Stimulated Raman scattering in a magnetized centrosymmetric semiconductor

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

I. INTRODUCTION

Stimulated Raman scattering induced by finite-amplitude coherent electromagnetic waves has received a great deal of attention in theoretical and experimental nonlinear optics research. This topic has been extensively studied by several authors and there exist some excellent reports on it.[1–8] The theory of the stimulated Raman effect has been discussed by many authors[4,5,7] both from the classical and from the quantum-mechanical points of view. In this paper a classical discussion on the coupling of vibrational waves and light waves via a molecular system is given. If there are many photons in the radiation field it can properly be described by classical waves.[9] 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 the uncertainty principle. The quantum analog of the classical treatment is obtained from the so-called coherent state of the field and has been recently investigated in much detail by Sen and Sen[7] in noncentrosymmetric crystals.

Recently the present authors[10] (hereafter referred to as paper I) have analytically investigated the possibility of optical phase conjugation via stimulated Brillouin scattering in a transversely magnetized semiconductor crystal. Motivated by this work[10] and by the intense interest in the field of stimulated Raman scattering (SRS) in the present paper we have attempted to study the phenomenon of stimulated Raman Stokes scattering in a narrow-band-gap *n*-type moderately doped semiconductor crystal irradiated by an intense uniform pump wave. The physical origin of the phenomenon lies in the nonvanishing, nonlinear polarization due to the coupling of the molecular vibrations having a frequency equal to that of the transverse-optical-phonon frequency ω_T with the pump frequency ω_0, as well as the electron plasma frequency ω_P in the presence of a magnetostatic field such that $\omega_T < \omega_P < \omega_0$ and the electron cyclotron frequency $\Omega_c\ (<\omega_0) \gg \omega_\tau\ [=\frac{1}{2}(\omega_T^2+\omega_P^2)^{1/2}]$. The analysis is based on the coupled mode theory which was employed earlier by Ghosh and Dixit.[11] The chief utility of the analysis in achieving the nonlinear optical phase conjugation has been discussed subsequently.

II. THEORY

This section deals with the theoretical formulation of the third-order nonlinear optical susceptibility $\chi^{(3)}$ for the Stokes component of the scattered electromagnetic wave in transversely magnetized semiconductors. We have used the particular geometry where the incident high-frequency spatially uniform laser radiation (pump wave) $\mathbf{E}_0\exp(-i\omega_0 t)$ is applied parallel to the propagation vector of the density perturbation k (along the x axis) and the external dc magnetic field $\mathbf{B}_0$ is taken normal to 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 hydrodynamical model of a homogeneous one-component (with electrons as carriers) system is considered. The semiconductor is assumed to possess an isotropic and nondegenerate conduction band and being centrosymmetric in nature the effect of any pseudopotential is neglected for analytical simplicity. 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 $u(x,t)$.

The optical-phonon mode is represented in a one-dimensional configuration as[4]

$$\frac{\partial^2 u(x,t)}{\partial t^2}+\Gamma\frac{\partial u(x,t)}{\partial t}+\omega_T^2 u(x,t)=\frac{F(x,t)}{M}\ , \qquad (1)$$

where $F(x,t)$ is the driving force per unit volume, which can be obtained by considering the electromagnetic energy in the presence of the molecules and in polarizable material it is given by

$$F(x,t)=\tfrac{1}{2}\epsilon(\partial\alpha/\partial u)_0\bar{E}^2(x,t) . \quad (2)$$

The notations used in Eqs. (1) and (2) are well explained in Yariv.[4] The other fundamental equations employed are Eqs. (2)–(4) of paper I and

$$\mathbf{P}=\epsilon N(\partial\alpha/\partial u)_0 u^* \mathbf{E}_{\text{eff}} , \quad (3)$$

$$\frac{\partial E_1}{\partial x}=\frac{n_1 e}{\epsilon}-N(\partial\alpha/\partial u)_0 E_{\text{eff}} . \quad (4)$$

These one-dimensional equations are appropriate for nondegenerate semiconductors. The molecular vibrations at 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., $(\omega_0 \pm p\omega)$ where $p=1,2,\ldots$]. The electric induction in the presence of an external dc magnetic field is given by $\mathbf{D}=\epsilon\mathbf{E}_{\text{eff}}+\mathbf{P}$.[12] 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_T) of the Raman-active medium due to molecular vibrations can be deduced from Eq. (4) as

$$n_T=\frac{i\epsilon k}{e}\left[\frac{\omega_T^2-\omega^2+i\omega\Gamma-(\epsilon N/2M)(\partial\alpha/\partial u)_0^2|E_0|^2}{(\epsilon/2M)(\partial\alpha/\partial u)_0 E_0^*}\right] \times u^* . \quad (5)$$

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

$$n_S=\frac{ik\bar{E}_0 n_T^*}{\bar{\omega}_R^2-\omega_S^2-i\nu\omega_S} , \quad (6)$$

where subscripts T and S denote the components of the perturbed carrier concentration associated with the molecular vibrations and Stokes mode, respectively, and $\bar{E}_0=[(e/m)E_0-\Omega_c v_{0y}]$; $\bar{\omega}_R^2=\nu^2\omega_R^2/(\nu^2+\Omega_c^2)$; $\omega_R^2=(\omega_P^2\omega_L^2)/\omega_T^2$, $\omega_L/\omega_T=\sqrt{\epsilon_L/\epsilon_\infty}$; $\omega_P^2=n_0e^2/m_0\epsilon_0\epsilon_L$ and $\Omega_c=eB_0/m$. The components of v_0 are given in Eq. (12) of paper I.

Following paper I, the resonant Stokes component of the current density due to finite nonlinear induced polarization becomes

$$J(\omega_S)=\frac{i\epsilon\omega_P^2\omega_0^2E_1(\omega_S)}{\omega_S(\omega_0^2-\omega_c^2)}+\frac{\epsilon k^2\bar{E}_0^2E_1}{(\delta_2^2+i\nu\omega)(\nu-i\omega_0)}\times\left[1-\frac{(\epsilon N/2M)(\partial\alpha/\partial u)_0^2|E_0|^2}{\delta_1^2+i\omega\Gamma}\right] , \quad (7)$$

where $\delta_1^2=\omega_T^2-\omega^2$; $\delta_2^2=\bar{\omega}_R^2-\omega_1^2$.

The first part of Eq. (7) represents the linear component of the induced current density while the latter term represents the nonlinear coupling among the three interacting waves via the nonlinear current density $J_{nl}(\omega_S)$. In this present investigation the effect of the transition dipole moment is neglected in order to study the effect of nonlinear current density on the induced polarization in a transversely magnetized Raman-active medium.

Henceforth treating the induced polarization $\mathcal{P}_{cd}$ as the time integral of the current density J_{nl}, we obtain the nonlinear induced polarization due to perturbed current density from Eq. (7) as

$$\mathcal{P}_{cd}(\omega_S)=\frac{-e^2k^2\epsilon_\infty|E_0|^2E_1(\omega_S)}{m^2\omega_0\omega_S(\delta_2^2+i\nu\omega_S)}\times\left[1-\frac{(\epsilon N/2M)(\partial\alpha/\partial u)_0^2|E_0|^2}{\delta_1^2+i\omega\Gamma}\right] . \quad (8)$$

The induced polarization at the Stokes frequency ω_S is defined by

$$\mathcal{P}_{cd}(\omega_S)=\epsilon_0\chi_R^{(3)}|E_0|^2E_1(\omega_S) . \quad (9)$$

It is well known that the origin of the SRS processes lies in that component of $\mathcal{P}_{cd}(\omega_S)$ which depends on $|E_0|^2E_1$ and the corresponding third-order susceptibility of the Raman-active medium which is known as the Raman susceptibility χ_R can be obtained using Eqs. (8) and (9),

$$(\chi_R^{(3)})_{cd}=\frac{-e^2k^2\epsilon_\infty\omega_0^3}{\omega_S m^2(\delta_2^2+i\nu\omega_S)(\omega_0^2-\Omega_c^2)^2} . \quad (10)$$

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

$$\mathcal{P}_{mv}(\omega_S)=\epsilon_0(\chi_R^{(3)})_{mv}|E_0|^2E_1 . \quad (11)$$

Using Eqs. (3) and (11), we obtain

$$(\chi_R^{(3)})_{mv}=\frac{\epsilon_\infty(\epsilon N/2M)(\partial\alpha/\partial u)_0^2\omega_0^2}{(\delta_1^2+i\omega\Gamma)(\omega_0^2-\Omega_c^2)} . \quad (12)$$

Thus the effective Raman susceptibility of the centrosymmetric medium is obtained as

$$(\chi_R^{(3)})_{\text{eff}}=(\chi_R^{(3)})_{mv}+(\chi_R^{(3)})_{cd}=\frac{\epsilon_\infty(\epsilon N/2M)(\partial\alpha/\partial u)_0^2\omega_0^2}{(\delta_1^2+i\omega\Gamma)(\omega_0^2-\Omega_c^2)}-\frac{e^2k^2\epsilon_\infty\omega_0^3}{\omega_S m^2(\delta_2^2+i\nu\omega_S)(\omega_0^2-\Omega_c^2)^2} . \quad (13)$$

We now estimate the Raman gain constant $|g_R(\omega_S)|$ of the Stokes mode which is related to the imaginary part of $(\chi_R^{(3)})$ $(=\chi_{Rr}^{(3)}+i\chi_{Ri}^{(3)})$ through

$$g_R(\omega_S)=-\frac{\omega_S}{\eta c_0}\chi_{Ri}^{(3)}|E|^2 , \tag{14}$$

where η is the refractive index of the medium and c_0 is the speed of light in a vacuum.

Substituting the value of $\chi_{Ri}^{(3)}$ from the imaginary part of Eq. (13) one finds

$$\begin{aligned} g_R(\omega_S)=&[\epsilon_\infty\omega_S\omega_0^2E_0^2/2m^2\eta c_0(\omega_0^2-\Omega_c^2)]\\ &\times[\omega m^2(\epsilon N/2M)(\partial\alpha/\partial u)_0^2(\omega_0^2-\Omega_c^2)(\delta_2^4+\omega_S^2\nu^2)-e^2k^2\omega_0\nu(\delta_1^4+\omega^2\Gamma^2)]\\ &\times[(\delta_1^2\delta_2^2-\nu\omega\omega_S\Gamma)^2+(\nu\omega_S\delta_1^2+\omega\Gamma\delta_2^2)^2]^{-1} . \end{aligned} \tag{15}$$

The effect of the external magnetic field is thus found to be quite significant on the Raman gain $[g_R(\omega_S)]$ which is also strongly dependent on the pump frequency through the terms ω_0^2 and ω_S^2.

III. RESULTS AND DISCUSSIONS

In order to establish the validity of the present model we have applied the analytical results obtained above to a nearly centrosymmetric semiconductor like n-type InSb at 77 K. The physical constants used are given in Ghosh and Dixit.[11]

From Eq. (15) we find

$$g_R\approx 4.5\times10^{-8}I_P , \tag{16}$$

where we have defined $I_P=\frac{1}{2}\eta\epsilon_0c_l|E_0|^2$; where c_l is the velocity of light in the crystal with I_P in W m^{-2} and g_R in m^{-1}. The steady-state gain constant g_R for InSb thus obtained is found to be in agreement with that quoted by Yariv.[4]

At high magnetic field the variation of g_R is found to be fairly striking. Figure 1 represents the qualitative behavior of the steady-state Raman gain factor g_R as a function of cyclotron frequency (Ω_c) corresponding to the transverse magnetic field B_0. It has been observed that g_R is nearly independent of feeble magnetic fields (i.e., in the imposed cyclotron frequency regime $\Omega_c\le10^{13}$ s^{-1}). However, g_R increases very rapidly for cyclotron frequencies above 2×10^{13} s^{-1} corresponding to a magnetic field greater than or equal to 1.0 T and attains explosive proportions.

To obtain a quantitative estimation about the effect of the large transverse magnetostatic field on the Raman instability, we can observe from Eqs. (15) and (16) that the threshold value of the pump can be greatly reduced by increasing the value of B_0. The effect of B_0 on the Raman growth rate can be studied from Eq. (15) and is found for chosen physical constants as

$$\frac{(g_R)_{B\neq0}}{(g_R)_{B=0}}\approx2\times10^2 , \tag{17}$$

at $k=10^7$ m^{-1}; $E_0=10^7$ Vm^{-1} with $\Omega_c=0.9\omega_0$ which is quite large.

In order to explore the possibility of the occurrence 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 $[g(\omega_S)_{\text{eff}}]L>30$,[13] L being the cell length. However, for pulse duration $\tau_P\ge10^{-9}$ s, the cell length can be taken to be equal to the interaction length x. It may be noted that in order to reach the SRS threshold, either the beam has to be focused in a nonlinear medium or a sufficient interaction length has to be provided by directing the beam into a light guide. In both cases 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 $\approx30/g_R(\omega_S)$. The external magnetic field induces a certain degree of nonlinearity into the propagating electromagnetic waves, which is favorable for the occurrence of phase conjuga-

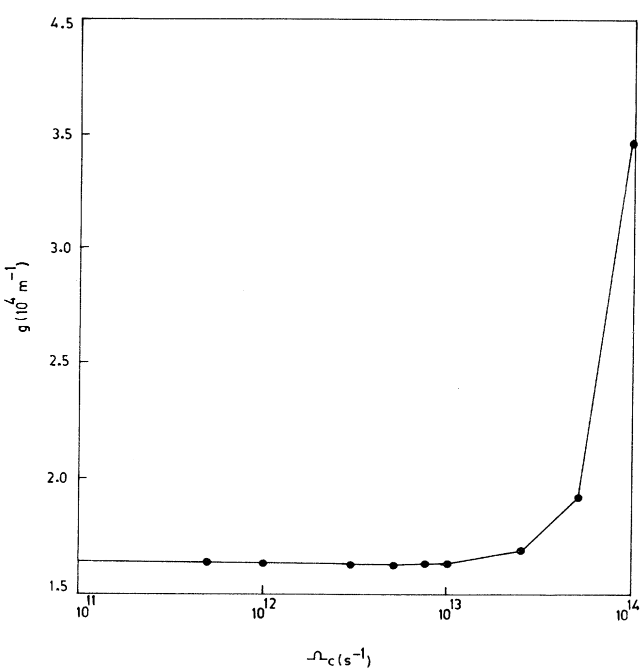

FIG. 1. Variation of the growth rate g_R with the cyclotron frequency Ω_c at $k=5\times10^6$ m^{-1} and $E_0=10^8$ V m^{-1}.

tion. 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 the excitation intensity, within the limit of the damage threshold.

IV. CONCLUSIONS

The present analytical investigation of SRS has yielded interesting results which can be categorized as follows.

(1) The third-order Raman susceptibility is found to be strongly dependent not only on the incident pump frequency ω_0 but also on the cyclotron frequency ω_c corresponding to the transverse magnetic field B_0.

(2) In a magnetoactive semiconductor plasma one can obtain a considerable growth rate of the Stokes component Raman mode at a much smaller values of the pump amplitude E_0. If B_0 is taken so large that $\omega_c^2 > \omega_0^2$ then from Eq. (15) one can note that the gain due to stimulated Raman scattering is not obtained.

(3) The possibility of OPC-SRS has been studied from the analytical investigation of the steady-state Raman growth and it has been found that the presence of a magnetostatic field effectively reduces the OPC threshold conditions.

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