Stimulated Raman Scattering in a Weakly Polar III-V Semiconductor: Effect of dc Magnetic Field and Free Carrier Concentration *

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Using the hydrodynamic model of semiconductor plasmas, we perform an analytical investigation of stimulated Raman scattering (SRS) of an electromagnetic pump wave in a transversely magnetized weakly polar semiconductor arising from electron-density perturbations and molecular vibrations of the medium both produced at the longitudinal optical phonon frequency. Assuming that the origin of SRS lies in the third-order susceptibility of the medium, we investigate the growth rate of Stokes mode. The dependence of stimulated Raman gain on the external dc magnetic field strength and free carrier concentration is reported. The possibility of the occurrence of optical phase conjugation via SRS is also studied. The steady-state Raman gain is found to be greatly enhanced by the presence of the strong external dc magnetic field.

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Experimental and theoretical investigations of stimulated Raman scattering (SRS) have already gained momentum with the advent of high power lasers. SRS has been extensively studied by several authors and there exist some excellent reports on it. The theory of SRS has been discussed by many authors[1−6] both from the classical and from the quantum mechanical point of view. In this Letter, a classical discussion on the coupling of vibrational waves and light waves via a molecular system is given. If there are many photons in a radiation field it can properly be described by classical waves.[7] In the treatment of coupled wave problems, the classical description is even more appropriate since then the decay or amplification of the waves depends on the relative phases among them, whereas in the quantum mechanical description, if the number of quanta is prescribed, the phases will be undetermined as required by uncertainty principle. Moreover, the classical electromagnetic approach can yield valuable information regarding the threshold value of the pump electric field for the onset of SRS in unmagnetized as well as strongly magnetoactive semiconductors.[8−10] The quantum analogue of the classical treatment is obtained from the so-called coherent state of the field and has been investigated by Sen and Sen[5] in noncentrosymmetric crystals.

Recently the present authors[11] (hereafter referred to as paper I) have analytically investigated the steady-state and transient Brillouin gain in magnetoactive narrow band-gap semiconductors. Motivated by this work[11] and by the intense interest in the field of nonlinear optics in the present study we have attempted to study the phenomenon of SRS in a narrow-band gap n-type moderately doped weakly polar semiconductor crystal irradiated by an intense uniform pump wave. We have used the particular geometry where the incident high-frequency spatially uniform laser radiation (pump beam) $\boldsymbol{E}_0(x,t) = \hat{x}E_0 \exp[i(k_0x - \omega_0 t)]$ is applied parallel to the propagation vector of the density perturbation $\boldsymbol{k}$ (along the x-axis) and the external static magnetic field is taken normal to $\boldsymbol{k}$ along the z-axis. In order to study the effective Raman susceptibility arising due to induced nonlinear current density and the vibrational polarization, the hydrodynamic model of a homogeneous one component (with electrons as carriers) system is considered. In a Raman active medium the scattering of the high frequency pump wave is enhanced due to the excitation of a molecular vibrational mode. In the present analysis the Raman medium is taken as consisting of N harmonic oscillators per unit volume: each oscillator being characterized by its position x, molecular weight M, and the normal vibrational coordinate $u(x,t)$.

$$[(\partial^2/\partial t^2) + \Gamma_{op}(\partial/\partial t) + \omega_{\rm opo}^2]s = (q/M)E_{op} + (F/M), \quad (1)$$

where $s = u_+ - u_-$ is the relative displacement of positive and negative ions, respectively. In Eq. (1), Γ_{op} is the damping constant corresponding to phonon collision frequency (about $10^{-2}\omega_{\rm opo}$); $\omega_{\rm opo}$ is the undamped resonance frequency which is taken to be equal to longitudinal phonon frequency. $F[= 0.5\varepsilon_0\varepsilon_\infty(\partial\alpha/\partial s)_0\bar{E}^2$; ε_0 and ε_∞ are the absolute and high frequencies permittivities, respectively, F is the driving force per unit volume, which can be obtained by considering the electromagnetic energy in the po-

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larizable materials. The bar over E indicates the averaging over a few optical periods, as the molecules cannot respond to optical frequencies. This infers that natural mode of vibrations within the medium can be driven by the optical electric field because of nonvanishing differential polarizability (DP) $(\partial\alpha/\partial s)_0$. Here $q[=\omega_{\rm opo}\{(M/N)\varepsilon_0(\varepsilon_s-\varepsilon_\infty)\}^{1/2}]$ is the Szigeti effective charge (SEC).[12]

It is convenient to write the relative displacements in terms of a new parameter u defined as[13,14] $u=(NM)^{1/2}s$. Accordingly, Eq. (1) can be rewritten as

$$[(\partial^2/\partial t^2)+\Gamma_{\rm op}(\partial/\partial t)+\omega_{\rm opo}^2]u = (N/M)^{1/2}qE+0.5(N/M)^{1/2}\varepsilon_0\varepsilon_\infty(\partial\alpha/\partial u)_0\bar{E}^2. \quad (2)$$

The other fundamental equations employed are Eqs. (1)–(3) of paper I and

$$(\partial E/\partial x)=-(n_1e/\varepsilon)-(1/\varepsilon)(\partial P_{mv}/\partial x), \quad (3)$$

with

$$P_{mv}=\varepsilon N(\partial\alpha/\partial u)_0u^*E_e. \quad (4)$$

These one-dimensional equations are appropriate for non-degenerate semiconductors. The vibrations at longitudinal optical phonon frequency ω modulate the effective dielectric constant of the medium leading to an exchange of energy among the electromagnetic fields separated in frequency by integral multiples of ω (i.e., $\omega_0\pm q\omega$, where $q=1,2,3\cdots$). The electric induction in the presence of an external static magnetic field is given by $\boldsymbol{D}=\varepsilon_0\boldsymbol{E}_e+\boldsymbol{P}_{mv}$.[15] The equations and notations are explained in paper I.

The high frequency pump field gives rise to a carrier density perturbation within the Raman active medium. Now following the procedure of paper I for the theoretical development the perturbed electron density n_L of the Raman active medium due to molecular vibrations can be deduced from Eq. (4) as

$$n_L=\frac{1}{(\omega_p^2\Gamma_1-\omega^2-i\nu\omega)}\cdot\Big[\frac{N\omega_p^2\varepsilon\Gamma_1kq(\partial\alpha/\partial u)_0|E_0|^2}{eM(\omega_{\rm opo}^2-\omega^2-i\Gamma_{\rm op}\omega)}+\{ik(e/m)\Gamma_0E_0\}n_s^*\Big]. \quad (5)$$

The density perturbation associated with the molecular vibrations at frequency ω beats with the pump at frequency ω_0 and produces fast components of density perturbations. Following paper I the Stokes mode of this component at frequency $\omega_s=\omega_0-\omega$ can be obtained as

$$n_s=\frac{1}{(\omega_p^2\Gamma_1-\omega_s^2+i\nu\omega_s)}\cdot\Big[\frac{N\omega_p^2\varepsilon_0^2\varepsilon_l\omega_\infty\Gamma_1k_s(\partial\alpha/\partial u)_0^2|E_0|^2E_s^*}{2eM(\omega_{\rm opo}^2-\omega^2-i\Gamma_{\rm op}\omega)}+\{ik_{\rm op}(e/m)\Gamma_0E_0\}n_L^*\Big], \quad (6)$$

where $\omega_p=(n_0e^2/m\varepsilon)^{1/2}$ is the electron-plasma frequency, $\omega_c=eB_0/m$ is the electron-cyclotron frequency, $\Gamma_0=1-\omega_c^2/(\Delta_0^2+\omega_c^2)$, $\Gamma_1=1-\omega_c^2/(\Delta_1^2+\omega_c^2)$, $\Delta_0=\nu-i\omega_0$, and $\Delta_1=\nu+i(k_sv_0-\omega_s)$.

The resonant Stokes component of current density due to finite nonlinear induced polarization can be given as

$$J_{NL}=n_L^*ev_0. \quad (7)$$

The time integral of nonlinear current density yields the expression for the nonlinear induced polarization as

$$P_{\rm cd}(\omega_s)=\int J_{NL}(\omega_s)dt. \quad (8)$$

In the present investigation we have restricted ourselves only to the infrared transparency regime where photon energies are less than the crystal band-gap energy. In other words, we have neglected the induced polarization due to transition dipole moment of the transversely magnetized Raman active medium. SRS is a third-order nonlinear phenomenon and the simplified expression for the third-order susceptibility, termed as Raman susceptibility, is obtained using Eqs. (5)–(8) as

$$\chi_{R,cd}(\omega_s)=\frac{1}{2\omega_sM\{\omega^2\omega_p^2\delta\}}\cdot\Big\{N\Gamma_0\Gamma_1\Delta_0^2\Delta_1^2(\nu-i\omega_0)\cdot\Big[\frac{q(\partial\alpha/\partial u)_0}{\Delta_0^2\Delta_1^2-\{2(e/m)^2k^2/\eta\varepsilon_0c\}I_0}-i\frac{2\{(\partial\alpha/\partial u)_0^2/\eta\varepsilon_0c\}}{(\Delta_0^2\Delta_1^2/I_0)-\Gamma_0(ek/m)^2}\Big]\Big\}, \quad (9)$$

where $\delta=\{1-(\omega_0/\omega_p)^2\}\{1-(\omega/\omega_p)^2\}$ may be termed as the coupling parameter and $I_0=0.5\varepsilon_0\eta c_l|E_0|^2$; with c_l being the velocity of light in the crystal.

Besides Raman susceptibility, the system should also possess a polarization created by the interaction of the pump wave with the molecular vibrations generated within the medium such that

$$\chi_{R,mw}(\omega_s)=\frac{\varepsilon_0\varepsilon_\infty\omega_c^2N(N/M)^{1/2}(\partial\alpha/\partial u)_0^2}{2(\omega_{\rm opo}^2-\omega^2-i\Gamma_{op}\omega)}. \quad (10)$$

Thus the effective Raman susceptibility of weakly polar semiconducting medium is obtained as

$$\chi_{R,\rm eff}(\omega_s)=\chi_{R,cd}(\omega_s)+\chi_{R,mv}(\omega_s). \quad (11)$$

The Raman gain coefficient $|g_R(\omega_s)|$ of the Stokes mode which is related to imaginary part of $\chi_{R,\rm eff}[=(\chi_{R,\rm eff})_r+i(\chi_{R,\rm eff})_i]$ is given by

$$g_R(\omega_s)=-(k/2\varepsilon_l)(\chi_{R,\rm eff})_i|E_0|^2. \quad (12)$$

In order to establish the validity of the present model, we have applied the analytical results obtained above to weakly-polar semiconductor-like n-type InSb at 77 K. We take the physical parameters given in Ref. [5].

From Eq. (12) we find

$$g_R \approx 9.25 \times 10^{-7} I_0, \tag{13}$$

where I_0 in units of $\mathrm{Wm^{-2}}$ and g_R in $\mathrm{m^{-1}}$. The steady-state gain constant g_R obtained for InSb is found to be in agreement with that quoted by Yariv.[3] Equation (13) clearly indicates that the Raman gain coefficient varies linearly with excitation intensity. This trend has also been experimentally observed in magnetized InSb crystal duly shined by 10.6 μm CO_2 laser.[1] The high excitation intensity may damage the medium. For example, Kruer *et al.*[16] pointed out that a typical excitation intensity (about $4.2 \times 10^{12}\,\mathrm{Wm^{-2}}$) of Q-switched 170 ns pulsed 10.6 μm CO_2 laser damages the InSb crystal at 300 K. Therefore, in the present report, the results have been obtained for $E_0 = 10^7\,\mathrm{Vm^{-1}}$ (corresponding to $I_0 = 5.3 \times 10^{11}\,\mathrm{Wm^{-2}}$) well below the damage threshold.

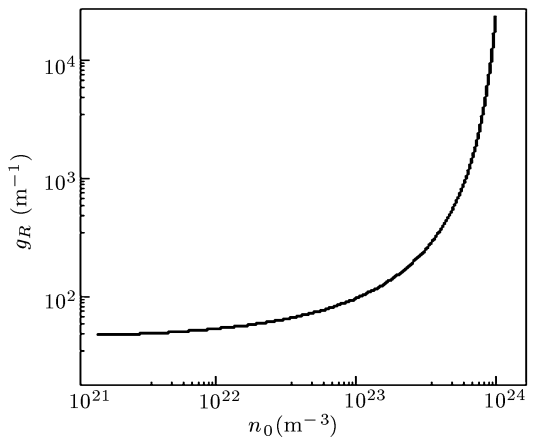


Fig. 1. Nature of variation of Raman gain coefficient g_R with free carrier concentration n_0 in the presence of dc magnetic field.

We have studied the nature of variation of effective Raman gain coefficient as a function of doping level in Fig. 1 in the presence of dc magnetic field. It exhibits almost doping-independent behaviour of g_R in lightly doped semiconductors. With further increase of doping level, g_R increases sharply. This type of variation can be understood from the fact that g_R depends upon longitudinal optical phonon-plasmon coupling which can be seen by the terms in curly bracket in denominator of Eq. (9). For $n_0 > 10^{23}\mathrm{m^{-3}}$ (corresponding to $\omega_p > 3.8 \times 10^{13}\mathrm{s^{-1}}$), the coupling parameter increases rapidly and thus g_R increases sharply. This result leads one to conclude that the longitudinal optical phonon-plasmon coupling increases the Raman gain coefficient in highly doped semiconducting crystals. In investigation of tunable stimulated Raman scattering from mobile carriers in semiconductors such as InSb, InAs, and GaAs, a similar type of variation of spin flip Raman gain coefficient with free carrier concentration is observed by Patel and Shaw,[17] where they computed the gain coefficient and found that it increases sharply for $n_0 > 10^{17}\mathrm{cm^{-3}}$.

Figure 2 displays the qualitative behaviour of the steady-state Raman gain factor g_R as a function of cyclotron frequency ω_c corresponding to a transverse magnetic field B_0 for $n_0 > 10^{22}\mathrm{m^{-3}}$. It has been observed that g_R increases sharply around $\omega_c \approx 3.75 \times 10^{13}\,\mathrm{s^{-1}}$ corresponding to static magnetic field $B_0 = 3$ Tesla. It is noteworthy that the enhancement is considerable only when the electron cyclotron frequency becomes nearly resonant with optical phonon frequency. One may also note from Eq. (13) that a higher Raman gain can be achieved by increasing the pump electric strength E_0. However, an arbitrary increment in B_0 may damage the medium. The most striking fact of the present report is that for $E_0 = 10^8\mathrm{Vm^{-1}}$, the first term insider the square bracket in Eq. (11) dominates over second and Raman gain is due to finiteness of both the SEC and DP. However, for $E_0 > 10^8\mathrm{Vm^{-1}}$ the contribution of SEC is nearly wiped off and the Raman gain spectrum becomes dependent only on DP.

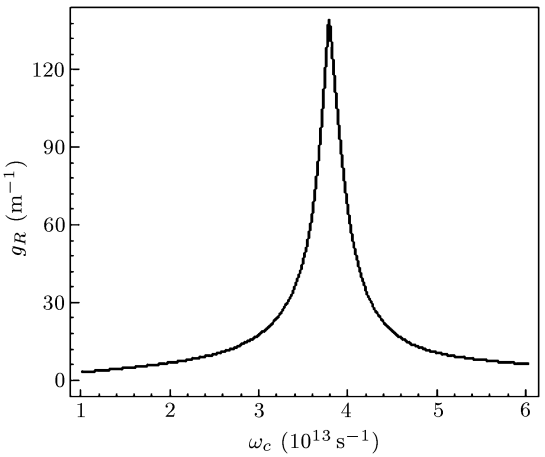


Fig. 2. Variation of Raman gain coefficient g_R with cyclotron frequency ω_c at $n_0 = 10^{22}\mathrm{m^{-3}}$.

To obtain a qualitative estimation of the effect of the large transverse static magnetic field on the Raman instability, we can observe from Eq. (12) that the threshold value of the pump can be greatly reduced by increasing the value of B_0. The effect of B_0 on Raman growth rate can be studied from Eq. (12) and is found for the parameters chosen as

$$\frac{(g_R)_{B_0 \neq 0}}{(g_R)_{B_0 = 0}} = 7.6 \times 10^2 \tag{14}$$

at $k = 10^7 \mathrm{m}^{-1}$, $E_0 = 10^7 \mathrm{Vm}^{-1}$ and $\omega_c \sim \omega$.

In order to explore the possibility of optical phase conjugation via stimulated Raman scattering (OPC-SRS) in the presence of a transverse magnetic field, we have employed the theoretical formulation of the Raman growth. The OPC threshold condition is given by $g_R(\omega_s)L > 30$ with L being the cell length.[18] However, for a pulse duration $\tau_p \geq 10^{-9}$ s, the cell length can be taken equal to the interaction length x. It may be noted that in order to reach the SRS threshold, either the beam must be focused in a nonlinear medium or a sufficient interaction length must be provided by directing the beam into the light guide. In both the cases, an appreciable amplification length is provided only in the longitudinal direction. The geometry thereby permits either forward or backward scattering. Thus one can achieve OPC-SRS in n-type InSb, if the cell length L is taken to be about $30/g_R(\omega_s)$. The external magnetic field induces a certain degree of nonlinearity into the propagating electromagnetic waves, which is favourable for occurrence of phase conjugation. The optimum OPC length can therefore be effectively reduced by the application of a strong magnetic field. The cell length can be further reduced by increasing the carrier concentration as well as excitation intensity, within the limit of the damage threshold.

In conclusion, the present theory explains satisfactorily the phenomenon of stimulated Raman scattering and possibility of optical phase conjugation via SRS in magnetoactive weakly polar III–V semiconductors under moderate pump electric field strengths.

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