Stimulated Raman scattering in weakly polar transversely magnetized doped semiconductors

M. Singh, P. Aghamkar,* and N. Kishore
Department of Applied Physics, Guru Jambheshwar University of Science & Technology, Hisar-125001, India

P. K. Sen
Department of Applied Physics, S.G.S. Institute of Science and Technology, Indore-452003, India

M. R. Perrone
Dipartimento di Fisica, Universita degli Studi di Lecce, Arnesano, I-73000 Lecce, Italy
(Received 23 October 2006; revised manuscript received 21 May 2007; published 31 July 2007)

Following the coupled-mode theory, stimulated Raman scattering (SRS) has been investigated in weakly polar, transversely magnetized doped semiconductor under off-resonant transition regime. The origin of the nonlinear interaction is considered to be in the Szigeti effective charge and the differential polarizability of the medium. Numerical estimates are made for a representative n-InSb crystal duly irradiated by a few nanosecond pulsed 10.6 μm CO_2 laser. Efforts are directed toward optimizing the doping level and an applied static-magnetic field for the occurrence of first-order SRS using minimum possible threshold to yield the maximum possible gain. It is found that, in the presence of a strong static-magnetic field, the backward stimulated Raman first-order Stokes mode can be substantially amplified under moderate pump field strengths in weakly polar III-V doped semiconducting crystals, which proves its potential as candidate materials for fabrication of cubic nonlinear devices. For field strengths ($E_0 > 10^8$ V m^{-1}), the results are found to be well in agreement with available literature.

DOI: 10.1103/PhysRevB.76.012302 PACS number(s): 42.65.−k, 42.65.Dr, 78.55.Cr

I. INTRODUCTION

The study of stimulated Raman scattering (SRS) is one of the major topics in the area of laser-matter interaction due to its manifold applications in a wide range of optoelectronics and laser spectroscopy. In the past, this topic has been studied extensively in gaseous, plasmas and liquids.[1–3] Due to the existence of the sophisticated fabrication technology and the experimental observations of large optical nonlinearities in the vicinity of band gap resonant transitions, semiconductors have been the natural choice as active media for the study of nonlinear optical phenomena[4,5] in preference to materials such as liquids and gases.

The optical properties of a material based on nonlinear optical susceptibility can be modified by the application of electric and magnetic fields from outside. This property can easily be exploited to understand the mechanisms involved in several nonlinear processes such as electro-optic and magneto-optic effects. Of late, several research groups[6–8] have observed a large enhancement in surface field terahertz emission from a variety of III-V semiconducting crystals when subjected to an externally applied magnetic field. The effect of a magnetic field on the growth rate of Raman back-scattered circularly polarized electromagnetic waves was reported by Maraghechi and Willett.[9] In the recent past, using the classical and semiclassical approaches, a significant amount of research work on SRS and its consequent instabilities in magnetized doped semiconductors has been reported by Sen and co-workers.[10–12] Neogi and Ghosh[13] have analytically investigated SRS and also made a comparison between the SRS and stimulated Brillouin scattering[14] in a centrosymmetric magnetized semiconductor. They pointed out that significant Raman growth is possible if the pump field strength is very large (i.e., $\sim 10^8$ V m^{-1}), which is the fundamental limitation of their theory. Moreover, in the previous reports, the origin of SRS in polar semiconductors, has been taken into finiteness of the differential polarizability only. However, in these semiconductors, the parametric coupling between the pump wave and the optical phonon mode also depends on the Szigeti effective charge. While studying the interaction of magnetoplasmons with optical phonons in polar semiconductors, it has been found that the coupling depends on the Szigeti effective charge of the compound semiconductors, and it is stronger for those that are more ionic but weakly polar.[15] As far as we know, no such attempt has yet been made to analyze critically the role of an externally applied magnetic field on the Raman growth in weakly polar doped III-V semiconductors, where the origin of the process is taken to be in the finiteness of the Szigeti charge as well as on the differential polarizability. In this Brief Report, using the hydrodynamic model of a semiconductor plasma[10] and following the coupled-mode approach, we have analyzed the effect of an external magnetic field on Raman growth in weakly polar doped semiconductors and found that the backward Stokes line can be substantially enhanced in a low doping regime by a nanosecond pulsed not-too-high power infrared laser irradiation.

II. THEORETICAL FORMULATIONS

This section deals with theoretical formulation of complex effective Raman optical susceptibility and consequent gain coefficient for the first-order Stokes component of backward scattered electromagnetic wave in weakly polar III-V doped semiconductors subjected to a transverse static-magnetic field B_0. The magnetic field is applied along the z axis, i.e., normal to propagation vectors $\vec{k}_0$, $\vec{k}_s$, and $\vec{k}_{op}$ (all

 ©2007 The American Physical Society

parallel to the x axis) of the three interacting waves, viz. pump $(\omega_0,\vec{k}_0)$, signal $(\omega_s,\vec{k}_s)$, and optical phonon $(\omega_{op},\vec{k}_{op})$. The energy exchange between these waves can be described by phase-matching conditions: $\hbar\omega_0=\hbar\omega_s+\hbar\omega_{op}$ and $\hbar\vec{k}_0=-\hbar\vec{k}_s+\hbar\vec{k}_{op}$. The Stokes mode originates from the coupling of the pump wave with density perturbations at an optical phonon frequency in the crystal. The optical phonon mode possesses both the transverse and longitudinal components. The polarization associated with the longitudinal vibration is much stronger than the transverse component in weakly polar III-V crystals with zinc blende structures.[16] Thus, one can neglect the transverse component of the electric field, and the optical phonon mode may be treated as a purely longitudinal mode with optical phonon frequency ω_{op}. We also assume the crystal sample to be irradiated by a laser with photon energy well below the band gap energy of the crystal. This choice allows the optical properties of the sample to be influenced considerably by the free carriers and to remain unaffected by the photoinduced interband transition mechanisms. Let us consider the Raman-active medium that consists of N harmonic oscillators per unit volume, each oscillator being characterized by its position x and normal vibrational coordinates $u(x,t)$. The equation of motion for a single oscillator (optical phonon) is then represented as[17]

$$(\partial^2u/\partial t^2)+\Gamma_{op}(\partial u/\partial t)+\omega_{opo}^2u = (q/M)E_{op}+(1/2M)\varepsilon(\partial\alpha/\partial u)_0E_0E_s^*, \tag{1}$$

ω_{opo}^2 is the optical phonon frequency at $k_{op}=0$. $\varepsilon=(\varepsilon_0,\varepsilon_\infty)$, and q and M are the dielectric constant effective charge and reduced mass of the diatomic system, respectively. The other fundamental equations of the model are

$$(\partial E_1/\partial x)=-(n_1e/\varepsilon)-(1/\varepsilon)(\partial P/\partial x), \tag{2}$$

$$(\partial\vec{v}_0/\partial t)=-(e/m)(\vec{E}_0+\vec{v}_0\times\vec{B}_0)-\nu v_0, \tag{3}$$

$$(\partial\vec{v}_1/\partial t)=-(e/m)(\vec{E}_1+\vec{v}_1\times\vec{B}_0)-\nu\vec{v}_1-(\vec{\nabla}\cdot\vec{v}_1)\vec{v}_0, \tag{4}$$

$$(\partial n_1/\partial t)+n_0(\partial v_1/\partial x)+n_1(\partial v_0/\partial x)+v_0(\partial n_1/\partial x)=0. \tag{5}$$

The notations used in Eq. (1)–(5) are well explained in the previous reports[10,18] The natural vibrations at frequency ω_{op} give rise to carrier density perturbation within the Raman-active medium. The density perturbation may oscillate at optical phonon (ω_{op}) and electromagnetic wave frequencies $(\omega_0\pm p\omega_{op})$, where $p=1,2,3,\ldots$. In general, as the interaction length of the amplifying medium or the pump power increases (>100 MW/cm^2), the energy transfer from the pump wave to the first-order Stokes wave $(\omega_S=\omega_0-\omega_{op})$ can become so large that the Stokes wave reaches intensity levels approaching those of the pump wave (i.e., pump depletion). Eventually, the first-order Stokes component acts as a source of amplification of the higher-order Stokes modes such as $\omega_0-2\omega_{op}$, $\omega_0-3\omega_{op}$, etc. In addition, the emission of the higher-order modes depends on the pump pulse duration. For example, for subnanosecond light pulse with intensity ~320 MW/cm^2 and interaction length ~30 cm, the transmitted laser power and first- and second-order Stokes emission are of approximately equal value (30%) while the third- and fourth-order Stokes components are smaller by a factor of 10^4 and 10^{12}, respectively.[19] These results establishes that even at high pump intensities, the lower Stokes components are approximately of equal intensity, while the higher Stokes waves have rapidly decreasing intensity values. In addition, the higher-order Stokes modes are generally emitted in cones and can be achieved by angular phase matching of four-wave mixing processes. The occurrence of this conical emission leads to beams with poor spatial quality, thereby reducing the conversion efficiency. In spite of all this fact, higher-order SRS holds the potential to generate ultrabroadband light pulses[20,21] and convert ultraviolet laser output to visible radiation[22] with energy conservation efficiencies approaching unity. In the present work, the analysis of SRS is confined for moderate pump power with pulse duration longer than the longitudinal optical phonon life time. The interaction length of the crystal sample is taken to be a few millimeters. Moreover, in steady-state SRS, only the mode with the maximum gain participates in the process. Under these approximations, the generation of the higher-order SRS modes may be neglected and the first-order component need to be considered.

Using phase-matching conditions, the perturbed density of the Raman-active medium associated with optical phonon (n_{op}) and first-order Stokes mode (n_{st}) can be deduced by following the procedure of Ref. 10,

$$n_{op}=\frac{ik_{op}}{\Delta_{op}^2}\left[\frac{\Omega_p^2\delta_1N\varepsilon_0\varepsilon_\infty q(\partial\alpha/\partial w)_0E_0E_s^*}{2eM\Delta^2}+\left(\frac{e}{m}\right)\delta_0E_0n_{st}^*\right] \tag{6a}$$

and

$$n_{st}=\frac{ik_s}{\Delta_{ps}^2}\left[\frac{\Omega_p^2\delta_1N\varepsilon_0^2\varepsilon_\infty^2(\partial\alpha/\partial w)_0^2|E_0|^2E_s}{2eM\Delta^2}+\left(\frac{e}{m}\right)\delta_0E_0n_{op}^*\right], \tag{6b}$$

where w is a new parameter and is defined as $w=(NM)^{1/2}u$, $\Omega_p=(n_0e^2/m\varepsilon)^{1/2}$ (electron plasma frequency), $\omega_c=(e/m)B_0$ (electron cyclotron frequency), $\delta_0=1-\omega_c^2/(\Delta_0^2+\omega_c^2)$, $\delta_1=1-\omega_c^2/(\Delta_1^2+\omega_c^2)$, $\Delta_0=\nu-i\omega_0$, $\Delta_1=\nu-i\omega_s+ik_sv_0$, $\Delta^2=(\omega_{opo}^2-\omega_{op}^2-i\Gamma_{op}\omega_{op})$, $\Delta_{ps}^2=(\Omega_p^2/\delta_1)-\omega_s^2+i\nu\omega_s$, and $\Delta_{op}^2=(\Omega_p^2/\delta_1)-\omega_{op}^2-i\nu\omega_{op}$.

Equations (6) represent the coupling between optical phonon and first-order Stokes mode via the pump electric field E_0 and the magnetic field B_0 (through ω_c). These coupling acts as a modulated source and feeds energy to the scattered Stokes component; as a result, the Stokes component amplifies with a large gain coefficient at the expense of the pump electric field and the external magnetic field. The first-order Stokes component of the induced current density due to modulated charge density can be expressed as

$$J(\omega_s)=(n_0ev_1)_L+(n_{1s}^*ev_0)_{NL}. \tag{7}$$

The time integral of the second term of Eq. (9) yields the expression for the nonlinear induced polarization due to the perturbed carrier density:

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

It is a well known fact that the origin of the SRS process lies in that component of $P_{cd}(\omega_s)$ which depends on $|E_0|^2E_s$, and the corresponding third-order susceptibility of the Raman-active medium, which is known as the Raman optical susceptibility $\chi_{R,cd}(\omega_s)$, is obtained using Eqs. (6)–(8):

$$\chi_{R,cd}(\omega_s) = \frac{\varepsilon_\infty(e/m)\Omega_p^2\delta_0\delta_1 N k_{op}\Delta_0}{2\omega_s M\Delta^2\Delta_{op}^2(\Delta_0^2+\omega_c^2)}\left[q\left(\frac{\partial\alpha}{\partial w}\right)_0 - 2i\varepsilon\left(\frac{k_s e\delta_0}{m\Delta_{ps}^2}\right)\times\left(\frac{\partial\alpha}{\partial w}\right)_0^2|E_0|^2\right]. \tag{9}$$

It is evident from Eq. (9) that both the free-carrier concentration (through Ω_p) and the external magnetic field (through ω_c) influence $\chi_{R,cd}$. In addition to free-carrier dependent Raman optical susceptibility, the medium also possesses the molecular vibration dependent optical susceptibility. This susceptibility can be separated from the free-carrier dependent susceptibility by considering solely differential polarizability. Following Shen,[1] we obtain

$$\chi_{R,mv}(\omega_s) = \frac{\varepsilon N(\partial\alpha/\partial w)_0^2}{2M\Delta^2}. \tag{10}$$

The effective Raman optical susceptibility of the weakly polar doped semiconductors can be given as

$$\chi_{R,eff}(\omega_s) = \chi_{R,cd}(\omega_s) + \chi_{R,mv}(\omega_s) = \frac{\varepsilon N(\partial\alpha/\partial w)_0^2}{2M\Delta^2} + \frac{\varepsilon_\infty(e/m)\Omega_p^2\delta_0\delta_1 N k_{op}\Delta_0}{2\omega_s M\Delta^2\Delta_{op}^2(\Delta_0^2+\omega_c^2)}\left[q\left(\frac{\partial\alpha}{\partial w}\right)_0 - 2i\varepsilon\left(\frac{k_s e\delta_0}{m\Delta_{ps}^2}\right)\times\left(\frac{\partial\alpha}{\partial w}\right)_0^2|E_0|^2\right]. \tag{11}$$

In order to obtain the effective Raman gain coefficient g_R, we employ the relation[1]

$$g_R(\omega_s) = -(k_s/2\varepsilon_l)[\chi_{R,eff}(\omega_s)]_{imag}|E_0|^2, \tag{12}$$

where $[\chi_{R,eff}(\omega_s)]_{imag}$ is the imaginary part of the effective Raman optical susceptibility. The threshold value of pump field $E_{0,th}$ required for the onset of SRS can be obtained by setting $[\chi_{R,eff}(\omega_s)]_{imag}=0$.

III. RESULTS AND DISCUSSION

The physical constants are taken from Refs. 10 and 18 (i) to investigate the effect of an externally applied static-magnetic field and the doping concentration on threshold condition for the onset of SRS process as well as Raman gain well above the threshold, and (ii) to test the validity of our model. The nature of variation of the threshold pump field strength $E_{0,th}$ with a magnetic field B_0 for two different values of doping concentrations $n_0=2\times10^{21}$ m^{-3} (dotted line) and $n_0=10^{23}$ m^{-3} (solid line) has been plotted in Fig. 1. In the case of a low doped semiconductor, it can be observed

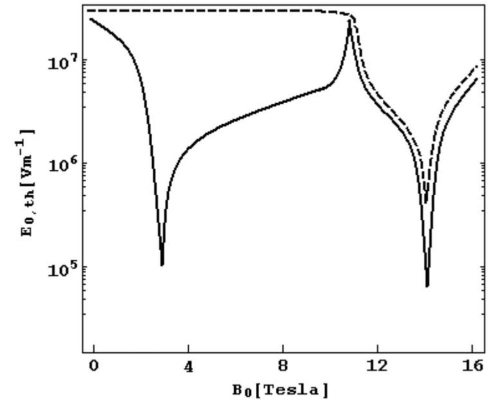

FIG. 1. The nature of dependence of threshold pump amplitude $E_{0,th}$ on static external magnetic field B_0 in n-InSb for $n_0=2\times10^{21}$ m^{-3} (dotted line) and $n_0=10^{23}$ m^{-3} (solid line).

that initially $E_{0,th}$ decreases with increasing magnetic field and reduces to 10^5 V m^{-1} when ω_c is in resonance with optical phonon frequency ω_{op}. With further increase in B_0, $E_{0,th}$ increases almost linearly due to increase in losses of the optical phonon mode and linear absorption (α_l) of the Raman medium. At $B_0\approx11.8$ T, α_L substantially increases as ω_c approaches ω_s. However, when $\omega_c\approx\omega_0>\omega_s$, loss of the medium decreases and, therefore, $E_{0,th}$ decreases sharply and attains a minimum value of about 5×10^4 V m^{-1}. However, in a highly doped semiconductor, threshold amplitude can be reduced significantly around $\omega_c\sim\omega_0$. It has been noticed that when $n_0\geqslant10^{24}$ m^{-3}, the $E_{0,th}$ increases quadratically, and therefore, in such semiconductors it is difficult to achieve SRS by a moderate power infrared laser irradiation. The physical interpretation of Fig. 1 enables one to infer that a strong first-order component of SRS can be achieved at lower pump fields in a weakly polar low doped III-V semiconductor subjected to a strong transverse static-magnetic field and makes this crystal a potential candidate for cubic nonlinear devices. Figure 2 represents the qualitative dependence of Raman gain coefficient g_R as a function of magnetostatic field B_0 for two different values of the pump amplitude $E_0=8\times10^6$ and 2×10^7 V m^{-1}. It is observed that for

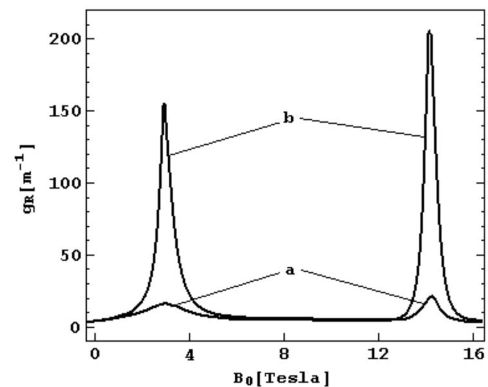

FIG. 2. Variation of Raman gain g_R with magnetic field B_0 for $n_0=2\times10^{21}$ m^{-3}. Curves a and b are for $E_0=8\times10^6$ and 2×10^7 V m^{-1}, respectively.

any pump amplitude above the threshold amplitude, the Raman gain spectrum increases sharply at B_0=3.0 T and B_0 =14.0 T due to resonance conditions $\omega_c \sim \omega_{op}$ and $\omega_c \sim \omega_0$, respectively. This result suggests that, around the resonance conditions, the Lorentz contribution is very effective in enhancing the Raman gain in III-V low doped semiconductors. However, the Raman gain is fairly independent of the magnetic fields between $4 \leqslant B_0 \leqslant 13$ T. This feature indicates that the Lorentz contribution is quite small when $\omega_{op} \leqslant \omega_c \leqslant \omega_0$. With further increase in B_0 resulting in $\omega_c > \omega_0$, g_R decreases sharply due to the departure and finally saturates at a small value. One may also note that a high Raman gain can also be achieved by increasing the pump field strength E_0. Practically, however, pump field strength cannot be increased arbitrarily because it may damage the sample. The most striking fact of this Brief report is that, for $E_0 \leqslant 10^8$ V m^{-1} the first term inside the square bracket in Eq. (11) dominates over the second term and Raman gain arises due to coupling between the Szigeti effective charge and differential polarizability of the medium. However, when $E_0 > 10^8$ V m^{-1}, the contribution of the Szigeti charge is wiped off and Raman gain spectrum depends only on the differential polarizability of the medium. The latter case supports the results of Neogi *et al.*[13,14]

Figure 3 represents the analytical dependence of Raman gain on doping concentration for B_0=3.0 T (curve 1) and B_0=14.0 T (curve 2). It is clearly evident that in a low doping regime and B_0=14.0 T, g_R increases significantly when the electron-plasma frequency Ω_p is almost in resonance with the modified optical phonon frequency (say, magneto-optical phonon frequency $\omega_m = \delta_1 \omega_{op}$). However, in the presence of a comparatively low magnetic field (B_0=3.0 T), g_R can be enhanced in a highly doped semiconductor, but the value of g_R in this case is smaller than the former case. Moreover, the positions of the peak of g_R shift toward lower values of n_0 (i.e., redshift) with increasing magnetostatic fields.

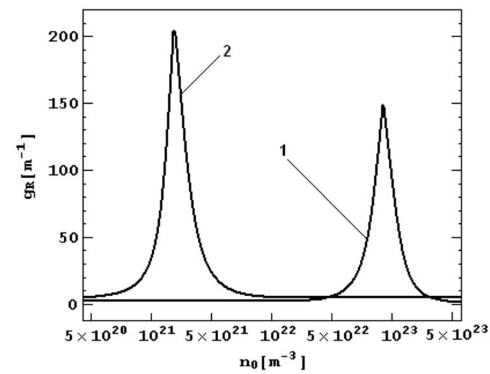

FIG. 3. Behavior of Raman gain g_R with doping concentration n_0 for $E_0 = 2 \times 10^7$ V m^{-1}. Curves 1 and 2 are for B_0=3.0 T and B_0=14.0 T, respectively.

From the above discussion, we may conclude that a substantial enhancement of Raman gain can be achieved in weakly polar, transversely magnetized, lightly doped III-V semiconductors by nanosecond pulsed moderate power infrared laser irradiation.

ACKNOWLEDGMENTS

Many fruitful discussions with P. Sen are thankfully acknowledged. One of the authors (P.A.) acknowledges financial support from the Council of Scientific and Industrial Research (CSIR) and the Department of Science and Technology (FIST), New Delhi, India.

*Corresponding author; prasut2003@yahoo.co.in

[1] Y. R. Shen, *Principle of Nonlinear Optics* (Wiley, New York, 1984), Chap. 10.
[2] J. F. Drake, P. K. Kaw, Y. C. Lee, G. Schmidt, C. S. Liu, and M. N. Rosenbluth, Phys. Fluids **17**, 778 (1974).
[3] M. S. Sodha, R. P. Sharma, and S. C. Kaushik, J. Appl. Phys. **47**, 3518 (1976).
[4] R. K. Jain, Opt. Eng. (Bellingham) **21**, 199 (1982).
[5] S. D. Smith, Nature (London) **316**, 319 (1982), and references therein.
[6] C. Weiss, R. Wallenstein, and R. Beigang, Appl. Phys. Lett. **77**, 4160 (2000).
[7] R. McLaughlin, A. Corchia, M. B. Johnston, Q. Chen, C. M. Ciesla, and D. D. Arnone, Appl. Phys. Lett. **76**, 2038 (2000).
[8] G. Meinert, L. Banyai, P. Gartner, and H. Haug, Phys. Rev. B **62**, 5003 (2000).
[9] B. Maraghechi and J. E. Willett, J. Plasma Phys. **20**, 163 (1979).
[10] P. K. Sen, N. Apte, and S. Guha, Phys. Rev. B **22**, 6340 (1980).
[11] P. Sen and P. K. Sen, Phys. Rev. B **31**, 1034 (1985).
[12] P. Sen and P. K. Sen, Phys. Rev. B **33**, 1427 (1986).
[13] A. Neogi and S. Ghosh, Phys. Rev. B **44**, 13074 (1991).
[14] A. Neogi, S. Ghosh, and D. K. Sinha, Phys. Rev. B **47**, 16590 (1993).
[15] K. S. Srivastava, R. Srivastava, A. Sinha, A. Tandon, and A. N. Nigam, Phys. Rev. B **38**, 1357 (1988).
[16] S. S. Mitra and N. D. Massa, in *Handbook of Semiconductors*, edited by T. S. Moss (North-Holland, Amsterdam, 1982), pp. 83–189.
[17] S. D. Kramer, F. G. Parsons, and N. Bloembergen, Phys. Rev. B **9**, 1853 (1974).
[18] P. Aghamkar and P. K. Sen, Mol. Cryst. Liq. Cryst. Sci. Technol., Sect. B: Nonlinear Opt. **8**, 97 (1994).
[19] D. von der Lind, M. Maier, and W. Kaiser, Phys. Rev. **178**, 11 (1969).
[20] E. Sali, P. Kinsler, G. H. C. New, K. J. Mendham, T. Halfmann, J. W. G. Tisch, and J. P. Marangos, Phys. Rev. A **72**, 013813 (2005).
[21] S. E. Harris and A. V. Soklov, Phys. Rev. Lett. **81**, 2894 (1998).
[22] H. Komine and E. A. Stapparerts, Opt. Lett. **4**, 398 (1979).