Stimulated scattering in magnetoactive semiconductors

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An analytical investigation of the stimulated Raman (SRS) and Brillouin (SBS) scatterings of the Stokes mode is undertaken in centrosymmetric semiconductors with a dc magnetic field applied along the direction of the uniform pump field. The threshold field for inciting SRS and SBS and the corresponding gain constants have also been identified. The magnetic field is found to reduce the threshold field and subsequently augment the gain constant of the scattered mode. The magnitude of the third-order nonlinear optical susceptibility determined from our analysis is found to agree with the theoretically and experimentally quoted values. The growth rates of the Raman and the Brillouin modes are found to be higher in the heavily doped regime. The SBS gain constant is found to be higher than SRS and its ratio is found to be equal to the ratio of the optical to acoustical wave frequency. The SBS and SRS gain constants are found to be effectively influenced by operating the magnetic field such that the cyclotron frequency becomes comparable to the acoustical (for SBS) or optical (for SRS) wave frequency.

I. INTRODUCTION

There has of late been a surge of interest in experimental and theoretical investigations of nonlinear optical susceptibilities in different materials in view of their potential uses in modern optoelectronic devices such as parametric amplifiers and oscillators, optical switches, phase conjugate mirrors, and, above all, in the development of newer coherent laser sources over a wide frequency spectrum. In centrosymmetric materials, the dominant nonlinear optical processes may be characterized by the third-order nonlinear optical susceptibility $\chi^{(3)}$. It is this nonlinear crystal property that has been extensively exploited for tuning lasers over the infrared spectrum,[1] optical bistability, phase conjugation,[2] etc.

In a crystalline medium, stimulated Raman (SRS) and Brillouin (SBS) scatterings are very closely related phenomena stemming from parametric interaction between light waves and material excitations. These phenomena are usually studied simultaneously considering that the second-order forces responsible for them are different, viz., the finite differential polarizability gives rise to SRS, while the electrostrictive strain produces SBS in the material.[3]

Recently, attention has been focused on the theoretical and experimental investigations of stimulated emission and resonant amplification of far-infrared radiation in the presence of electric and magnetic fields.[4,5] In the present paper, we have attempted to investigate the stimulated Raman and Brillouin Stokes scattering in a narrow-band-gap n-type semiconductor crystal duly irradiated by a uniform pump wave in the presence of a dc magnetic field applied along the pump wave. The nonlinearities taken into account in the present analysis are the electronic nonlinearity due to the nonlinear current density and the nonlinear polarization which is the cause of the nonlinear coupling between the resulting acoustical-optical mode and the scattered electromagnetic wave.

II. THEORETICAL FORMULATION OF NONLINEAR CURRENT DENSITY

We consider a homogeneous medium with electrons as carriers being subjected to an external magnetostatic field $\mathbf{B}_0$ (along the x axis), parallel to the propagation vector $\mathbf{k}$ and the spatially uniform pump wave $\mathbf{E}_0\exp(-i\omega_0 t)$. The intense laser excitation is responsible for the generation of a material excitation within the crystal which then leads to the stimulated scattering process; the molecular vibrations are responsible for SRS and the acoustic mode for SBS. Due to the electromagnetic field, the ions within the lattice move into nonsymmetrical position, usually producing a contraction in the direction of the field and an expansion across it. The electrostatic force thus produced is the origin of the differential polarizability and electrostriction in the medium. The polarizations due to molecular vibrations and the electrostriction are given by

$$\mathbf{P}_{mv}=\epsilon_0 N(\partial\alpha/\partial u)_0 u(r,t)^*\mathbf{E}_0 , \tag{1}$$

$$\mathbf{P}_{es}=-\gamma\mathbf{E}_0\frac{\partial u(r,t)^*}{\partial r} , \tag{2}$$

where $u(r,t)$ denotes the relative displacement of oscillators from the mean position of the lattice. Equation (1) shows that due to the nonvanishing differential polarizability $(\partial\alpha/\partial u)_0$ in the Raman active medium, the optical field drives the molecular vibrations, which in turn modulate the dielectric constant or the macroscopic susceptibility of the medium, resulting in a nonlinear induced polarization $\mathbf{P}_{mv}$. Similarly, in a Brillouin active centrosymmetric medium, an acoustic mode is generated due to the electrostrictive strain, leading to the energy exchange between electromagnetic and acoustic fields, and gives rise to $\mathbf{P}_{es}$ [Eq. (2)].

Now, applying the procedure adopted in our earlier work[5] to the present field configuration, the Stokes mode of this scattered component at $\omega_s=\omega_0-\omega$ can be ob-

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tained from (9) as

$$n_s^* = \frac{-\omega_M^2 P_{nl}}{e|\bar{E}|} \times \left[1 - \frac{(\omega_s^2-\omega_M^2-i\omega_s\nu)(\omega^2-\omega_M^2+i\omega\nu)}{k^2|\bar{E}|^2}\right]^{-1}, \quad (3)$$

with $\bar{\omega}=\omega-kv_0$,

$$\bar{E}=(eE_0/m)\ ,$$

$$\omega_M^2 \equiv \omega_R^2 \left[1-\frac{\omega_P^2\bar{\omega}\varepsilon_L}{\varepsilon_1\omega_c(k^2c_L^2-\omega^2)}\right],$$

$$\omega_R^2 = \left[\frac{\omega_P^2\bar{\omega}\omega_c\varepsilon_L}{\varepsilon_1\omega(\bar{\omega}+i\nu-\omega_c)}\right],$$

$$\omega_P = \left[\frac{n_0 e^2}{m\varepsilon_0\varepsilon_L}\right]^{1/2}$$

is the plasma frequency, and P_{nl} stands for the induced nonlinear polarization.

The resonant Stokes component of the induced current density due to the finite nonlinear polarization of the medium has been deduced neglecting the transition dipole moment which can be represented as

$$J(\omega_s)=\frac{i\varepsilon\omega_P^2\bar{\omega}}{\omega(\bar{\omega}+i\nu-\omega_c)} - \frac{\omega_M^2 P_{nl}}{(\nu-i\omega_0)} \times \left[1+\frac{(\omega_s^2-\omega_M^2-i\omega_s\nu)(\omega^2-\omega_M^2+i\omega\nu)}{k^2|\bar{E}|^2}\right]. \quad (4)$$

The first part of $J(\omega_s)$ represents the linear component of the induced current density and is strongly influenced by the external magnetic field, while the latter term represents the nonlinear current density $J_{nl}(\omega_s)$.

III. STIMULATED RAMAN SCATTERING AND RAMAN GAIN CONSTANT

In the present analysis the Raman active medium is taken as consisting of N independent harmonic oscillators per unit volume, each of these being characterized by its position x and normal vibrational coordinates $u(x,t)$. In a Raman active medium the scattering of the incident pump wave is enhanced due to the excitation of a molecular vibrational mode. In case of SRS, the vibrational frequency is taken to be equal to the transverse optical-phonon frequency, i.e., $\omega_v \approx \omega_T$ and is very large in comparison with $k(C/\rho)^{1/2}$. As $(C/\rho)^{1/2}$ is the acoustic velocity in the crystal, we have the equation of motion of an independent oscillator as

$$\frac{\partial^2 u(x,t)}{\partial t^2}+\omega_T^2 u(x,t)+2\Gamma_v\frac{\partial u(x,t)}{\partial t} = \frac{1}{2M}\varepsilon_\infty(\partial\alpha/\partial u)_0 E_0 E_1^* \ . \quad (5)$$

Treating the induced nonlinear polarization P_{cd} as the time integral of the nonlinear current density J_{nl}, we obtain the nonlinear Raman polarization due to the finite perturbed current density from (4) and (5) as

$$[P_r]_{cd}(\omega_s)=\frac{\omega_{Mr}^2\varepsilon_0\varepsilon_\infty N(\partial\alpha/\partial u)_0^2 E_0^2 E_1}{2\omega_0\omega_s M(\omega_T^2-\omega^2+i\omega\Gamma_u)}\left[1+\frac{(\omega_s^2-\omega_{Mr}^2-i\omega_s\nu)(\omega_T^2-\omega_{Mr}^2+i\omega_T\nu)}{k^2|\bar{E}|^2}\right], \quad (6)$$

where

$$\omega_{Mr}^2=\omega_{Rr}^2\left[1-\frac{\varepsilon_L\omega_P^2\bar{\omega}_v}{\varepsilon_\infty\omega_c(k^2c_L^2-\omega_v^2)}\right],$$

$$\omega_{Rr}^2=\left[\frac{\varepsilon_L\omega_P^2\bar{\omega}_v\omega_c}{\varepsilon_\infty\omega_v(\bar{\omega}_v+i\nu-\omega_c)}\right].$$

The nonlinear polarization induced into the system due to the finite molecular vibrations can be obtained from (1) and (5):

$$[P_r]_{mv}(\omega_s)=\frac{\varepsilon_0\varepsilon_\infty N(\partial\alpha/\partial u)_0^2 E_0^2 E_1}{(\omega_T^2-\omega^2+i\omega\Gamma_u)} \ . \quad (7)$$

Following Neogi and Ghosh,[5] the third-order Raman susceptibility is obtained from (6) and (7) as

$$[\chi_r^{(3)}]_{\text{eff}}=\frac{\varepsilon_\infty N(\partial\alpha/\partial u)_0^2}{2M(\omega_T^2-\omega^2+i\omega\Gamma_u)}\left[1+\frac{\omega_{Mr}^2}{\omega_0\omega_s}\right], \quad (8)$$

with

$$[\chi_r^{(3)}]_{\text{eff}}=[\chi_r^{(3)}]_{cd}+[\chi_r^{(3)}]_{mv} \ .$$

Further, from the imaginary part of (8) one obtains the Raman gain constant $g_r(\omega_s)$ as

$$[g_r(\omega_s)]=\frac{k\varepsilon_\infty N(\partial\alpha/\partial u)_0^2\Gamma_v|E_0|^2}{16\varepsilon_L M\omega_T[\{\omega_T-(\omega-\omega_s)\}^2+\Gamma_v^2/4]} \times \left[1+\frac{\omega_{Mr}^2}{\omega_0\omega_s}\right]. \quad (9)$$

Thus, the Raman gain is found to be strongly dependent on the pump frequency through ω_0 and ω_s. It is also influenced by the cyclotron frequency ω_c and the carrier concentration of the medium through the term ω_{Mr}. The SRS gain constant is found to be identical with Eqs. (18.4)–(23) of the formulation by Yariv[6] by setting $\omega_c=0$ in (9). It can also be inferred that the presence of the external magnetic field makes the magnitude of the carrier concentration component ω_{Mr} significant, yielding a favorable gain constant of the Stokes mode. Thus the external dc magnetic field can be utilized to achieve a tunable laser source at frequency $\omega_s=\omega_0-\omega_v$ in a Raman active medium.

However, in order to incite SRS, the input field should be pumped above a certain threshold field, obtained by setting $[P_r]_{cd}(\omega_s)=0$. This yields

$$|E_{0\text{th}}|_r=\frac{m}{ek}[(\omega_s^2-\omega_{Mr}^2-i\omega_s\nu)^{1/2}\times(\omega_{Mr}^2-\omega_T^2+i\omega_T\nu)^{1/2}] . \tag{10}$$

It can be observed from (10) that $|E_{0\text{th}}|_r$ is much lower in the lightly doped regime as compared to the heavily doped one.

IV. STIMULATED BRILLOUIN SCATTERING AND BRILLOUIN GAIN CONSTANT

The phenomenon of SBS is analogous to SRS with the acoustic waves due to electrostriction playing the role of the molecular vibrations. In a Brillouin active medium it is found that $\omega_v=\omega_\alpha$ and $\omega_T=k_v=0$, since such a mode does not exist for acoustic waves. Hence, the equation of motion for $u(x,t)$ in the acoustical branch becomes

$$\frac{\partial^2 u(x,t)}{\partial t^2}-\frac{C_\alpha}{\rho}\frac{\partial^2 u(x,t)}{\partial x^2}+2\Gamma_\alpha\frac{\partial u(x,t)}{\partial t}\doteq\frac{\gamma}{2\rho}\frac{\partial}{\partial x}(E_0E_1^*) , \tag{11}$$

with γ being the phenomenological electrostrictive coefficient ($\approx 10^{-11}$ mks units).[6]

We proceed in an almost similar way as followed in Sec. III to study SBS and the Brillouin gain constant of the Stokes mode. The nonlinear Brillouin polarization induced by a finite nonlinear current density is

$$[P_b]_{cd}(\omega_s)=\frac{-\omega_{Mb}^2\gamma^2k^2|E_0|^2E_1}{2\omega_0\omega_s\rho(\omega_\alpha^2-k^2v_s^2-2i\omega_\alpha\Gamma_\alpha)}\times\left[1+\frac{(\omega_s^2-\omega_{Mb}^2-i\omega_s\nu)(\omega_\alpha^2-\omega_{Mb}^2+i\omega_\alpha\nu)}{k^2|\bar{E}|^2}\right] , \tag{12}$$

where

$$\omega_{Mb}^2=\omega_{Rb}^2\left[1-\frac{\omega_P^2\bar{\omega}_\alpha}{\omega_c(k^2c_L^2-\omega_\alpha^2)}\right] ,$$

$$\omega_{Rb}^2=\left[\frac{\omega_P^2\bar{\omega}_\alpha\omega_c}{\omega_\alpha(\bar{\omega}_\alpha+i\nu-\omega_c)}\right] .$$

The electrostrictive strain interacts with the pump wave in the Brillouin active medium, giving rise to a electrostrictive polarization $[P_b]_{es}$ analogous to polarization due to molecular vibrations in SRS. $[P_b]_{es}$ is obtained from (2) and (11) as

$$[P_b]_{es}(\omega_s)=\frac{-\gamma^2k^2|E_0|^2E_1}{2\rho(\omega_\alpha^2-k^2v_s^2-2i\Gamma_\alpha\omega_\alpha)} . \tag{13}$$

Thus, the total third-order Brillouin susceptibility is obtained as

$$[\chi_b^{(3)}]_{\text{eff}}=\frac{-\gamma^2k^2}{2\rho\varepsilon_0(\omega_\alpha^2-k^2v_s^2-2i\omega_\alpha\Gamma_\alpha)}\left[1+\frac{\omega_{Mb}^2}{\omega_0\omega_s}\right] , \tag{14}$$

where

$$[\chi_b^{(3)}]_{\text{eff}}=[\chi_b^{(3)}]_{cd}+[\chi_b^{(3)}]_{es} .$$

The steady-state Brillouin gain constant $g_b(\omega_s)$ has been obtained as

$$[g_b(\omega_s)]=\frac{\Gamma_\alpha\gamma^2k^3|E_0|^2}{8\rho\varepsilon\omega_\alpha[(\omega_\alpha-kv_s)^2+\Gamma_\alpha^2]}\left[1+\frac{\omega_{Mb}^2}{\omega_0\omega_s}\right] . \tag{15}$$

Thus, it is found from (15) that the maximum Brillouin gain is obtained when $\omega_\alpha\approx kv_s$, i.e., in the dispersionless regime of the acoustic wave in the crystal. $[g_b(\omega_s)]$ is also found to depend on the pump frequency through ω_0 and ω_s and on cyclotron frequency ω_0 and carrier plasma frequency ω_P through ω_{Mb}.

The threshold condition required for inciting SBS is obtained (by setting $[(P_b)_{cd}(\omega_s)=0]$) as

$$|E_{0\text{th}}|_b=\frac{m}{ek}[(\omega_s^2-\omega_{Mb}^2-i\omega_s\nu)^{1/2}\times(\omega_\alpha^2-\omega_{Mb}^2-i\omega_\alpha\nu)^{1/2}] . \tag{16}$$

Both the acoustic and the stimulated Brillouin scattered optical beams are emitted along specific directions and their generation occurs only above $|E_{0\text{th}}|_b$. However, in most dielectric media, the phenomenon of beam trapping gives rise to intensities exceeding the threshold value for SBS even at moderate input powers. As a result, the experimentally observed threshold in most semiconducting materials is that of beam trapping.[7]

V. RESULTS AND DISCUSSIONS

The physical constants considered for the analytical investigation in order to establish the validity of our model and to study the effects of a longitudinal magnetic field can be obtained from Refs. 5 and 8.

From (10) and (16), it is found the external magnetic field reduces the threshold fields for both processes to an appreciable extent in the highly doped medium. This trend has also been observed experimentally by Murav'jov and Shastin.[4] The threshold fields for both stimulated scattering processes increase linearly with the increase in carrier concentration of the medium.

The nonlinear optical susceptibility due to stimulated scattering processes is found to increase with a rise in the carrier concentration of the medium. The third-order Raman susceptibility of a medium with carrier concentration 10^{24} m^{-3} is found to be 3.9×10^{-7} esu, whereas the cubic Brillouin susceptibility is 8.1×10^{-11} esu. However, at lower doping level, the magnitude of $\chi_r^{(3)}$ and $\chi_b^{(3)}$ is lowered by about five orders and are potentially not very useful in the fabrication of cubic nonlinear devices. The Raman susceptibility due to molecular vibrational polarization at $\omega_c=0$ is 2.59×10^{-11} esu, whereas the third-order Brillouin susceptibility due to the electrostrictive coupling mechanism is around 5.43×10^{-11} esu. The magnitude of the third-order susceptibilities agrees well with the experimentally observed[9] and theoretically quoted values.[10] It should be noted from (8) and (14) that the magnetic field also produces an increase in the magnitude of $\chi_{\rm eff}^{(3)}$ when $\omega_M^2 \gg \omega_0\omega_1$.

Equations (9) and (15) can be used to estimate the ratio (g_r/g_b) between the steady-state gain constants for SRS and SBS in terms of material parameters. This enables one to estimate qualitatively the gain constant of one of the stimulated scatterings in terms of the gain constant of the other. Figure 1 depicts the dependence of $g_{r,b}$ on the externally induced magnetic field via the cyclotron frequency ω_c. $g_{r,b}$ is fairly independent of ω_c at a lower magnitude of B_0. But as ω_c approaches the molecular vibrational-acoustic wave frequency, $g_{r,b}$ increase rapidly and then saturates at a higher magnitude of B_0. Thus a higher magnitude of $g_{r,b}$ is obtained at cyclotron frequencies comparable to the pump frequency (i.e., $\omega_c \leq 0.1\omega_0, \omega_0$), which is independent of the external magnetic field. The magnetic field B_0 is effective only when the cyclotron frequency ω_c is comparable to the molecular vibrational or the acoustical wave frequency as is evident from the enhancement parameter ω_M (via ω_R). And for $\omega_c \gg \omega_v, \omega_\alpha$, it is found that ω_M (via ω_R) becomes independent of ω_c. Hence, the stimulated laser may be tuned by operating the magnetic field (cyclotron frequency) around the vibrating phonon frequency. In the case of a transversely applied magnetic field,[5] it has been found that the magnetic field is effective only when the cyclotron frequency ω_c exceeds the vibrational phonon frequency of the scattering medium. Thus the circular polarization induced by the longitudinal magnetic field saturates the Stokes gain at higher magnetic fields. It can be further inferred from (9) and (10) and subsequently from Fig. 1 that stimulated Brillouin scattering has a larger steady-state gain as compared to a competing stimulated Raman process. This outcome is in agreement with the investigations carried out by Maier, Wendl, and Kaiser,[11] where they had compared both steady-state gain and the corresponding transient response of the competing stimulated processes. It has been observed from the preceding analysis that the ratio of the SRS and SBS gain constants under the same pump intensity indicates that SBS exhibits higher gain than SRS by a magnitude $\approx\omega_v/\omega_\alpha$. This result is in conformity with those obtained with the semiclassical theory by Sen and Sen.[12]

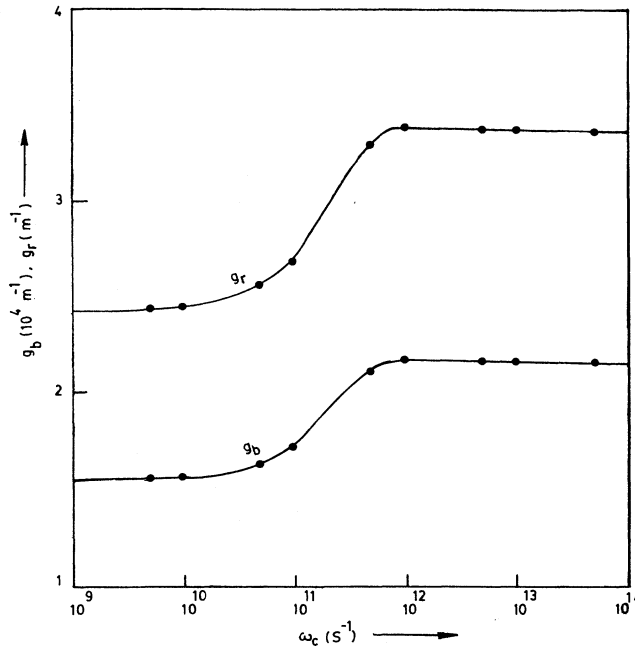

FIG. 1. Variation of gain constants g_r and g_b with magnetic field in terms of cyclotron frequency ω_c.

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