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Numerical modeling of stimulated Raman scattering with selective amplification

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ABSTRACT

This work suggests a mathematical model to simulate Stimulated Raman Scattering (SRS) with selective amplification of specific Stokes lines. This phenomena was observed experimentally when the laser excitation energy (E_L) was above the dissociation energy of the Raman medium (D_R), while the Stokes line showing amplification was in close proximity of an atomic emission line. The proposed numerical model suggests an influence of a stimulated emission factor in addition to the normal SRS behavior. This model was applied to the Raman medium of H_2 gas, with the fourth harmonic of the Nd:YAG laser, 266 nm. At this wavelength, the energy of the laser excitation source is above the dissociation limit of H_2 gas. In addition, the Stokes 3 line is in close proximity of the Balmer H_ε line. A comparison between simulated and experimental results was undertaken which showed good agreement. This concluded that the proposed model, which took stimulated emission into consideration, was a good explanation for the high conversion efficiency of the third Stokes lines as compared to other observed Stokes lines. The model also aided in the understanding of the selective amplification process of specific Stokes lines and the factors that influenced it.

1. Introduction

The discovery of lasers is regarded as the most important discovery of the century. The wide use of lasers in medical applications, industry, communication, science and numerous fields is very apparent. For new applications to be feasible, there is an urgent need for new lasers with multiple wavelengths and high intensity. One of the methods to produce radiations with different wavelengths is to convert wavelength from one region of the electromagnetic spectrum to another through the process of Stimulated Raman Scattering (SRS). SRS is an inelastic scattering of radiation falling on molecules which generates Stokes and anti-Stokes lines that are separated by the vibrational frequency of the molecule.

Among the first work of SRS in gases was that of Minck et al. in 1963 where the pump light used was a ruby laser [1]. SRS spectra excited with frequencies below the dissociation energy of the Raman media was observed by many workers [2–6]. To the contrary, only a few researcher have studied SRS when the exciting laser energy is above the dissociation limit of the Raman medium [7–9]. However, none of these investigations [7–9] have reported any abnormal behavior in the intensity of the observed Stokes lines.

In the work of Gondal and Dastageer [10], the SRS spectrum of H_2 gas as a Raman medium was observed using the Nd:YAG fourth harmonic wavelength of 266 nm (37,594 cm^{-1}). The energy of the exciting photons is higher than the dissociation energy of the H_2 gas (36,118 cm^{-1}). The experimental spectra of the SRS of H_2, when excited by a 266 nm laser wavelength, showed an amplification of the Stokes 3 line. Moreover, they observed an amplification of Stokes 3 line that increased with increasing laser energy. They have reported a conversion efficiency of 36% for the Stokes 3 line when laser energy reached 22 mJ, while no such amplification was observed in any of the other Stokes lines in the same experiment. Even though the work of Tzortzakis et al. [9] have investigated the SRS spectrum of H_2 gas using the fourth harmonic wavelength of 266 nm as did Gondal and Dastageer [10], they have not reported any selective amplification in any of the Stokes lines. The main reason behind this is that Tzortzakis et al. [9] recorded up to Stokes 2 line only, whereas Gondal and Dastageer [10] have extended their range of observation up to Stokes 5 line. Hence, the selective amplification phenomena in the Stokes 3 line was observed in Gondal and Dastageer [10] work and not in that of Tzortzakis et al. [9]. To our knowledge, the work of Gondal and Dastageer [10] is the only example for SRS when laser wavelength is above the

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dissociation energy of the Raman medium where selective amplification of SRS lines was observed.

Numerous numerical models [11–13] were suggested to simulate SRS spectra when the laser excitation energy (E_L) is below the dissociation energy of the Raman medium (D_R). The basics of these models will be discussed in Section 2.1. To our knowledge, no numerical model was derived that simulates SRS at excitation energy above the dissociation limit of the Raman medium. Hence, this paper mainly aims at providing a numerical model to simulate SRS when E_L is greater than D_R. This will require investigating other processes that might be present, such as atomic emission and stimulated emission, which might explain any abnormal behavior. This case will be covered in Section 2.2.

Many workers have confined their investigation of the SRS to hydrogen gas as a Raman medium because it has a maximum frequency shift of (4155 cm^{-1}) and sufficiently high Raman gain [14]. We have chosen to apply our simulation to hydrogen gas for the same reasons. Moreover, experimental results of Gondal and Dastageer [10] were present for analysis and it is the only experimental observation of selective amplification of a Stokes line. A comparison between simulated and experimental results will be undertaken in Section 3. We believe that such simulation will aid in interpreting the experimental results and understanding the physical processes that took place when the exciting laser energy was above the dissociation energy of the Raman media. Moreover, such simulation will assist in determining the conditions for selective amplification of a specific Stokes line.

2. Simulation model

2.1. SRS model ($E_L < D_R$)

Numerous works [11–13] have proposed numerical models to simulate SRS spectra when the laser excitation energy, E_L, is below the dissociation energy of the Raman medium, D_R. The starting point in these models is the wave propagation equation in nonlinear optical media which is given by:

$$\nabla^2 \boldsymbol{\epsilon} - \frac{1}{c^2}\frac{\partial^2 \boldsymbol{\epsilon}}{\partial t^2} = -\delta \tag{1}$$

Here, δ is a radiation source which is defined as:

$$\delta = -\boldsymbol{\mu}_\circ \frac{\partial^2 \mathscr{P}_{NL}}{\partial t^2} \tag{2}$$

where $\mathscr{P}_{NL}$ is a nonlinear component of the polarization density and is given by:

$$\boldsymbol{\mathscr{P}}_{NL} = \varepsilon_\circ X^{(3)} \boldsymbol{\epsilon}^3 \tag{3}$$

Here, $X^{(3)}$ is the third-order nonlinear susceptibility. The field $\epsilon(t)$ is a superposition of four frequencies $\omega_1, \omega_2, \omega_3$ and ω_4, with complex amplitudes. Therefore, the polarization density expands to $8^3 = 512$ terms [15]. By analyzing these terms and reducing them through the use of approximations, a set of coupled differential equation can be reached. These coupled differential equations describe the change in intensities of the exciting laser line, I_0, and that of the Stokes lines, I_{Si}, with the length of the Raman cell (z). These are partly shown below in Eq. (4).

$$\begin{aligned}
\frac{dI_0}{dz} &= -g_0 I_{S1} I_0 - \alpha_0 I_0 \\
\frac{dI_{S1}}{dz} &= g_{S1}[I_{S1}I_0 - I_{S1}I_{S2}] - \alpha_{S1}I_{S1} \\
\frac{dI_{S2}}{dz} &= g_{S2}[I_{S2}I_{S1} - I_{S2}I_{S3}] + g_{42}[I_0 I_{S1}^2] - \alpha_{S2}I_{S2}
\end{aligned} \tag{4}$$

where g_0 is the Raman gain coefficient and g_{Si} are the gain coefficients which controls the generation of the Stokes waves; α_i are additional terms that effect the change in intensity with length due to other losses. The last equation includes a Four Wave Mixing term (FWM) and the FWM gain coefficient, g_{42}. FWM process occurs as a result of mixing one pump wave, of frequency ω_0, with two first Stokes waves, of frequency ω_1, to create a second Stokes wave at frequency $\omega_2 = 2\omega_1 - \omega_0$, where this process seed the SRS conversion [13]. Throughout the above derivation, a plane wave model of the incident radiation was assumed.

For a better description of the experimental beam, a transformation of the beam characteristics from a plane wave approximation to a Gaussian beam is required. Furthermore, a conversion from intensity to power is made to coincide with experimental equipment. In order to achieve this, two ratios $K_0(z) = \frac{I_0(Z)}{I_0(0)}$ and $K_i(z) = \frac{I_i(Z)}{I_0(0)}$ are introduced, which give the laser transmission and Stokes conversion efficiencies, respectively, as a function of a dimensionless parameters Z such that [12]:

$$Z = z g_0 I_0(0),$$

Therefore, by taking $\alpha_0 = \alpha_{S1} = \alpha_{S2}$, Eq. (4) can be transformed to:

$$\begin{aligned}
\frac{dK_0(Z)}{dI_0(0)} &= z * g_0[-K_1(I_0(0))K_0(I_0(0)) - \beta K_0(I_0(0))] \\
\frac{dK_1(Z)}{dI_0(0)} &= z * g_0\left[g_1[K_1(I_0(0))K_0(I_0(0)) - K_1(I_0(0))K_2(I_0(0))] - \beta K_1(I_0(0))\right] \\
\frac{dK_2(Z)}{dI_0(0)} &= z * g_0\Big[g_2[K_2(I_0(0))K_1(I_0(0)) - K_2(I_0(0))K_3(I_0(0))] \\
&\quad + \frac{g_{42}}{g_0} I_0(0)(K_1(I_0(0))K_2(I_0(0))^2) - \beta K_2(I_0(0))\Big]
\end{aligned} \tag{5}$$

where $\beta = \alpha_0 / g_0 I_0(0)$ and $g_i = g_{Si}/g_0$ for $i = 1, 2, \ldots$.

Moreover, the output to input power ratio using a Gaussian distribution is given by [11]:

$$R_j = \frac{P_j(Z)}{P_L(0)} = \frac{1}{I_L(0,0)} \int_0^{I_L(0,0)} K_j[I]dI, \quad j = L, S_i \tag{6}$$

where $P = IA = \int I r dr\, d\theta$, A the area and r is a radial coordinate. L and S_i refer to the laser and Stokes waves, respectively.

The above model was used to simulate the SRS spectra of hydrogen gas at wavelengths that are below dissociation energy [11–13], where it showed good agreement between simulated and experimental results. The proposed model reported in this work builds on the above mentioned model while taking into account other processes that might occur when laser excitation energy is above the dissociation energy of the Raman medium, such as atomic emission and the interaction of this emission with the SRS radiation present in the medium. Namely, the probability of the occurrence of stimulated emission which could selectively amplify some Stokes lines. In the following subsection, the proposed model will be explained.

2.2. SRS model ($E_L > D_R$)

As mentioned previously, Gondal and Dastageer [10] observed an amplification of the Stokes 3 line only when the hydrogen gas

Table 1
Wavelengths of the first five Stokes lines using a 266 nm laser source along with the Blamer series of hydrogen.

	Stokes lines		Atomic lines		
	cm^{-1}	nm	Line name	cm^{-1}	nm
Stokes 1	33,438	299	H_α	15,233	656.3
Stokes 2	29,282	341.4	H_β	20,567	486.1
Stokes 3	<u>25,126</u>	<u>397.8</u>	H_γ	23,036	434
Stokes 4	20,970	476.7	H_δ	24,372	410.2
Stokes 5	16,814	594.5	H_ε	<u>25,182</u>	<u>397</u>