.$$

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

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

It should be noted that the SBS component was suppressed at I_p^{\max} because of the phase mismatch associated

with the two-photon heating [5]. For $I_p \approx 10^9 \text{ W/cm}^2$ we have $(\gamma_p) \approx 0.1 \text{ cm}^{-1}$ (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 γI_p to the total absorption coefficient at $\lambda = 308 \text{ nm}$ in hexane for the three values of the pump intensity I_p

| $I_p, \text{ W/cm}^2$ | $\gamma_p, \text{ cm}^{-1}$ |
|-----------------------|-----------------------------|
| $\geq 10^{10}$ | ≥ 1.0 |
| 10^9 | ≈ 0.1 |
| 2.5×10^8 | ≈ 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 component is 0.007 cm^{-1} . The spectral resolution of a Fabry-Perot etalon is limited by several MHz (or 10^{-3} 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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