Theory of stimulated Raman and Brillouin scattering in noncentrosymmetric crystals

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Stimulated Raman and Brillouin scattering (SRS and SBS) phenomena have been investigated analytically with use of density-matrix formulations and assuming parity indefiniteness for the eigenfunctions of the energy eigenstates of a two-level system in noncentrosymmetric (NCS) crystals. In crystals, when a molecule is excited, the disturbance travels through the crystal as a vibrational wave (phonon mode). We have considered the scattering of the incident electromagnetic wave by phonon modes on a common footing, assuming that vibrations should correspond to optical phonons for SRS and to acoustic phonons for SBS. The retardation effect due to finite $\chi^{(2)}$ in NCS crystals is found to play a more significant role in the case of SRS than for SBS. The classical differential polarizability $(\partial\alpha/\partial u)_0$ that causes SRS has been determined from the present formulations in terms of the material constants of the crystal and the photon pump energy. Applying Heisenberg's uncertainty relation, we have also obtained a general formula for the determination of the electrostrictive coefficient γ that is responsible for SBS in crystals. We find $(\partial\alpha/\partial u)_0/\gamma \sim 10^{-8}$ with $\gamma \sim 10^{-10}$–10^{-11} mks units for important III-V semiconductors. Both Raman and Brillouin gain constants are studied over a wide frequency spectrum in the semiconductors. The ratio between the two gain constants indicates that for the same pump intensity SBS exhibits higher gain than SRS by a magnitude $\sim\omega_{OP}/\omega_{AP}$.

I. INTRODUCTION

Stimulated Raman and stimulated Brillouin scattering (SRS and SBS) have been two of the most extensively studied nonlinear-optical processes in solid materials since the advent of lasers. Since their discoveries,[1,2] these processes have been studied theoretically in many ways in order to gain clear insight into the physical mechanisms that are expected to be present in solids. These treatments may be classified into three groups: classical electromagnetic, quantum mechanical, and semiclassical. Each treatment has its own merits and drawbacks. The classical electromagnetic approach[3] can yield valuable information regarding the threshold value of the pump electric field for the onset of both Raman and Brillouin instability in unmagnetized as well as strongly magnetoactive semiconductors,[4–6] but it is unable to provide any information on the transition mechanisms as well as the range of the pump energy under which SRS and SBS can be achieved most efficiently. Quantum-mechanical formulations[7] based upon the energy eigenstates describe the emission and absorption of photons, but phase information is lost and the theory cannot predict threshold conditions. The semiclassical treatment[8–10] has so far been regarded as the best approach in dealing with both SRS and SBS in crystals. This is essentially based on density-matrix formalisms where the electrons are treated quantum mechanically, while both photons and phonons are described classically. This approach[8–10] can explain the results obtainable from an electromagnetic treatment by incorporating the nonlinear current density arising from nonlinearity in the electron density; moreover, it gives insight into the transition mechanisms. The retardation effects arising from the lack of inversion symmetry in noncentrosymmetric (NCS) crystals are very important and must be included in the formulation for the third-order optical susceptibility $\chi^{(3)}$.[11] The electromagnetic approach cannot deal with such effects, while the semiclassical approach has been found to be useful in the study of retardation effects, incorporating $\chi^{(3)}$. Very recently, we have studied analytically the second-order susceptibility $\chi^{(2)}$ in NCS crystals.[12] We noted that the NCS effect could be very significant in the estimation of $\chi^{(3)}$, especially when the investigation is made in the near-band-gap resonant transition regime in NCS crystals. $\chi^{(3)}$ and the third-order material response to a suitable laser have become the most important tools in the analysis of a wide range of nonlinear-optical processes, such as optical bistability,[13] degenerate four-wave mixing,[14] phase conjugation,[15] etc., in addition to SRS and SBS. In this paper we have confined our attention to SRS and SBS arising from the first-order Stokes component of the scattered electromagnetic wave $(\omega_s, \vec{k}_s)$. A generalized attempt has been made to investigate both these processes in NCS semiconducting crystals on the basis that SRS is caused by the coupling of the pump-laser photons $(\omega_p, \vec{k}_p)$ with optical-phonon (OP) modes, while SBS originates from the coupling of the former with the acoustic-phonon (AP) modes in the crystals.[7]

In all crystalline solids, SRS arises from the finite differential polarizability, while SBS arises from the finite electrostriction of the medium. The response of electrons and nuclei to the laser irradiation changes the dielectric constant of the medium. Weak nuclear motions treated by the electromagnetic force of the radiation give rise to phonon modes $(\omega_v, \vec{k}_v)$ in the crystal. The stimulated scattering of the incoming radiation by these phonon modes results in SRS and SBS. Due care has been taken

to satisfy the rotating-wave approximation throughout the analysis.

In Sec. II we have derived a general expression for the third-order optical susceptibility $\chi^{(3)}$ that describes both the SRS and SBS of the Stokes component of the scattered electromagnetic wave in an NCS crystal. In Sec. III we deal with the phenomenon of SRS and the Raman gain constant in the crystals, taking into account the retardation effect. SBS and the appropriate gain constant is the subject of Sec. IV. The most important conclusions of the present investigation are listed in Sec. V.

II. THEORETICAL FORMULATION OF $\chi^{(3)}$

Here we address the formulation of $\chi^{(3)}$ for the Stokes component of the scattered electromagnetic wave to study both SRS and SBS. The scattering is the consequence of the phonon-mode generation (optical and acoustic for the SRS and the SBS, respectively) in the crystal in the presence of laser excitation. It may be noted in this connection that in a crystal molecular vibrations no longer remain localized and can travel throughout the material, which makes it necessary to replace conventional treatment of SRS due to molecular vibrations by an approach that explains SRS on the basis of existence of OP modes in the crystals. The generalized phonon mode can be represented in the one-dimensional configuration as

$$\frac{\partial^2 u(x,t)}{\partial t^2}+\frac{C_a}{d}\frac{\partial^2 u(x,t)}{\partial x^2}+\omega_0^2 u(x,t)+2\Gamma\frac{\partial u(x,t)}{\partial t}=\frac{\langle F\rangle}{d}\ . \quad (1)$$

Here,

$$u(x,t)\ (=u\exp[i(\omega_v t-k_v x)])$$

denotes the relative displacement of the nuclear positions within the lattice. C_a is the linear elastic modulus of the crystal and d is the mass density. ω_0 is the phonon frequency at $k_v=0$; Γ takes into account the damping and is introduced here phenomenologically as a constant parameter. $\langle F\rangle$ is the generalized force per unit volume experienced by the nuclei due to the electromagnetic pump wave and is defined as $\langle F\rangle=N\langle f\rangle$, with

$$\langle F\rangle=\langle f^{(1)}\rangle E+\langle f^{(2)}\rangle E^2+\langle f^{(3)}\rangle E^3+\cdots\ , \quad (2)$$

and N is the number of elementary cells per unit volume assuming that each cell contains one electron.[16] Thus with a_l being a lattice constant, one can consider $N=a_l^{-3}$. From expression (2), it is clear that for a centrosymmetric system, the odd-order force components, viz., $\langle f^{(1)}\rangle E$, $\langle f^{(3)}\rangle E^3, \ldots, =0$, while these are finite for an NCS system.

In the case of the OP branch, the phonon frequency $\omega_v=\omega_{\rm OP}$ and is very large in comparison with $k_v(C_a/d)^{1/2}$, as $(C_a/d)^{1/2}$ is the acoustic velocity in the crystal. This enables one to neglect the second term on the left-hand side of (1), yielding

$$\frac{\partial^2 u(x,t)}{\partial t^2}+\omega_0^2 u(x,t)+2\Gamma\frac{\partial u}{\partial t}=\frac{\langle F\rangle}{d} \quad (3)$$

for the OP-mode propagation in the crystal. For the AP branch, $\omega_v=\omega_{\rm AP}$ and $\omega_0=0$ since at $k_v=0$ such a mode does not exist. This results in

$$\frac{\partial^2 u(x,t)}{\partial t^2}+\frac{C_a}{d}\frac{\partial^2 u(x,t)}{\partial x^2}+2\Gamma\frac{\partial u(x,t)}{\partial t}=\frac{\langle F\rangle}{d}\ . \quad (4)$$

The phonon modes thus induced due to finite $\langle F\rangle$ modify the dielectric constant and, hence, the susceptibility of the medium.

We consider the validity of the formulation of the scattering of light by phonons based upon Placzek's approximation[17] that the pump frequency is too high for ions to be affected. Thus the scattering is produced by the modulation of electronic polarizability due to lattice vibrations. Within this approximation, we have neglected the direct interaction between the electromagnetic pump and the ions. We have dealt with the problem semiclassically, where the electronic system is treated quantum mechanically and both the pump and vibrational fields are described classically by waves.

Considering the interaction of the electrons with the pump photons as well as with the phonons in the crystal, the interaction Hamiltonian $H_{\rm int}$ in terms of the present one-dimensional configuration can be written as

$$H_{\rm int}=H_{e\text{-}v}+H_{e\text{-}r}\ , \quad (5)$$

with

$$H_{e\text{-}v}=-fu\exp[i(k_v x-\omega_v t)] \quad (6)$$

and

$$H_{e\text{-}r}=-\mu E_0\exp[i(k_p x-\omega_p t)]\ . \quad (7)$$

$H_{e\text{-}v}$ and $H_{e\text{-}r}$ are the interaction Hamiltonians for the electron-vibration and electron-radiation interactions, respectively, $\vec{\mu}$ is the dipole moment matrix,

$$\vec{\rm E}=\vec{\rm E}_0\exp[i(k_p x-\omega_p t)]\ ,$$

$\vec{\rm E}_0$ being the electric field amplitude of the pump wave. Both $\vec{\mu}$ and $\vec{\rm E}$ are considered to be parallel to each other (along the X axis). f is the generalized force given by $f=\langle F\rangle/N$.

For a two-level electronic system, we represent the ground level by $|\,g\,\rangle$ and assume it to be completely occupied, while assuming the excited level $|\,n\,\rangle$ to be empty. The total Hamiltonian for such a system when irradiated by a laser beam will consist of a part that determines the unperturbed energy levels of the atomic system, with $H_{\rm int}$ accounting for the interactions in the system in the presence of the applied fields.

The ensemble average of the induced dipole moment is expanded as

$$\langle\mu\rangle=\langle\mu^{(1)}\rangle+\langle\mu^{(2)}\rangle+\langle\mu^{(3)}\rangle+\cdots\ , \quad (8)$$

where the ensemble averages of both the generalized force and dipole moment of various orders are defined as

$$\langle f^{(j)}\rangle={\rm Tr}(f\rho^{(j)}) \quad (9a)$$

and

$$\langle\mu^{(j)}\rangle={\rm Tr}(\mu\rho^{(j)})\ , \quad (9b)$$

with j a positive integer.

In order to evaluate the matrix elements of f and μ, we consider the one-dimensional Schrödinger equation given by

$$\left[-\frac{\hbar^2}{2m_r}\frac{\partial^2}{\partial x^2}+U(x)\right]\psi(x)=\epsilon\psi(x)\ . \tag{10}$$

The ensemble average of $\langle F\rangle$ and $\langle\mu\rangle$ can be written as

$$\langle F\rangle=A\int\psi_n^* x^{-2}\psi_g dx \tag{11a}$$

and

$$\langle\mu\rangle=-e\int\psi_n^* x\psi_g dx\ , \tag{11b}$$

with A having the dimension of (mass)[(time)$^{-2}$]. The origin of the characteristic representation of $\langle F\rangle$ by Eq. (11a) is contained in the definition of $\langle F\rangle$ as the generalized force per unit volume having the dimension (mass)[(length)$^{-2}$][(time)$^{-2}$].

For centrosymmetric systems, $U(x)$ possesses definite parities such that

$$f_{nn},f_{gg}\neq 0,\quad f_{ng},f_{gn}=0$$

and

$$\mu_{nn},\mu_{gg}=0,\quad \mu_{ng},\mu_{gn}\neq 0$$

for opposite parities of the wave function ψ_g and ψ_n. The NCS crystals lack inversion symmetry, and hence the wave functions ψ_g and ψ_n will have mixed (rather indefinite) parities and, consequently, one finds all the matrix elements of f and μ to be finite.[12]

The density-matrix elements of various orders can be obtained by using a straightforward perturbation expansion of the density matrix of the material system as

$$\rho_{ng}^{1}(\omega_p)=H_{ng}(\omega_p)/\hbar(\omega_p-\omega_{ng}-i\Gamma)\ , \tag{12a}$$

$$\rho_{ng}^{(2)}(\omega_s)=\frac{H_{ng}(\omega_p)[H_{nn}(\omega_v)-H_{gg}(\omega_v)]}{[\hbar^2(\omega_s-\omega_{ng}-i\Gamma)(\omega_p-\omega_{ng}-i\Gamma)]}+\frac{H_{gn}(\omega_v)[H_{nn}(\omega_p)-H_{gg}(\omega_p)]}{[\hbar^2(\omega_s-\omega_{ng}-i\Gamma)(\omega_v-\omega_{ng}-i\Gamma)]}\ , \tag{12b}$$

$$\rho_{gg}^{(2)}(\omega_v)=-\rho_{nn}^{(2)}(\omega_v)=\frac{H_{gn}(\omega_s)H_{ng}(\omega_p)}{\hbar^2(\omega_v-i\Gamma)}\left[\frac{1}{\omega_s-\omega_{ng}-i\Gamma}-\frac{1}{\omega_p-\omega_{ng}-i\Gamma}+\text{c.c.}\right]\ , \tag{12c}$$

with ω_s $(=\omega_p-\omega_v)$ and $\vec{k}_s$ $(=\vec{k}_p-\vec{k}_v)$ representing the Stokes component of the scattered electromagnetic wave. In obtaining Eqs. (12), we have taken into account the role of various relaxation processes owing not only to the photons and phonons but also to the applied random fields through the introduction of a phenomenological damping parameter Γ into the density-matrix formalisms which is assumed to be the same as that introduced in Eq. (1). Γ plays a significant role, especially when the NCS crystal is subjected to laser irradiation in the near-band-gap resonant transition regime. Other terms are standard and defined elsewhere.[12] It must be noted that, in deriving the above set of equations, we have neglected the complex-conjugate terms in order to avoid mathematical complexities without loss of generality.

One obtains, from the above formulations,

$$\langle f^{(1)}\rangle\langle E\rangle=\frac{f_{gn}\mu_{ng}E_v}{\hbar\Omega_v}\ , \tag{13}$$

$$\langle f^{(2)}\rangle\langle E^2\rangle=\frac{|\mu_{ng}|^2(f_{nn}-f_{gg})E_0E_1^*}{\hbar^2\Omega_s\Omega_p}+\frac{\mu_{ng}f_{gn}(\mu_{nn}-\mu_{gg})\omega_vE_0E_1^*}{\hbar^2\Omega_s\Omega_p\Omega_v}\ . \tag{14}$$

Similarly,

$$\langle\mu^{(1)}\rangle=\frac{|\mu_{ng}|^2E_0}{\hbar\Omega_p} \tag{15}$$

and

$$\langle\mu^{(2)}\rangle=\frac{|\mu_{ng}|^2(f_{nn}-f_{gg})\mu^*E_0}{\hbar^2\Omega_s\Omega_p}+\frac{\mu_{ng}f_{gn}(\mu_{nn}-\mu_{gg})\omega_vE_0u^*}{\hbar^2\Omega_s\Omega_p\Omega_v}\ , \tag{16}$$

where

$$\Omega_{p,s,v}=\omega_{p,s,v}-\omega_{ng}-i\Gamma\ .$$

$\langle f^{(1)}\rangle\langle E\rangle$ in Eq. (13) describes the first-order force component which is finite only in the NCS crystals with $f_{ng}\neq 0$. This force is related to the crystal deformation potential, piezoelectric property, etc. The classical counterpart of Eq. (13) for a real piezoelectric crystal is described by $\beta\partial E/\partial x$ with β a piezoelectric constant. Equation (14), one the other hand, represents the second-order force component which arises from the application of the electromagnetic field on the crystal. This force consists of two parts. The first part corresponds to the second-order force arising in a centrosymmetric system, and the second one denotes the contribution from the NCS properties of the system. Owing to the application of the electromagnetic field, the ions within the lattice move into nonsymmetrical positions, usually producing a contraction in the direction of the field and an expansion across it. The electrostatic force thus produced is the origin of differential polarizability and electrostriction in the medium.

Equation (15) describes the induced laser polarization and consequent linear refraction and absorption phenome-

na in the crystals, and is independent of the polar nature of the medium. $\langle \mu^{(2)} \rangle$ as given by Eq. (16) is comprised of two parts, the former being finite in any crystal having centrosymmetric characteristics, while the latter is an additional contribution due to the NCS property of the crystal.

The set of Eqs. (13)—(16) arises as a result of the response of electrons to the phonon and pump fields. It is the NCS property of the material systems that is responsible for the finite strain-associated electric field, and hence the first-order electronic response due to the phonon mode is finite for such media only. Although Eqs. (14) and (16) are similar in nature, in the near-band-edge region, $\langle \mu^{(2)} \rangle \gg \langle f^{(2)} \rangle \langle E_0 E_1^* \rangle$. It can be further observed that the NCS contributions to these terms are equal for both ω_v and ω_s.

To our knowledge, no systematic analysis of the role of the NCS contribution to $\langle \mu^{(2)} \rangle$ has been undertaken so far in the study of SRS and SBS, although its importance has been mentioned very often in the available literature.[11,18]

We now consider the equation for the Stokes component of the scattered electromagnetic wave, the frequency (ω_s) and wave vector ($\vec{k}_s$) of which are obtainable from the energy- and momentum-conservation relations in the present investigation as

$$\hbar\omega_p = \hbar\omega_s + \hbar\omega_v \tag{17a}$$

and

$$\hbar\vec{k}_p = \hbar\vec{k}_s + \hbar\vec{k}_v \ , \tag{17b}$$

yielding $\omega_s = \omega_p - \omega_v$ and $\vec{k}_s = \vec{k}_p - \vec{k}_v$. When the crystals are irradiated by infrared or near-infrared lasers, one can easily neglect the pump photon momentum and, consequently, it can be assumed that $\vec{k}_v = -\vec{k}_s$ with $|\vec{k}_v| = |\vec{k}_s| = |\vec{k}|$ and $|\vec{k}_p| \sim 0$.

The electromagnetic-wave equation for the Stokes component can be written as

$$\vec{\nabla}\times\vec{\nabla}\times\vec{E}_s - \frac{\omega_s^2}{c_l^2}\vec{E}_s = \frac{1}{c_l^2}\frac{\partial^2\vec{P}(t)}{\partial t^2} \ , \tag{18}$$

with $c_l^2 = c^2/\epsilon$, c being the velocity of light in free space, E_s the amplitude of the wave, and $\vec{P}(t)$ the total induced polarization. The nonlinear contribution of the induced current density to Eq. (18) is found to be negligible and increases the mathematical complexities. Although its incorporation can yield valuable information regarding the threshold conditions for SBS and SRS,[2–4] our basic aim is to obtain the contribution of noncentrosymmetricity to the induced polarization which causes these scattering phenomena, and, hence, the nonlinear current density has been neglected. The total induced polarization $\vec{P}(t)$ for an NCS crystal can be described by

$$\vec{P}(t) = \epsilon_0(\chi^{(1)} + \chi^{(2)}|E| + \chi^{(3)}|E^2| + \cdots)\vec{E}(t) \ , \tag{19}$$

where $\chi^{(1)}, \chi^{(2)}, \chi^{(3)}, \ldots$ are various orders of susceptibility. $\chi^{(1)}$ corresponds to the linear component, while $\chi^{(2)}$ is the first-order nonlinearity in the optical susceptibility that describes various passive optical processes. $\chi^{(2)}$ also accounts for the retardation effects on the higher-order nonlinearities. Our principal aim is to study $\chi^{(3)}$ and the retardation effect of $\chi^{(2)}$ on it in NCS crystals.

To obtain $\chi^{(3)}$ from the above formulations, we substitute the value of u^* from Eq. (1) in Eq. (16). It can be seen that if only the first-order force $\langle f^{(1)} \rangle \langle E \rangle$ is considered in Eq. (1), the nonlinear polarization obtained gives rise to $\chi^{(2)}$.[12] The substitution of the second-order force $\langle f^{(2)} \rangle \langle E_0 E_1^* \rangle$ for $\langle f \rangle$ in Eq. (1) explains the origin of $\chi^{(3)}$ in the crystal possessing parity indefiniteness of electronic eigenstates. Thus, one obtains

$$\chi^{(3)} = \frac{N^2 C}{\epsilon_0 dD}\left[|\mu_{ng}|^2(f_{nn}-f_{gg})^2 + \frac{f_{gn}^2(\mu_{nn}-\mu_{gg})\omega_v^2}{(\omega_{ng}^2+\Gamma^2)} - \frac{2\mu_{ng}f_{gn}(\mu_{nn}-\mu_{gg})(f_{nn}-f_{gg})\omega_v}{(\omega_{ng}^2+\Gamma^2)^{1/2}}\right] , \tag{20}$$

with

$$C = \frac{|\mu_{ng}|^2}{\hbar^4|\Omega_s|^2|\Omega_p|^2}$$

and

$$D = [-\omega_v^2 + k^2 v_s^2 + \omega_0^2 + 2i\omega_v\Gamma] \ ,$$

where we have neglected ω_v in comparison with ω_{ng}, and have also neglected the complex-conjugate terms in obtaining $\langle f \rangle$ and $\langle \mu \rangle$ for the sake of mathematical simplification. As can be noticed from the above expression, the first term is finite for a CS (centrosymmetric) system. The second term is a positive contribution from the NCS property of the system and its contribution is considered to be very small in comparison with the third term that represents the retardation effect on $\chi^{(3)}$. The first term in Eq. (20) corresponds to the Raman susceptibility arising from the finite differential polarizability if we choose $D = (\omega_0^2 - \omega_{\rm OP}^2 + 2i\omega_{\rm OP}\Gamma)$ and ω_v ($\gg kv_s$) replaced by $\omega_{\rm OP}$. The same factor can explain the SBS phenomena due to electrostriction by defining $D = k^2 v_s^2 - \omega_{\rm AP}^2 + 2i\omega_{\rm AP}\Gamma$, where $\omega_{\rm AP}$ corresponds to the acoustic-phonon frequency. These replacements are compatible with the phonon propagation represented by Eqs. (2) and (3). It must be remembered here that such a simple relationship between the two (Raman and Brillouin) susceptibilities could be obtained only because of the assumption that each unit cell contains only one electric dipole and one simple harmonic oscillator.

III. SRS AND RAMAN GAIN CONSTANT

As discussed in Sec. II, here we have attempted to bring forth an interesting correspondence between the classical finite differential polarizability $(\partial\alpha/\partial u)_0$, which is re-

garded as the origin of SRS in an electromagnetic treatment[3,5] and $f^{(2)}$ in our analysis. It is well known that the electrostatic, stored energy density responsible for the occurrence of SRS in crystals is proportional to $|E|^2$, and can be represented also as $N\langle f^{(2)}\rangle\langle E^{(2)}\rangle u$, with $\langle f^{(2)}\rangle\langle E^2\rangle$ given by Eq. (14). Upon neglecting the NCS contribution in Eq. (14), one can obtain a simple expression for $(\partial\alpha/\partial u)_0$ in the form

$$\left[\frac{\partial\alpha}{\partial u}\right]_0=\frac{2|\mu_{ng}|^2(f_{nn}-f_{gg})}{\epsilon_0\hbar^2\Omega_p\Omega_s}\ . \tag{21}$$

From this equation we note that the differential polarizability is not a constant, but instead depends very much on the pump-laser frequency as well as the transition frequency. This is because of the fact that $\Omega_p=\omega_p-\omega_{ng}-i\Gamma$, and $|\mu_{ng}|^2=|ep_{ng}(0)/m_r\omega_g|^2$, with $p_{ng}(0)$ and m_r being the band-to-band transition momentum matrix and free-electron mass, respectively; ω_g is the crystal band-gap frequency. $(\partial\alpha/\partial u)_0$ behaves like a physical constant of the crystal only when one chooses off-resonant transitions with ω_{ng} $(=\omega_g)\gg\omega_p$, $\omega_{\rm OP}$, and Γ. During near-resonant transitions ($\omega_p\sim\omega_{ng}\sim\omega_s$), one can achieve a considerably large differential crystal polarizability. This leads one to infer that SRS would be much more efficiently achieved under near-resonant transitions in the crystal, irrespective of its symmetry properties. One can also observe from the same equation [Eq. (21)] that $(\partial\alpha/\partial u)_0$ will be significantly small for $\omega_p\gg\omega_{ng}$ when $(\partial\alpha/\partial u)_0$ varies nearly as ω_p^{-2} (under such circumstances, $\omega_{\rm OP}\ll\omega_p$).

Remembering the above correspondence, we now proceed to study SRS and the consequent Raman gain constant in a crystal exhibiting a lack of inversion symmetry. The Raman susceptibility can accordingly be obtained from Eq. (20) as

$$\chi_R=\frac{N|\mu_{ng}|^3(f_{nn}-f_{gg})}{\hbar^4|\Omega_s|^2|\Omega_p|^2\epsilon_0 dD_R}\times\left[\mu_{ng}(f_{nn}-f_{gg})-\frac{2f_{gn}(\mu_{nn}-\mu_{gg})\omega_{\rm OP}}{\omega_{ng}}\right], \tag{22}$$

with

$$D_R=\omega_0^2-\omega_{\rm OP}^2+2i\omega_{\rm OP}\Gamma\ .$$

In obtaining the above expression, we have neglected the positive contribution of the NCS property. The first part of Eq. (22) corresponds to the Raman susceptibility in the crystal when the NCS effect is absent or neglected, and has been studied extensively.[8–10] The second component accounts for the contribution of the NCS property to χ_R. The same component can be expressed in terms of $\chi^{(2)}$ and, consequently, can be designated the retardation effect on Raman susceptibility due to finite $\chi^{(2)}$. Examining expression (22), it is seen that the NCS effect would be higher in crystals having a stronger polar nature. If the transition frequency is in the vicinity of the optical-phonon frequency, the NCS effect becomes very significant [as $\omega_{\rm OP}\gg\Gamma$, e.g., $\omega_{\rm OP}\gtrsim 10^2\,\Gamma$ (Ref. 3)], although the percentage effect remains the same.

We now estimate the Raman gain constant $|g(\omega_s)|_R$ of the Stokes mode, which is related to the imaginary part of χ_R $(=\chi_{Rr}+i\chi_{Ri})$ through[3]

$$|g(\omega_s)|_R=-\frac{k}{2c_l}\chi_{Ri}|E|^2\ . \tag{23}$$

Substituting the value of χ_{Ri} from the imaginary part of Eq. (22), one finds

$$|g(\omega_s)_R=\frac{kN|\mu_{ng}|^3(f_{nn}-f_{gg})\omega_{\rm OP}\Gamma|E|^2}{2\epsilon_l\hbar^4|\Omega_s|^2|\Omega_p|^2\epsilon_0 d|D_R|^2}\times\left[\mu_{ng}(f_{nn}-f_{gg})-\frac{2f_{gn}(\mu_{nn}-\mu_{gg})\omega_{\rm OP}}{\omega_{ng}}\right], \tag{24}$$

with $\epsilon=\epsilon_0\epsilon_l$, ϵ_0 and ϵ_l being the permittivity of free space and lattice dielectric constant, respectively. From Eq. (24), one can note that the effect of the NCS property (represented by the second part) will be higher in crystals having a stronger polar nature. It is also significant in narrow-band-gap semiconductors. $|g(\omega)|_R$ is found to depend very strongly on the pump frequency through the terms $|\Omega_p|^2$ and $|\Omega_s|^2$. For off-resonant transitions much below the band edge, both $|\Omega_p|^2$ and $|\Omega_s|^2$ are simply equal to ω_{ng}^2 (because $\omega_s=\omega_p-\omega_{\rm OP}$, $\Gamma\ll\omega_{\rm OP}\ll\omega_p\ll\omega_{ng}$), such that the Raman gain reduces to an appreciably low constant value. During off-resonant transitions far above the band edge ($\omega_p\gg\omega_{ng}$), $|g(\omega_s)|_R$ varies almost as ω_p^{-4}, and will be even smaller than that obtainable under the former transition regime. The most interesting situation occurs when the NCS crystal is subjected to near-band-gap resonant laser excitation. This yields a giant gain constant of the Stokes component of the scattered electromagnetic wave indicating its potentiality in the achievement of a tunable laser source at frequency $\omega_s=\omega_p-\omega_{\rm OP}$. The optical-phonon mode considered in the study of SRS can be treated either as the longitudinal or the transverse OP mode, the frequencies being easily obtainable from a knowledge of the Debye temperatures of the crystals as well as the usage of Lyddane-Sachs-Teller relation.

IV. SBS AND BRILLOUIN GAIN CONSTANT

We proceed, in an almost similar way as followed in Sec. III, to study SBS and the Brillouin gain constant of the Stokes mode, modifications being made only in the replacement of D in Eq. (20) by D_B [obtainable from Eq. (3)]. As discussed in Sec. II, the electrostrictive force acting per unit volume of the crystal,[3] $-(\gamma/2)(\partial/\partial x\langle E^2\rangle)$, corresponds to $\langle f^{(2)}\rangle\langle E^{(2)}\rangle$, given by Eq. (14), ω_v being replaced by $\omega_{\rm AP}$. The difference between the two expressions is that the latter includes the NCS effect. Thus, one can write

$$-\frac{\gamma}{2}\frac{\partial}{\partial x}\langle E^2\rangle=\frac{N|\mu_{ng}|^2(f_{nn}-f_{gg})\langle E^2\rangle}{\hbar^2\Omega_p\Omega_s}\ . \tag{25}$$

Equation (25) can be used to estimate the value of γ. In

order to achieve this goal, we have applied Heisenberg's uncertainty relation for the linear momentum and position coordinate in the present quantum-mechanical formulations. It is well known[19] that for a linear dimension u of a region in which a particle can move, the uncertainty in the coordinate is $\sim u$; the uncertainty in the momentum, and therefore the order of magnitude of the momentum itself, is $p_u \sim \hbar/u$. Now, $-(\gamma/2)(\partial/\partial x)\langle E^2\rangle$ can be written as

$$\left\langle -\frac{i\gamma}{2\hbar} p_u \frac{\partial u}{\partial x} u \right\rangle \quad \text{with } p_u = -i\hbar \frac{\partial}{\delta u} .$$

Application of the uncertainty relation in the present context yields $\partial u/\partial x = 1$ such that

$$\gamma = \left[\frac{2N |\mu_{ng}|^2 (f_{nn} - f_{gg}) u}{\hbar^2 \Omega_p \Omega_s} \right]_{\mathrm{QL}} . \tag{26}$$

The suffix QL denotes the quantum limit. For a particular crystal in the off-resonant—transition regime with $\omega_{ng} > \omega_p$ and ω_s, one can obtain a constant value of γ due to the fact that the quantity $(f_{nn} - f_{gg})u$ can be expressed as[10] $\eta\hbar\omega_{\mathrm{AP}}$ with η ($\sim\frac{1}{2}$ to $\frac{1}{10}$) being a measure of the perturbing strength of the vibrational wave on the electronic system. Thus the electrostrictive coefficient γ is obtained in terms of the physical constants of the crystal as

$$\gamma = \left[\frac{2 |\mu_{ng}|^2 \eta \omega_{\mathrm{AP}}}{\hbar \omega_{ng}^2 a_l^3} \right]_{\mathrm{QL}} . \tag{27}$$

The concept of finite electrostrictive force is applicable only for $|k| > 0$. One can also note that under the quantum limit as discussed earlier, $p_u u \sim \hbar$, such that $k_v \sim 1/u$, and for a lattice, u can be replaced by a_l. This enables one to treat ω_{AP} as a constant in the quantum limit, given by $\omega_{\mathrm{AP}} = k_v v_s = v_s/a_l$, and one obtains

$$\gamma = \frac{2\pi\eta |ep_{ng}(0)/m_r\omega_g|^2 v_s}{\hbar \omega_g^2 a_l^4} , \tag{28}$$

where we have replaced $|\mu_{ng}|^2$ by $|ep_{ng}(0)/m_r\omega_g|^2$. From Eq. (28), one can determine the value of γ for any crystal, remembering that it is a constant only under off-resonant—transition situations. The numerical estimations for the important III-V semiconductors yield values of $\gamma \sim 10^{-10}$ to 10^{-11} mks units. This is in agreement with the value quoted by Yariv.[3] Its variation with respect to pump frequency can be studied from Eq. (26). Our next objective is to obtain the susceptibility and gain constant of the Brillouin mode. For this purpose, we use Eq. (20) with N being replaced by a_l^{-3}, ω_v by ω_{AP}, and choosing $|k| > 0$. Thus one obtains

$$\chi_B = \frac{|\mu_{ng}|^3 (f_{nn} - f_{gg})}{\epsilon_0 d D_B a_l^6 \hbar^4 |\Omega_p|^2 |\Omega_s|^2} \times \left[\mu_{ng}(f_{nn} - f_{gg}) - \frac{2 f_{gn}(\mu_{nn} - \mu_{gg})\omega_{\mathrm{AP}}}{\omega_{ng}} \right] \tag{29}$$

with

$$D_B = -\omega_{\mathrm{AP}}^2 + k^2 v_s^2 + 2i\Gamma\omega_{\mathrm{AP}} .$$

The Brillouin gain constant is

$$|g(\omega_s)|_B = \frac{|\mu_{ng}|^3 (f_{nn} - f_{gg}) |E|^2}{\epsilon_0 d a_l^3 \hbar^4 |\Omega_p|^2 |\Omega_s|^2 v_s \Gamma} \times \left[\mu_{ng}(f_{nn} - f_{gg}) - \frac{2 f_{gn}(\mu_{nn} - \mu_{gg})\omega_{\mathrm{AP}}}{\omega_{ng}} \right] , \tag{30}$$

where it is assumed that $\omega_{\mathrm{AP}}^2 = k^2 v_s^2$.

The NCS effect reduces the gain constant in the case of SBS almost like that for SRS. This retardation effect on $|g(\omega_s)|_B$ is found to be less than that on $|g(\omega_s)|_R$ [Eq. (24)] as $\omega_{\mathrm{AP}} < \omega_{\mathrm{OP}}$. The qualitative behavior of $|g(\omega_s)|_B$ on the pump frequency is similar to that in the case of $|g(\omega_s)|_R$.

When the effect of lack of inversion symmetry is neglected in a crystal, the gain constant of the Stokes component of the scattered electromagnetic wave is found to be larger for SBS than that for SRS, the ratio being $\sim \omega_{\mathrm{OP}}/\omega_{\mathrm{AP}}$.

V. CONCLUSIONS

The present analytical investigation of SRS and SBS has yielded the following interesting results which were not pointed out categorically in earlier theoretical attempts.

(i) The propagation behavior of the vibrational modes has been represented by a general phonon-mode wave equation with the optical and acoustic phonons being responsible for the SRS and the SBS, respectively.

(ii) The differential polarizability $(\partial\alpha/\partial u)_0$ that is normally treated as a constant parameter of the crystal for the study of SRS is found to depend on the pump photon energy. This dependence should be taken into account especially when the crystal is irradiated with a near-band-gap resonant laser beam to obtain a very large Raman gain of the Stokes mode. While drawing an analogy between the electromagnetic approach and the present semiclassical approach, we note that $(\partial\alpha/\partial u)_0$ should be modified due to the NCS properties of the crystal. An appropriate expression has been derived to estimate the same in the centrosymmetric medium.

(iii) Quite interestingly, in the case of SBS, the classical electrostrictive coefficient γ has been calculated using the present formulation. The universal validity of Heisenberg's uncertainty relation has been employed in deriving an expression for γ in terms of the material parameters of the crystal. An order-of-magnitude agreement with available values has also been achieved. The frequency dependence of γ is similar to that of $(\partial\alpha/\partial u)_0$.

(iv) A comparison between $(\partial\alpha/\partial u)_0$ and γ for a centrosymmetric crystal using the present formulation gives

$(\partial\alpha/\partial u)_0/\gamma \sim 10^{-8}$.

(v) Both Raman and Brillouin gain constants are affected by the finite retardation produced due to $\chi^{(2)}$ in the NCS crystals, and they should not be neglected in strongly polar crystals. This retardation effect is more important in the study of SRS than in SBS.

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