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Original Research Article

Interplay of Raman and Brillouin scattering in semiconductors under short- and long-pulse excitation

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ABSTRACT

A straightforward analytical approach, grounded in the hydrodynamic model of plasmas, has been formulated to examine both steady-state and transient phenomena of stimulated Raman and Brillouin scattering in semiconductors that are either centrosymmetric or weakly non-centrosymmetric. The study provides estimates for gain coefficients, threshold pump intensities, and the optimal pulse durations required to trigger Raman and Brillouin instabilities. The observed trends of transient gain factors show good agreement with experimental results and other theoretical predictions. The analysis also effectively elucidates the interplay and competition between stimulated Raman and Brillouin scattering under both short- and long-pulse duration conditions.

1. Introduction

With the development of lasers, nonlinear optics (NLO) has grown into a diverse and multidisciplinary field, attracting researchers from both fundamental and applied areas. NLO phenomena can generally be classified into two main categories: (i) steady-state effects and (ii) coherent transient optical effects. Many NLO phenomena—such as parametric interactions, stimulated scattering, and their applications in parametric amplifiers, oscillators, and optical phase conjugation—occur under continuous-wave (cw) laser operation or when laser pulse durations are significantly longer than the medium's dephasing or recombination times. These phenomena are typically categorized as steady-state NLO effects. Longer interaction times also make it possible to manipulate light with light, which underlies various all-optical signal processing techniques. In crystals, these effects primarily originate from free-carrier states and the photogeneration of carriers.

On the other hand, coherent transient optical effects, such as optical nutation and free induction decay, have become an important focus in the field of coherent optical spectroscopy. These effects occur when the material's response to an intense laser field is slower than the changes in the light intensity. In other words, if the duration of the pump pulse is much shorter than the dephasing time of the resonant excited state, the medium can temporarily retain the coherence induced by the light even after the pulse has ended.

Among the various nonlinear optical (NLO) phenomena, the nonlinear scattering of laser radiation in gaseous and solid-state plasmas has attracted significant attention in recent years [1–4], primarily because these processes strongly influence

laser energy absorption in plasmas. When a high-intensity light beam interacts with an active medium, the scattered wave can experience strong optical amplification at the Stokes-shifted frequency. Such phase-coherent processes are known as stimulated scattering.

Particularly, the study of stimulated Raman scattering (SRS) and stimulated Brillouin scattering (SBS) in solids has been extensively explored both theoretically and experimentally, due to their technological relevance and their role in understanding light-matter interactions in quantum electronics. One important application of these effects in modern optics is the generation of optical phase conjugation (OPC) in active media [5]. Fundamentally, Raman and Brillouin scatterings are similar: both occur because a nonlinear property of the medium couples a mechanical variable (such as interatomic distances in molecules or density variations in solids or liquids) to the polarization induced by an electromagnetic wave. In these processes, the electromagnetic wave frequency is altered as energy is exchanged with the medium, generating phonons—optical phonons in SRS and acoustic phonons in SBS.

Despite these similarities, SRS and SBS are often analyzed separately, since the second-order forces driving them differ: SRS arises from finite differential polarizability, whereas SBS originates from electrostrictive strain in the medium. Quantitative studies indicate that the steady-state gain for SBS is generally an order of magnitude higher than that for SRS. Consequently, SBS often dominates over SRS in many materials, especially for longer laser pulses, where SRS is suppressed and a significant fraction of the laser energy is



converted into Brillouin-shifted light. Previous studies have shown that achieving the full steady-state gain for SBS requires a duration comparable to the phonon lifetime; for shorter interaction times, SBS generation is limited, allowing SRS to develop more freely.

Recent independent investigations of SRS and SBS under steady-state conditions, employing the hydrodynamic model and methodology proposed by Singh et al. [6], have been carried out in Ref. [7, 8].

Until now, the focus of most studies has been on steady-state solutions [7–9], with researchers typically assuming monochromatic waves that grow spatially. Similar methods can be applied to analyze pure temporal growth as well. An example of a transient nonlinear effect is the temporal deformation of a laser pulse as it propagates through a medium whose refractive index depends on intensity [10]. The investigation of such transient coherent optical effects has become a significant area of research within the field of coherent optical spectroscopy [11].

The widespread use of semiconductors in electronics and the need to understand their fundamental properties have motivated many solid-state physicists to explore their roles in areas such as spectroscopy, lasers, and device fabrication. In the pursuit of optical memories and switching devices, it was observed that the optical characteristics of semiconductors change significantly when electrons are optically excited. Semiconductors exhibit electrical properties that fall between those of metals, with nearly free electrons, and insulators, with tightly bound electrons. This intermediate behavior makes them particularly suitable as nonlinear devices in both electronics and optics, since their properties can be easily modified by external fields, chemical composition, or microstructuring. Consequently, semiconductors have become indispensable as active media in laser communications, modern optoelectronic devices, optical computing [12–14], and all-optical signal processing [15]. Therefore, understanding the mechanisms underlying transient effects in these crystals is of fundamental importance.

Furthermore, contrary to the large steady-state Raman gain predicted experimentally, it has been observed that this gain is almost completely diminished, while Brillouin gain reaches a significant value when the pulse duration of the incident laser radiation exceeds $\tau_p > 10^{-9}$ s [16]. The propagation of pulses through a resonant medium can give rise to a variety of intriguing phenomena. Transient SRS, for instance, has been employed to investigate the relaxation dynamics of material excitations in the picosecond regime using mode-locked laser pulses [16,17]. Since pulsed lasers are commonly used in stimulated scattering experiments, it becomes essential to account for the temporal behavior of the output, i.e., to consider transient solutions of the SRS and SBS processes in crystals subjected to pulsed laser irradiation.

In solids, particularly semiconductors, transient coherent optical effects hold promise for a variety of applications, ranging from optical computing and information storage to the investigation of hot-carrier dynamics and intraband carrier-carrier scattering processes [14]. These coherent transient phenomena typically occur on sub-picosecond to picosecond timescales and remained largely inaccessible in the past due to the lack of laser sources with sufficiently short pulse durations. With the advent of experimentally available short-pulse lasers capable of generating sub-picosecond to femtosecond pulses [15], it is now possible to perform time-resolved measurements of coherent transient effects—such as optical nutation, free

induction decay, and self-induced transparency—in semiconductors with very high temporal and spectral resolution [16].

To the best of the authors' knowledge, a straightforward analytical approach based on the hydrodynamic model of plasmas has not yet been developed to study transient stimulated Raman and Brillouin scattering processes, or the competition between them, in centrosymmetric or weakly noncentrosymmetric semiconductors. The present investigation is motivated by this gap in the existing literature.

In this work, the authors present analytical results on stimulated Raman (SRS) and Brillouin (SBS) scattering of a hybrid pump wave propagating obliquely in a magnetoactive semiconductor plasma that is either centrosymmetric or weakly noncentrosymmetric. The analysis is based on the premise that the nonlinear optical behavior of the medium originates from its third-order nonlinear optical susceptibility. By employing the coupled-mode theory of plasmas, steady-state gain coefficients for both SRS and SBS are derived analytically. Transient Raman and Brillouin gains are subsequently obtained from the corresponding steady-state values. Additionally, numerical estimates of threshold pump intensities and optimal pulse durations have been calculated using parameters suitable for a centrosymmetric or weakly noncentrosymmetric semiconductor crystal irradiated by a frequency-doubled 10.6 μm CO₂ laser, thereby validating the present theoretical approach.

2. Theoretical formulation

This section presents the theoretical framework for calculating transient Raman and Brillouin gains using the corresponding steady-state gain constants. The analysis is based on a hydrodynamic model of a homogeneous n-type semiconductor plasma, where electrons act as the charge carriers and the system is assumed to be in thermal equilibrium. To investigate the competition between stimulated Raman scattering (SRS) and stimulated Brillouin scattering (SBS), let us consider the propagation of a hybrid pump wave

$$\vec{E}_0 = [E_{0x}\hat{x} + E_{0y}\hat{y}] \exp[i(k_0x - \omega_0t)] \quad (1)$$

in a homogeneous semiconductor plasma subjected to an external magnetostatic field \vec{B}_s oriented at an angle θ with respect to the x -axis (using the same field configuration as described in Ref. [7] and [8]).

The authors have selected this particular field configuration because most previous studies have considered the pump wave propagating exactly parallel to the applied magnetic field, which may not be experimentally practical. In addition, earlier analyses often assume that the pump's electric field is either entirely parallel or perpendicular to the propagation direction, a simplification that does not generally reflect realistic conditions [18]. In a finite solid-state plasma, the pump field E_0 typically has components both parallel and perpendicular to the propagation direction. Therefore, the most realistic scenario, adopted in the present study, is a hybrid mode propagating obliquely with respect to the external magnetic field.

The coupled-mode approach is employed to derive simplified expressions for both steady-state and transient gains. For the same field configuration, the authors have previously reported the analytical expressions for the steady-state gain

coefficients of stimulated Brillouin (g_B) and stimulated Raman (g_R) scattering in Ref. [7] and [8], respectively.

Hence, based on equation (21) from Ref. [7], the steady-state Brillouin gain coefficient (g_B) can be expressed as:

$$g_B = \frac{-\gamma^2 k_s}{4\rho\epsilon_0\epsilon_1\omega_a\Gamma_a} \left[\frac{(k_0 - k_a)}{k_a} + \frac{\bar{\omega}_p^2}{\omega_0\omega_s} \right] |k_a(E_e)_x|^2. \quad (2)$$

Here, $\vec{E}_e = \vec{E}_0 + (\vec{v}_0 \times \vec{B}_s)$ in which \vec{v}_0 and \vec{B}_s denote the oscillatory velocity of the electrons and the static magnetic field, respectively. In this expression, we define

$$\bar{\omega}_p^2 = \frac{\omega_p^2(\omega_{cx}^2 + v^2)}{(\omega_c^2 + v^2)}.$$

in which ω_{cx} represents the x -component of the cyclotron frequency ω_c , and ω_p is the electron plasma frequency of the medium. Since the focus is on the first Stokes component of the scattered electromagnetic wave, the phase-matching conditions used in deriving equation (2) are $\omega_s = \omega_0 - \omega_a$ and $\vec{k}_s = \vec{k}_0 - \vec{k}_a$. In this context, (ω_a, \vec{k}_a) and (ω_s, \vec{k}_s) correspond to the generated acoustic mode and the Stokes electromagnetic mode, respectively.

The wave numbers k_0 and k_a have approximate magnitudes of 10^5 m and 10^7 m, respectively, making backward scattering apparent. Γ_a represents the phenomenological damping parameter, while ρ , ϵ_0 and ϵ_1 denote the mass density, absolute permittivity, and relative dielectric constant of the crystal, respectively. γ is the electrostrictive coefficient of the medium.

Similarly, based on equations (17) and (24) from Ref. [8], the steady-state Raman gain coefficient (g_R) can be expressed as:

$$g_R = \frac{\epsilon_0 e^2 v \omega_0 \omega_s k (k_0 - k) Z |(E_e)_x|^2}{\eta c \omega_0^2 m^2}. \quad (3)$$

$$\text{Here } Z = \frac{(\delta_2^4 + \omega^2 \Gamma^2) - \omega_0^2 \omega m^2 \Gamma (\delta_1^2 + v^2 \omega_s^2) \left(\frac{\epsilon N}{2M} \right) \left(\frac{\partial \alpha}{\partial u} \right)_0^2}{(\delta_1^2 \delta_2^2 - v \omega \omega_s \Gamma)^2 + (v \omega_s \delta_2^2 + \Gamma \omega \delta_1^2)^2},$$

$$\delta_1^2 = (\bar{\omega}_R^2 - \omega_s^2), \quad \delta_2^2 = (\bar{\omega}_R^2 - \omega_0^2),$$

$$\bar{\omega}_R^2 = \omega_R^2 \left(\frac{\omega_{cx}^2 + v^2}{\omega_c^2 + v^2} \right), \quad \omega_R^2 = \frac{\omega_p^2 \omega_L^2}{\omega_T^2}, \quad \frac{\omega_L}{\omega_T} = \sqrt{\frac{\epsilon_L}{\epsilon_\infty}},$$

in which ϵ_L and ϵ_∞ are the lattice dielectric constant and the high-frequency permittivity of the medium. ω_L and ω_T are the longitudinal and transverse optical phonon frequencies respectively. η , v and Γ are the background refractive index, collision frequency and phenomenological damping constant of the medium.

In deriving equation (3), it is assumed that the Raman-active medium consists of N harmonic oscillators per unit volume, each of mass M , oscillating at their natural vibrational frequencies. These oscillators are driven by the optical electric field due to the nonzero differential polarizability $(\partial \alpha / \partial u)_0$.

The above expressions for the steady-state gain coefficients can be reformulated in terms of the input pump intensity for a weakly noncentrosymmetric semiconductor plasma, such as n-InSb, as follows:

$$g_R = 2.07 \times 10^{-8} I_{in} \quad (4)$$

and

$$g_B = 1.55 \times 10^{-4} I_{in}. \quad (5)$$

Here, $I_{in} = 0.5 \eta \epsilon_0 c |E_e|^2$ is expressed in W m^{-2} , while the gain coefficients are in m^{-1} . The relevant physical parameters are taken from Ref. [7] and [8].

From equations (4) and (5), it is evident that only a high-power laser pump can produce significant gain coefficients for both SBS and SRS. Therefore, the laser source should either be pulsed with a duration on the order of 10^{-9} s for Q-switched lasers or consist of a pulse train with individual pulses of about 10^{-12} s for mode-locked lasers. Since these pulse durations are comparable to or shorter than the phonon lifetimes (acoustic phonons $> 10^{-9}$ s and optical phonons $> 10^{-12}$ s) [18], the investigation of transient effects becomes essential.

However, the previous steady-state formulations are limited, as they cannot accurately predict the threshold pump intensity (I_{pth}) required for the onset of SRS or SBS with positive gains, nor can they determine the optimum pulse durations for observing these instabilities. This indicates that SRS and SBS must be analyzed by including transient effects. In general, following Wang [18], the transient gain factors can be related to the corresponding steady-state gain coefficients through the following relation:

$$g_{TR,B} = (2g_{R,B}x\Gamma_{R,B}\tau_p)^{1/2} - \Gamma_{R,B}\tau_p. \quad (6)$$

Here, $\Gamma_R(\Gamma_B)$ represents the damping constant of the Raman or Brillouin wave, x is the interaction length, and τ_p denotes the pulse duration.

For backward scattering, the effective interaction length is very short because the Stokes pulse and the laser pulse travel in opposite directions, limiting their overlap to the length of the laser pulse. For picosecond pump pulses, this overlap region is typically only a fraction of a millimeter.

Therefore, according to Wang [19], for extremely short pulse durations ($\tau_p < 10^{-10}$ s), the interaction length should be taken as $(c_1 \tau_p / 2)$, where c_1 is the speed of light within the crystal lattice.

Consequently, by setting $g_{TR,B} = 0$ in the above expression, the threshold pump intensity for the onset of SRS and SBS can be determined as:

$$(I_{pth})_{R,B} = \frac{\Gamma_{R,B}}{2G_{R,B}c_1}, \quad (7)$$

where $G_{R,B} = g_{R,B} / I_{in}$ represents the gain per unit of pump intensity.

However, for relatively long pulse durations ($\tau_p > 10^{-9}$ s), the interaction length can be approximated by x , and under these conditions, one obtains

$$g_{TR,B} = (\Gamma_{R,B}\tau_p)^{1/2} [-(\Gamma_{R,B}\tau_p)^{1/2} + (g_{R,B}x)^{1/2}]. \quad (8)$$

Using the above expression, the optimum pulse duration, $(\tau_p)_{opt}$, beyond which no gain is obtained, can be determined by setting $g_{TR,B} = 0$, giving

$$(\tau_{p,opt})_{R,B} \approx \frac{g_{R,B}}{\Gamma_{R,B}}. \quad (9)$$

3. Results and discussion

To validate the present model and examine the competition between SRS and SBS, a weakly noncentrosymmetric crystal (n-InSb) is chosen as the medium, which is assumed to be irradiated by a pump wave of frequency $\omega_0 = 1.78 \times 10^{14} \text{ s}^{-1}$. The physical parameters used are as follows:

$$\begin{aligned} m &= 0.015 m_0 \text{ being the rest mass of an electron,} \\ \rho &= 5.8 \times 10^3 \text{ kgm}^{-3}, \varepsilon_L = 17.8, \varepsilon_\infty = 15.68, \\ v &= 3.5 \times 10^{11} \text{ s}^{-1}, n_e = 10^{23} \text{ m}^{-3}, N = 1.48 \times 10^{28} \text{ m}^{-3}, \\ \omega_T &= 3.7 \times 10^{13} \text{ s}^{-1}, (\partial\alpha/\partial u)_0 = 1.68 \times 10^{-16} \text{ mks units,} \\ M &= 236.47, \gamma = 5 \times 10^{-10} \text{ Fm}^{-1} \text{ and } \Gamma_a = 2 \times 10^{10} \text{ s}^{-1}. \end{aligned}$$

Equations (4) and (5) can be employed to estimate the ratio of the steady-state gain coefficients for SRS and SBS, (g_R/g_B) , in terms of the material parameters. Therefore, for a given pump intensity I_{in} , we have

$$(g_R/g_B) = 1.36 \times 10^{-4}. \quad (10)$$

Equation (10), which provides the ratio of the steady-state gain coefficients for SRS and SBS under the same pump intensity, shows that SBS has a significantly higher steady-state gain than SRS, by a factor on the order of ω_T/ω_a , where ω_T is the transverse optical-phonon frequency and ω_a is the acoustic-phonon frequency. This finding is consistent with results obtained using semiclassical theory by Sen and Sen [20] and with the coupled-mode approach by Singh et al. [9].

The influence of the magnetostatic field B_s on the Raman and Brillouin gains can be analyzed using equations (2) and (3), and for the chosen parameters, the results are as follows:

$$\frac{(g_B)_{B_s=0}}{(g_B)_{B_s=0}} = 4.9 \times 10^2 \quad (11a)$$

and

$$\frac{(g_R)_{B_s=0}}{(g_R)_{B_s=0}} = 10^2 \quad (11b)$$

at $k = 10^{-7} \text{ m}^{-1}$, $E_e = 10^7 \text{ Vm}^{-1}$ with $\omega_c = 0.9\omega_0$ which is inclined at an angle $\theta = 45^\circ$.

From equations (11), it is observed that the magnetic field enhances both Raman and Brillouin gains. Using $\Gamma_R = 3.7 \times 10^{11} \text{ s}^{-1}$, $\Gamma_B = 2 \times 10^{10} \text{ s}^{-1}$ and equations (4)–(7), the threshold pump intensities for the onset of Raman and Brillouin instabilities are calculated as $1.26 \times 10^{11} \text{ Wm}^{-2}$ and 10^6 Wm^{-2} , respectively. The presence of the magnetic field lowers the threshold intensity required for both processes. These results further confirm the well-known fact that Brillouin instability occurs at significantly lower pump intensities compared to Raman instability.

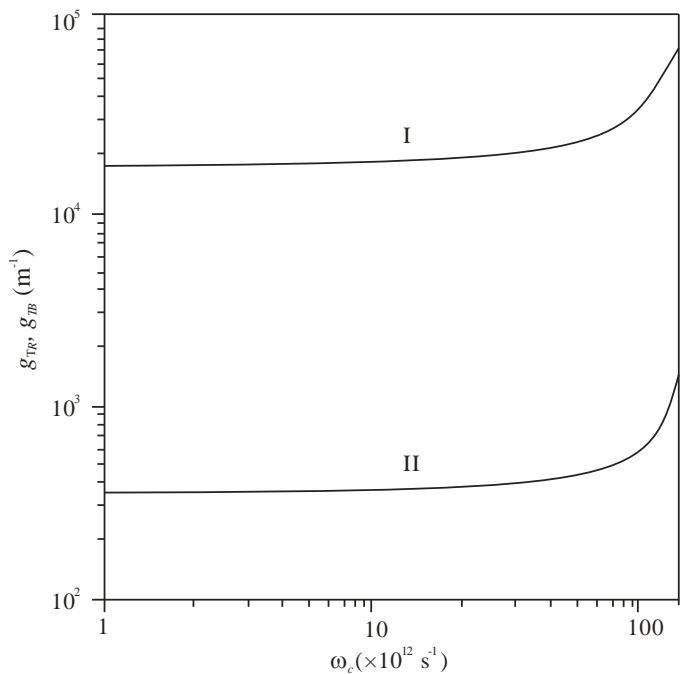


Figure 1: Transient gain factors for Brillouin (g_{TB} , curve I) and Raman (g_{TR} , curve II) scattering as functions of the magnetic field, expressed in terms of the cyclotron frequency ω_c .

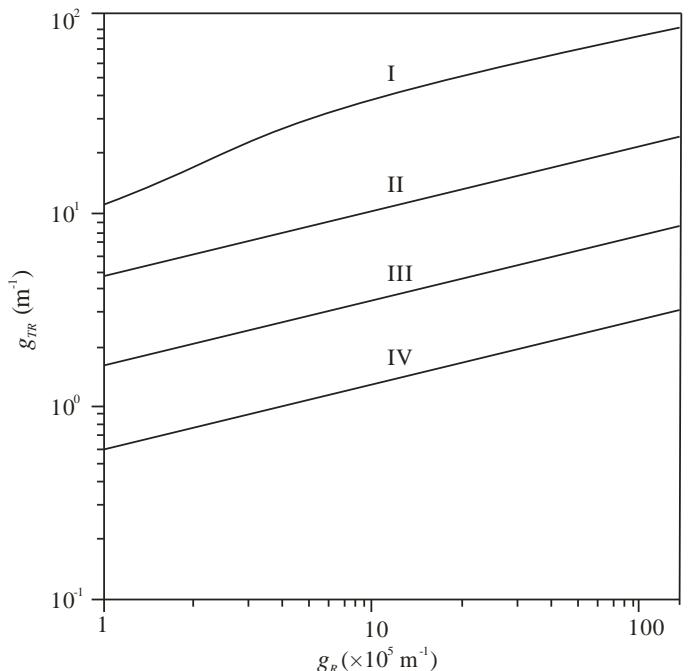


Figure 2: Transient Raman gain (g_{TR}) versus steady-state Raman gain (g_R) for different pulse durations. Curves I, II, III, and IV correspond to $\tau_p = 10, 1, 0.1$, and 0.01 , respectively.

Using equation (9), the optimum pulse duration, $(\tau_p)_{opt}$, for a weakly noncentrosymmetric crystal (n-InSb) can be estimated numerically by applying the previously calculated values of $g_{R,B}$, with $x = 10^{-4} \text{ m}$, as follows:

$$(\tau_{p,opt})_R = 5.6 \times 10^{-20} I_{in} \text{ s} \quad (12)$$

and

$$(\tau_{p,opt})_B = 7.7 \times 10^{-15} I_{in} \text{ s.} \quad (13)$$

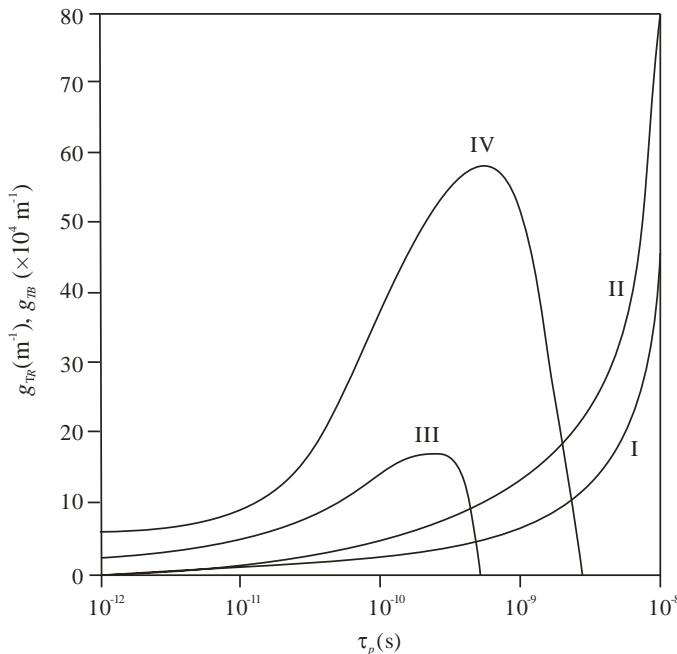


Figure 3: Transient gain factors (g_{TR} and g_{TB}) as functions of the pump pulse duration τ_p , with pump intensity I_{in} as a parameter. Curves I and II represent Brillouin gain factors, while curves III and IV represent Raman gain factors. Curves I and III correspond to $I_{in} = 1.2 \times 10^{13} \text{ W m}^{-2}$, and curves II and IV correspond to $I_{in} = 5 \times 10^{13} \text{ W m}^{-2}$.

The calculated values of $\tau_{p,opt}$ indicate that the optimum pulse duration increases with higher pump intensity. Equation (12) accounts for the diminishing Raman gain at longer pulse durations. The next focus is on the behavior of the transient gain factors, $g_{TR,B}$, as functions of various physical parameters.

Figure 1 illustrates the qualitative dependence of the transient Raman (g_{TR}) and Brillouin (g_{TB}) gain factors on the externally applied magnetostatic field, expressed in terms of the cyclotron frequency, ω_c . In this figure, curve I corresponds to g_{TB} , while curve II represents g_{TR} . Both g_{TR} and g_{TB} are relatively insensitive to ω_c at low values of the magnetostatic field, B_s . However, as ω_c approaches the molecular vibrational or acoustic wave frequency, the respective transient gain factors increase sharply. Consequently, the maximum $g_{TR,B}$ occurs when ω_c is comparable to the pump frequency, ω_0 , which is independent of the magnetic field. This implies that stimulated laser output can be tuned by adjusting the magnetic field (cyclotron frequency) around the phonon vibrational frequency, consistent with earlier observations on steady-state gain factors by Singh et al. [9].

In configurations with a transversely applied magnetic field [21], it has been reported that the magnetic field only significantly influences the system when ω_c exceeds the vibrational phonon frequency of the medium. Figure 1 also indicates that SBS exhibits a higher transient gain than the competing SRS process. This conclusion aligns with the studies of Maier, Wendt, and Kaiser [22], who compared both steady-state and transient responses of these stimulated scattering processes.

Figure 2 illustrates the variation of the transient Raman gain, g_{TR} , with the steady-state Raman gain, g_R , for different pulse durations. For a given value of g_R , g_{TR} increases as the pulse duration becomes longer. At $\tau_p = 10^{-2} / \Gamma_R$, for small values of g_R , the transient gain g_{TR} is less than 1. For all other pulse durations, g_{TR} remains greater than 1. The qualitative trend shown in this figure is consistent with the experimental findings of Carman et al. [23].

Figure 3 illustrates the qualitative behavior of the transient gain factors g_{TR} and g_{TB} as functions of the pump pulse duration, τ_p , with the pump intensity, I_{in} , treated as a parameter. Pulse durations in the range $10^{-12} < \tau_p < 10^{-8}$ s have been considered. For SRS, the interaction length is taken as the cell length x , whereas for backward SBS, it is the shorter of $c_1 \tau_p / 2$ or x .

For a fixed I_{in} , g_{TR} increases with increasing pulse duration, reaching a maximum at a certain τ_p . This maximum persists over a specific range of τ_p , representing quasi-steady-state or quasi-saturation regimes. Curves III and IV indicate that increasing I_{in} shifts the gain saturation regime to higher values of τ_p . If τ_p exceeds the quasi-saturation regime while I_{in} is kept constant, g_{TR} drops sharply. This trend closely resembles experimental observations in CS_2 [24], where Raman conversion efficiency, proportional to the exponential of the gain factor, decreases significantly at longer pulse durations.

Curves I and II show the transient Brillouin gain factor, g_{TB} , as a function of τ_p . For a fixed I_{in} , g_{TB} is negligible at very short pulse durations ($\tau_p < 0.1$ ns) but rises sharply as τ_p increases. For n-InSb, g_{TB} is found to dominate over g_{TR} across all considered pulse durations.

4. Conclusions

The present theoretical framework provides a satisfactory explanation of the competition between the two transient stimulated scattering processes. A key feature of this approach is that both SRS and SBS, in steady-state and transient regimes, can be analyzed in centrosymmetric or weakly non-centrosymmetric dielectric materials using a straightforward classical treatment.

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