# Theory of resonant Brillouin scattering in population-inverted semiconductors. I. General formulation

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We have developed a general theory of the resonant Brillouin scattering in populationinverted semiconductors by solving the equation of motion for one-particle density matrix. The general expressions for the photoelastic constants obtained can be applied to (i) the analysis of the dispersion of photoelastic constants when the photon energies come very close to the energy gap and (ii) the analysis of the operation of acoustical distributed feedback tasers. We found the importance of a new contribution to the photoelastic constants directly induced by electrondensity fluctuation due to phonons. We also found the importance of the imaginary part of photoelastic constants, which has been overlooked so far.

#### I. INTRODUCTION

The possibility of the resonant enhancement of the Brillouin- and Raman-scattering cross sections for photons with energies close to the band gap in semiconductors has been predicted by Loudon.<sup>1</sup> After his prediction, the resonant enhancement and cancellation have been observed experimentally by many researchers. Experimental results reported so far on resonant Brillouin scattering are summarized as follows. Garrod and Bray<sup>2</sup> observed Brillouin scattering in GaAs and found the resonant enhancement and cancellation of the photoelastic constant  $p_{44}$ . The resonant enhancement of the Brillouin-scattering cross section in CdS has been observed by Pine.<sup>3</sup> Many people have observed this kind of resonant phenomena in CdS,<sup>4-7</sup> ZnO,<sup>7</sup> CdSe,<sup>8</sup> ZnSe,<sup>9-11</sup> ZnTe,<sup>12</sup> and GaP.<sup>13</sup> Also, Loudon's theory has been extended to take the effects of excitons<sup>14</sup> and polaritons<sup>15</sup> into account. The experimental results have been satisfactorily explained by the theories.

On the other hand, the resonant phenomena similar to the resonant Brillouin scattering have been observed in piezobirefringence. Feldman and Horowitz<sup>16</sup> have found the resonant cancellation of the piezobirefringence coefficient  $\pi_{11} - \pi_{12}$  in GaAs. Cardona and his co-workers<sup>17-20</sup> measured the dispersions of piezoelectroreflectance and piezobirefringence in many semiconductors and explained theoretically their experimental results. Berkowicz *et al.*,<sup>7</sup> Ando *et al.*,<sup>10</sup> and Tada *et al.*<sup>11</sup> have compared their results on the Brillouin scatterings by acoustical phonons with the results on piezobirefringence by static stresses. As a result, it was concluded that the dispersion of the Brillouinscattering cross section by low-frequency acoustical phonons is in good agreement with the dispersion of piezobirefringence coefficient.

Although the resonant enhancement and cancellation of the Brillouin-scattering cross section have been observed and explained theoretically, it is difficult to obtain the experimental data of the Brillouin scattering when the energies of incident and scattered photons come very close to the energy gap, since the photons are strongly absorbed by the electronic interband transitions in semiconductors. Even in such a case, however, it is possible to obtain the scattering cross section in population-inverted semiconductors, since the photons can be amplified instead of absorbed. One of the motivations of this paper is to give a theoretical basis for such an investigation. The second motivation is related to acoustical distributed feedback (ADFB) lasers<sup>21, 22</sup> proposed and demonstrated by us. In our ADFB lasers, a set of distributed Bragg reflectors (DBR's) are formed at both ends of GaAs wafers and surface acoustical waves (SAW's) are propagated on the planar region between DBR's. The light beam radiated by population inversion is deflected by DBR's and SAW's, if the Bragg conditions are satisfied. The photon energies of ADFB oscillation modes are very close to the energy gap of GaAs. Therefore, the values of photoelastic constants in the resonant Brillouin scattering in population-inverted semiconductors must be used for the theoretical analysis of the operation of ADFB lasers.

In this paper, we develop a general theory of the Brillouin scattering in population-inverted semicon-

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ductors by solving the equation of motion for oneparticle density matrix. The density-matrix formulation has merits that the effects of relaxation and of population inversion can be introduced into the basic equations in a straightforward manner. In Sec. II, we describe the assumptions and basic equations in our treatment. In Sec. III, we derive the expressions for first-order terms of density matrix, which correspond to the absorption (or amplification) of photons and the damping of phonons. Also, the increment of the dielectric constants due to the transitions between conduction and valence bands is calculated. In Sec. IV, we derive the expression for the contribution to photoelastic constants from the transition between conduction and valence bands, taking account of the effects of population inversion and relaxation. The physical meaning of each term in the expressions are discussed. We emphasize the importance of a new contribution to photoelastic constants directly induced by electron-density fluctuation due to phonons. We also point out the importance of the imaginary part of photoelastic constants, which has been overlooked so far. In Sec. V, the validity of the assumptions used in this paper is discussed. In Appendix A, the concept of a local equilibrium value of the density fluctuation due to low-frequency phonons introduced in Sec. II is discussed in some detail. The discussion on the screening effects of deformation potentials by free carriers is given in Appendix B.

In a separate paper,<sup>23</sup> we shall derive the explicit expressions of photoelastic constants for zinc-blende crystals based on the general results obtained in this paper and present numerical examples for GaAs.

#### **II. BASIC EQUATIONS**

We make the following assumptions throughout this paper.

(a) We consider a direct-gap semiconductor with one (s-like) conduction and three (p-like) valence bands, as shown in Fig. 1. In this paper, the effects of excitons are neglected. In the case of some III-V compounds such as GaAs and InP, this assumption is valid under population-inverted condition, since excitons disappear by the plasma-screening effects.<sup>24</sup>

(b) By using the density-matrix method,<sup>25</sup> we derive expressions for the contribution to photoelastic constants from the conduction and valence bands  $(M_0 \text{ critical point})$  taking account of the effect of population inversion. In the derivation, the interband transitions between the conduction and valence bands due to photons, the intraband transitions due to phonons and the interband transitions between the valence bands due to the phonons are considered, while the interband transitions between the conduction and valence bands due to the phonons are neglected.



FIG. 1. Illustration of energy-band diagram of a semiconductor considered in this paper.

(c) The effects of intraband transitions due to photons are considered separately as free-carrier plasma screening and free-carrier absorption in the separate paper.<sup>23</sup> The contributions to the photoelastic constants from higher bands  $(M_1, M_2, \ldots$  critical points) are also considered separately.

(d) We take account of the effects of electronphonon coupling due to deformation potential but neglect piezoelectric coupling. Screening effects for the deformation potential are discussed separately.

(e) The photon wave number  $\beta$  and the phonon wave number K are much smaller than the electron wave number k. The phonon frequency  $\Omega$  is sufficiently low so that the phonons can be dealt with as a classical sound wave.

(f) The photon energy  $\hbar \omega$  is nearly equal to the transition energy  $(\epsilon_C - \epsilon_{V_I})$ .

(g) The bands are isotropic and the energies of the bands are represented by isotropic effective masses.

The equation of motion of the density operator is written

$$\frac{i\hbar\partial\rho}{\partial t} = [H,\rho] - \frac{1}{2}[\Gamma(\rho-\bar{\rho}) + (\rho-\bar{\rho})\Gamma] \quad , \quad (1)$$

where the total Hamiltonian  $H = H_0 + H_e^{(1)} + H_s^{(1)}$  is the sum of the unperturbed Hamiltonian  $H_0$  in the semiconductor, the electron-photon interaction Hamiltonian  $H_e^{(1)}$ , and electron-phonon interaction Hamiltonian  $H_s^{(1)}$ . The three Hamiltonians are given by

$$H_{0} = \vec{\mathbf{P}}^{2}/2m_{0} + V(\vec{\tau}) ,$$

$$H_{e}^{(1)} = -e\vec{\mathbf{A}} \cdot \vec{\mathbf{P}}/m_{0} ,$$

$$H_{s}^{(1)} = e\left[C_{ij}^{(CC)} + \sum_{l} C_{ij}^{(V_{l}V_{l})} + \sum_{\substack{l \ j \ j \ (l \neq l)}} \sum_{(l \neq l)} C_{ij}^{(V_{l}V_{l})}\right]S_{ij} ,$$
(2)

where  $e, m_0$ , and  $\vec{P}$  are the charge of an electron, the free-electron mass, and the momentum operator, respectively. In the expressions for  $H_0$  and  $H_e^{(1)}$ ,  $V(\vec{r})$  is the periodic potential, and  $\vec{A}$  is the vector potential due to the electromagnetic waves (the photons). In the third expression,  $C_{ij}^{(CC)}$ ,  $C_{ij}^{(V_I V_I)}$ , and  $C_{ij}^{(V_I V_J)}$  are the tensor components of the deformation potentials for the conduction band, the *I*th valence band, and the deformation potential which induces the transition between the *I*th and *J*th valence bands, respectively.  $S_{ij}$  is the strain tensor due to the phonons.  $\Gamma$  on the left-hand side of Eq. (1) is an operator representing the effect of intraband relaxation by electron-electron, electron-phonon, or electron-impurity scatterings.  $\bar{\rho}$  is an equilibrium value of  $\rho$ .

We use the Bloch function  $|C, \vec{k}\rangle$  for the conduction band and  $|V_l, \vec{k}\rangle$  for the valence bands. The functions are the eigenfunctions for the unperturbed Hamiltonian  $H_0$ ; i.e.,

$$H_{0}|C,\vec{k}\rangle = \epsilon_{C}(\vec{k})|C,\vec{k}\rangle ,$$

$$H_{0}|V_{I},\vec{k}\rangle = \epsilon_{V_{I}}(\vec{k})|V_{I},\vec{k}\rangle ,$$
(3)

where the energy  $\epsilon_C(\vec{k})$  in the conduction band and  $\epsilon_{V_I}(\vec{k})$  in the *I*th valence band are given as

$$\epsilon_C(\vec{k}) = \hbar^2 k^2 / 2m_C + \epsilon_c ,$$
  

$$\epsilon_{V_I}(\vec{k}) = -\hbar^2 k^2 / 2m_{V_I} + \epsilon_{V_I} .$$
(4)

Here,  $m_C$  and  $m_{V_I}$  are the effective masses of the electrons, and  $\epsilon_C$  and  $\epsilon_{V_I}$  are the band-edge energies, as shown in Fig. 1.

The current density operator  $\vec{j}(\vec{r})$  for the electron is defined as

$$\vec{j}(\vec{r}) = (e/2m_0)[(\vec{P} - e\vec{A})\delta(\vec{r}' - \vec{r}) + \delta(\vec{r}' - \vec{r})(\vec{P} - e\vec{A})] , \quad (5)$$

where the spin current density is ignored and the position operator<sup>25</sup> is defined as  $\delta(\vec{r}' - \vec{r}) = |\vec{r}'\rangle \langle \vec{r}|$ .

As is the usual procedure, we expand the elements of the density matrix:

$$\rho_{ll'} = \rho_{ll'}^{(0)} + \rho_{ll'}^{(1)} + \rho_{ll'}^{(2)} + \cdots , \qquad (6)$$

in successive orders of approximation corresponding to ascending powers of the amplitudes of the vector potential  $\vec{A}$  due to the photons and the strain tensor  $S_{ij}$  due to the phonons. In Eq. (6), the suffixes *l* and *l'* denote the eigenfunctions for the unperturbed Hamiltonian  $H_0$  and the matrix element  $\rho_{ll'}$  is defined as

$$\rho_{\mu'} = \langle l | \rho | l' \rangle$$

The *n*th-order element of the density matrix  $\rho_{ll}^{(n)}$  is given by the iterative equation

$$\frac{i\hbar\partial\rho_{ll}^{(n)}}{\partial t} = \left[H_0, \rho^{(n)}\right]_{ll'} + \left[H_e^{(1)} + H_s^{(1)}, \rho^{(n-1)}\right]_{ll'} - \left(\frac{1}{2}i\hbar\right) \left(\frac{1}{\tau_l} + \frac{1}{\tau_{l'}}\right) \rho_{ll'}^{(n)} , \qquad (7)$$

where  $\tau_{l}$  and  $\tau_{l'}$  are the intraband relaxation times.

The vector potential  $\vec{A}(\vec{r},t)$  and the strain tensor  $S_{ij}(\vec{r},t)$  are written in the plane-wave form as

$$\vec{A}(\vec{r},t) = \vec{A} \exp[i(\vec{\beta} \cdot \vec{r} - \omega t)] ,$$

$$S_{ij}(\vec{r},t) = S_{ij} \exp[i(\vec{K} \cdot \vec{r} - \Omega t)] .$$
(8)

From Eqs. (5) and (8), we obtain the Fourier component of the matrix element of the current density  $\vec{j}_{u'}(\vec{\beta})$ :

$$\vec{j}_{u'}(\vec{\beta}) = (e/2m_0) \langle l | e^{-i(\vec{\beta} \cdot \vec{\tau})} \vec{\mathbf{P}} + \vec{\mathbf{P}} e^{-i(\vec{\beta} \cdot \vec{\tau})} | l' \rangle \quad .(9)$$

The *n*th-order component of the current density is given by  $\int dx dx dx dx$ 

$$\vec{J}^{(n)} = \text{Tr}(\vec{j} \rho^{(n)}) = \sum_{ll'} \vec{j}_{ll'} \rho^{(n)}_{ll'} .$$
(10)

Finally, the zeroth-order components of the density matrix are given as

$$\rho_{C_{\vec{k}}C_{\vec{k}}}^{(0)} = f_{C}(\vec{k}) ,$$

$$\rho_{l_{\vec{k}}}^{(0)} = f_{V_{l}}(\vec{k}) ,$$

$$(11)$$

$$\rho_{l_{l}'}^{(0)} = 0 \text{ for } l \neq l' ,$$

where  $f_C(\vec{k})$  and  $f_{V_I}(\vec{k})$  are the Fermi-Dirac distribution functions for the conduction and valence bands, respectively. The effects of the population inversion can be taken into account by replacing the Fermi levels with the quasi-Fermi levels in the distribution functions; i.e.,  $f_C(\vec{k})$  and  $f_{V_I}(\vec{k})$  are given by

$$f_C(\vec{\mathbf{k}}) = (1 + \exp\{[\epsilon_C(\vec{\mathbf{k}}) - \epsilon_{C_F}]/\kappa T\})^{-1} ,$$
  

$$f_{V_I}(\vec{\mathbf{k}}) = (1 + \exp\{[\epsilon_{V_I}(\vec{\mathbf{k}}) - \epsilon_{V_F}]/\kappa T\})^{-1} ,$$
(11')

where  $\epsilon_{C_F}$  and  $\epsilon_{V_F}$  are the quasi-Fermi levels of the conduction and valence bands, respectively. When

the separation of the quasi-Fermi levels  $(\epsilon_{C_F} - \epsilon_{V_F})$  is larger than the band gap of the semiconductor  $E_g$ , the population of electrons is inverted and the photons with the energies smaller than  $(\epsilon_{C_F} - \epsilon_{V_F})$  can be amplified.

#### **III. FIRST-ORDER TERMS**

Here we derive the expressions for the first-order terms of the density matrix induced by the perturbation energies  $H_e^{(1)}$  and  $H_s^{(1)}$ . Also, the first-order terms of the current density induced by the photons are derived. From the results, we obtain the increment of the dielectric constants due to the transitions between the conduction and valence bands.

Replacing the time derivative with  $-i\omega$  in Eq. (7), we obtain the first-order terms of the density matrix for the perturbation  $H_e^{(1)}$ :

$$\rho_{e}^{(1)}(\omega)_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}},V_{I}\overrightarrow{\mathbf{k}}} = \frac{-(e/m_{0})(M_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}},V_{I}\overrightarrow{\mathbf{k}}})_{i}|\overrightarrow{\mathbf{A}}|[f_{C}(\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}})-f_{V_{I}}(\overrightarrow{\mathbf{k}})]}{\epsilon_{C}(\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}})-\epsilon_{V_{I}}(\overrightarrow{\mathbf{k}})-\hbar\omega-i\hbar(1/\tau_{CV_{I}})},$$

$$\rho_{e}^{(1)}(\omega)_{V_{I}\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}},C_{\overrightarrow{\mathbf{k}}} = \frac{-(e/m_{0})(M_{V_{I}\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}},C_{\overrightarrow{\mathbf{k}}})_{i}|\overrightarrow{\mathbf{A}}|[f_{C}(\overrightarrow{\mathbf{k}})-f_{V_{I}}(\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}})]}{\epsilon_{C}(\overrightarrow{\mathbf{k}})-\epsilon_{V_{I}}(\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}})+\hbar\omega+i\hbar(1/\tau_{CV_{I}})},$$
(12)

where the momentum matrix element is defined as

$$(M_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}},V_{I}\overrightarrow{\mathbf{k}}})_{i} = \langle C, \overrightarrow{\mathbf{k}} + \overrightarrow{\boldsymbol{\beta}} | e^{i\overrightarrow{\boldsymbol{\beta}}\cdot\overrightarrow{\mathbf{r}}} (\overrightarrow{\mathbf{A}}/|\overrightarrow{\mathbf{A}}|) \cdot \overrightarrow{\mathbf{P}} | V_{I}, \overrightarrow{\mathbf{k}} \rangle \quad , \tag{12'}$$

where the suffix *i* denotes the direction of the vector potential  $\vec{A}$ . The relaxation time  $\tau_{CV_I}$  is represented by the intraband relaxation times in the conduction and valence bands

$$1/\tau_{CV_{I}} = \frac{1}{2} (1/\tau_{C} + 1/\tau_{V_{I}}) \quad . \tag{12"}$$

Using a similar procedure, we obtain the first-order terms of the density matrix for the perturbation  $H_s^{(1)}$ :

$$\rho_{s}^{(1)}(\Omega)_{C_{\overrightarrow{k}+\overrightarrow{K}},C_{\overrightarrow{k}}} = \frac{[f_{C}(\overrightarrow{k}+\overrightarrow{k}) - f_{C}(\overrightarrow{k}) - i\hbar\partial f_{C}(\overrightarrow{k})/\partial\epsilon_{C}\tau_{C}]H_{s}^{(1)}C_{\overrightarrow{k}+\overrightarrow{k}},C_{\overrightarrow{k}}}{\epsilon_{C}(\overrightarrow{k}+\overrightarrow{k}) - \epsilon_{C}(\overrightarrow{k}) - \hbar\Omega - i\hbar(1/\tau_{C})}$$

$$\approx \left(\frac{\partial f_{C}(\overrightarrow{k})}{\partial\epsilon_{C}}\right) \frac{e(C_{ij}^{(CC)} - C_{ij}^{(\nu_{1}\nu_{1})})S_{ij}}{2} ,$$

$$\rho_{s}^{(1)}(\Omega)_{\nu_{1\overrightarrow{k}+\overrightarrow{K}},\nu_{1\overrightarrow{k}}} = \frac{[f_{\nu_{1}}(\overrightarrow{k}+\overrightarrow{k}) - f_{\nu_{1}}(\overrightarrow{k}) - i\hbar\partial f_{\nu_{1}}(\overrightarrow{k})/\partial\epsilon_{\nu_{1}}\tau_{\nu_{1}}]H_{s}^{(1)}}{\epsilon_{\nu_{1}}(\overrightarrow{k}+\overrightarrow{k}) - \epsilon_{\nu_{1}}(\overrightarrow{k}) - \hbar\Omega - i\hbar(1/\tau_{\nu_{1}})}$$

$$\approx \left(\frac{\partial f_{\nu_{1}}(\overrightarrow{k})}{\partial\epsilon_{\nu_{1}}}\right) \frac{e(C_{ij}^{(\nu_{1}\nu_{1})} - C_{ij}^{(CC)})S_{ij}}{2} ,$$
(13)

$$\rho_s^{(1)}(\Omega)_{V_I \overrightarrow{k} + \overrightarrow{K}, V_I \overrightarrow{k}} \simeq 0 \quad (\text{for } I = 2, 3)$$

and

$$\rho_{s}^{(1)}(\Omega)_{V_{I\vec{k}}+\vec{K},V_{J\vec{k}}} = \frac{[f_{V_{I}}(\vec{k}+\vec{K}) - f_{V_{J}}(\vec{k})]H_{sV_{I\vec{k}}+\vec{K},V_{J\vec{k}}}^{(1)}}{\epsilon_{V_{I}}(\vec{k}+\vec{K}) - \epsilon_{V_{J}}(\vec{k}) - \hbar\Omega - i\hbar(1/\tau_{V_{I}}V_{J})}$$
(14)

for  $I \neq J$ , where the energy matrix element  $H_{s,u'}^{(1)}$  is given by

$$H_{sll'}^{(1)} = \langle l | e C_{ij} S_{ij} e^{i \overrightarrow{K} \cdot \overrightarrow{\tau}} | l' \rangle \quad .$$

$$(14')$$

In the derivation of Eq. (13), the local equilibrium value of the density matrix  $\tilde{\rho}^{(0)}$  was introduced into the relaxation term of Eq. (7) and the long-wavelength approximation for the phonons was used (see Appendix A).

From Eqs. (9), (10), and (12), we obtain the *j*-directional component of the current density induced by the

photons

$$[\vec{J}^{(1)}(\vec{\omega},\vec{\beta})]_{j} = \sum_{I} \sum_{\vec{k}} \{ [\vec{j}(\vec{\beta})_{V_{I}\vec{k}}, c_{\vec{k}+\vec{\beta}}]_{j} \rho_{e}^{(1)}(\omega)_{C_{\vec{k}+\vec{\beta}}}, V_{I\vec{k}} + [\vec{j}(\vec{\beta})_{C_{\vec{k}}}, V_{I\vec{k}+\vec{\beta}}]_{j} \rho_{e}^{(1)}(\omega)_{V_{I}\vec{k}+\vec{\beta}}, c_{\vec{k}} \}$$

$$\approx -\left(\frac{e}{m_{0}}\right)^{2} |\vec{A}| \sum_{I} [M_{CV_{I}}]_{i} [M_{V_{I}C}]_{j} \sum_{\vec{k}} \frac{f_{C}(\vec{k}) - f_{V_{I}}(\vec{k})}{\epsilon_{C}(\vec{k}) - \epsilon_{V_{I}}(\vec{k}) - \hbar\omega - i\hbar(1/\tau_{CV_{I}})} .$$
(15)

In the derivation of Eq. (15), it was assumed that the photon energy  $\hbar \omega$  is close to the energy gap of the semiconductor and then  $\rho_e^{(1)}(\omega)_{V_{l\vec{k}+\vec{\beta}},C_{\vec{k}}}$  is negligibly small compared with  $\rho_e^{(1)}(\omega)_{C_{\vec{k}+\vec{\beta}},V_{l\vec{k}}}$ . Also, we assumed that  $|\vec{\beta}|$  is much smaller than  $|\vec{k}|$  and that the momentum matrix element is independent of the electron wave number  $\vec{k}$ . By using the macroscopic relations,

$$E_i = -\frac{\partial A_i}{\partial t} = i\,\omega A_i \quad , \quad J_j = \frac{\partial P_j}{\partial t} = -i\,\omega P_j \quad , \quad P_j = \Delta \epsilon_{ij} E_i \quad , \tag{16}$$

(where P is the electric polarization) with the help of Eq. (15), we obtain the increment of the dielectric constants due to the interband transitions between the conduction and valence bands as

$$\Delta \epsilon_{ij}^{(1)}(\omega, \vec{\beta}) = -\left(\frac{e}{m_0}\right)^2 \left(\frac{1}{\pi\omega}\right)^2 \left[\sum_{l} (M_{CV_l})_i (M_{V_lC})_j \int_0^{k_m} \frac{[f_C(\vec{k}) - f_{V_l}(\vec{k})]k^2 dk}{\epsilon_C(\vec{k}) - \epsilon_{V_l}(\vec{k}) - \hbar\omega - i\hbar/\tau_{CV_l}}\right]$$
(17)

where  $k_m$  is the maximum wave number, whose order is the inverse of the atomic distance of the semiconductor. The real part of  $\Delta \epsilon_{ij}^{(1)}(\omega, \vec{\beta})$  represents the enhancement of the dielectric constant by the conduction-to-valence-band transitions. The imaginary part of  $\Delta \epsilon_{ij}^{(1)}(\omega, \vec{\beta})$  is proportional to the absorption (or amplification) coefficient of the photons.

## **IV., SECOND-ORDER TERMS: PHOTOELASTIC CONSTANTS**

We can derive the expressions for the photoelastic constants from the second-order terms of the current density due to the mixing of the fields  $\vec{A}$  and  $S_{ij}$ , because two photons and single phonons are involved in the ordinary Brillouin scatterings.

The iterative relation of Eq. (7) is written for n = 2 as

$$\frac{i\hbar\partial\rho_{ll'}^{(2)}}{\partial t} = \left[H_0, \rho^{(2)}\right]_{ll'} + \left[H_e^{(1)} + H_s^{(1)}, \rho_e^{(1)} + \rho_s^{(1)}\right]_{ll'} - \left(\frac{i\hbar}{\tau_{ll'}}\right)\rho_{ll'}^{(2)} \quad .$$
(17)

From Eq. (7'), one can see that there are two kinds of second-order terms of the density matrix due to the mixing of  $\vec{A}$  and  $S_{ij}$ . One is due to the mixing of  $\rho_s^{(1)}$  and  $H_e^{(1)}$ , and the other is due to the mixing of  $\rho_e^{(1)}$  and  $H_s^{(1)}$ . Let us denote the former as  $\rho_{se}^{(2)}$  and the latter as  $\rho_{es}^{(2)}$ . The terms due to the mixings of  $\rho_e^{(1)}$  and  $H_e^{(1)}$  and of  $\rho_s^{(1)}$  and  $H_s^{(1)}$  represent nonlinear effects by large-signal photon and phonon fields, so that these terms are neglected in this paper.

From Eq. (7') with Eq. (2), we get

$$\rho_{se}^{(2)}(\omega+\Omega)_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}+\overrightarrow{\mathbf{K}}},V_{I\overrightarrow{\mathbf{k}}}} = -\frac{e}{m_{0}} \left| \vec{A} \right| \left[ \left( \rho_{s}^{(1)}(\Omega)_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}+\overrightarrow{\mathbf{K}}},C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}}}(M_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}},V_{I\overrightarrow{\mathbf{k}}}})_{i} - (M_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}+\overrightarrow{\mathbf{K}}},V_{I\overrightarrow{\mathbf{k}}+\overrightarrow{\mathbf{k}}}})_{i}\rho_{s}^{(1)}(\Omega)_{V_{I\overrightarrow{\mathbf{k}}+\overrightarrow{\mathbf{K}}},V_{I\overrightarrow{\mathbf{k}}}} - \sum_{j\neq I} (M_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}+\overrightarrow{\mathbf{K}}},V_{J\overrightarrow{\mathbf{k}}+\overrightarrow{\mathbf{k}}}})_{i}\rho_{s}^{(1)}(\Omega)_{V_{J\overrightarrow{\mathbf{k}}+\overrightarrow{\mathbf{K}}},V_{I\overrightarrow{\mathbf{k}}}} \right) / \left[ \epsilon_{C}(\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}+\overrightarrow{\mathbf{k}}) - \epsilon_{V_{I}}(\overrightarrow{\mathbf{k}}) - \hbar(\omega+\Omega) - i\hbar/\tau_{CV_{I}} \right] \right]$$

$$\rho_{se}^{(2)}(\omega+\Omega)_{V_{I\vec{k}+\vec{\beta}+\vec{K}},C_{\vec{k}}}$$

$$= -\frac{e}{m_{0}} |\vec{A}| \left[ \left[ \left( M_{V_{I\vec{k}+\vec{\beta}+\vec{K}},C_{\vec{k}+\vec{K}}} \right)_{i}\rho_{s}^{(1)}(\Omega)_{C_{\vec{k}+\vec{K}},C_{\vec{k}}} - \rho_{s}^{(1)}(\Omega)_{V_{I\vec{k}+\vec{\beta}+\vec{K}},V_{I\vec{k}+\vec{\beta}}} \left( M_{V_{I\vec{k}+\vec{\beta}},C_{\vec{k}}} \right)_{i} - \sum_{J \neq I} \rho_{s}^{(1)}(\Omega)_{V_{I\vec{k}+\vec{\beta}+\vec{K}},V_{J\vec{k}+\vec{\beta}}} \left( M_{V_{J\vec{k}+\vec{\beta}},C_{\vec{k}}} \right)_{i} \right] / \left[ \epsilon_{C}(\vec{k}) - \epsilon_{V_{I}}(\vec{k}+\vec{\beta}+\vec{k}) + \hbar(\omega+\Omega) + i\hbar/\tau_{CV_{I}} \right] \right]$$

$$(18a)$$

(18a)

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$$\begin{split} \rho_{es}^{(2)}(\omega+\Omega)_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}+\overrightarrow{\mathbf{k}}},V_{I}\overrightarrow{\mathbf{k}}} \\ &= \left[ \left( \rho_{e}^{(1)}(\omega)_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}+\overrightarrow{\mathbf{k}}},V_{I}\overrightarrow{\mathbf{k}}+\overrightarrow{\mathbf{k}}} H_{s}^{(1)}_{V_{I}\overrightarrow{\mathbf{k}}+\overrightarrow{\mathbf{k}}},V_{I}\overrightarrow{\mathbf{k}}} - H_{s}^{(1)}_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}}+\overrightarrow{\mathbf{k}}},C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}} \rho_{e}^{(1)}(\omega)_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}},V_{I}\overrightarrow{\mathbf{k}}} \\ &+ \sum_{j\neq l} \rho_{e}^{(1)}(\omega)_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}+\overrightarrow{\mathbf{k}}},V_{J}\overrightarrow{\mathbf{k}}+\overrightarrow{\mathbf{k}}} H_{s}^{(1)}_{V_{J}\overrightarrow{\mathbf{k}}+\overrightarrow{\mathbf{k}}},V_{I}\overrightarrow{\mathbf{k}}} \right] / \left[ \epsilon_{C}(\overrightarrow{\mathbf{k}}+\overrightarrow{\boldsymbol{\beta}}+\overrightarrow{\mathbf{k}}) - \epsilon_{V_{I}}(\overrightarrow{\mathbf{k}}) - \hbar(\omega+\Omega) - i\hbar/\tau_{CV_{I}} \right] \right] \,, \end{split}$$

$$\rho_{es}^{(2)}(\omega+\Omega)_{V_{l\vec{k}}+\vec{\beta}+\vec{K}},c_{\vec{k}}$$

$$= \left[ \left( H_{s}^{(1)}v_{l\vec{k}+\vec{\beta}+\vec{K}},v_{l\vec{k}+\vec{\beta}}\rho_{e}^{(1)}(\omega)_{V_{l\vec{k}}+\vec{\beta}},c_{\vec{k}} - \rho_{e}^{(1)}(\omega)_{V_{l\vec{k}}+\vec{\beta}+\vec{K}},c_{\vec{k}+\vec{k}}H_{s}^{(1)}c_{\vec{k}+\vec{K}},c_{\vec{k}} + \sum_{j\neq l} H_{s}^{(1)}v_{l\vec{k}+\vec{\beta}+\vec{K}},v_{j\vec{k}+\vec{\beta}}\rho_{e}^{(1)}(\omega)_{V_{j\vec{k}+\vec{\beta}}},c_{\vec{k}} \right) / [\epsilon_{C}(\vec{k}) - \epsilon_{V_{l}}(\vec{k}+\vec{\beta}+\vec{K}) + \hbar(\omega+\Omega) + i\hbar/\tau_{CV_{l}}] \right]$$

From the approximation (f), we obtain the inequalities

$$\begin{aligned} |\rho_{se}^{(2)}(\omega+\Omega)_{C_{\overrightarrow{k}+\overrightarrow{\beta}+\overrightarrow{K}},V_{\overrightarrow{k}}}| >> |\rho_{se}^{(2)}(\omega+\Omega)_{V_{\overrightarrow{k}+\overrightarrow{\beta}+\overrightarrow{K}},C_{\overrightarrow{k}}}| \\ |\rho_{es}^{(2)}(\omega+\Omega)_{C_{\overrightarrow{k}+\overrightarrow{\beta}+\overrightarrow{K}},V_{\overrightarrow{k}}}| >> |\rho_{es}^{(2)}(\omega+\Omega)_{V_{\overrightarrow{k}+\overrightarrow{\beta}+\overrightarrow{K}},C_{\overrightarrow{k}}}| \end{aligned}$$

Making use of Eq. (10) with the above assumption, we obtain the second-order term of the current density as

$$\vec{\mathbf{J}}^{(2)}(\omega+\Omega,\vec{\beta}+\vec{\mathbf{K}}) \simeq \sum_{I} \sum_{\vec{\mathbf{k}}} [\vec{\mathbf{j}} (\vec{\beta}+\vec{\mathbf{K}})_{V_{I\vec{\mathbf{k}}},C_{\vec{\mathbf{k}}+\vec{\beta}+\vec{\mathbf{K}}}} \rho_{se}^{(2)}(\omega+\Omega)_{C_{\vec{\mathbf{k}}+\vec{\beta}+\vec{\mathbf{K}}},V_{I\vec{\mathbf{k}}}} + \vec{\mathbf{j}} (\vec{\beta}+\vec{\mathbf{K}})_{V_{I\vec{\mathbf{k}}},C_{\vec{\mathbf{k}}+\vec{\beta}+\vec{\mathbf{K}}}} \rho_{es}^{(2)}(\omega+\Omega)_{C_{\vec{\mathbf{k}}+\vec{\beta}+\vec{\mathbf{K}}},V_{I\vec{\mathbf{k}}}}] .$$

Substituting Eq. (9) into  $\vec{j} (\vec{\beta} + \vec{K})_{\mu'}$ , Eq. (18a) into  $\rho_{se}^{(2)}(\omega + \Omega)_{\mu'}$ , and Eq. (18b) into  $\rho_{es}^{(2)}(\omega + \Omega)_{\mu'}$ ) in the above equation, the *j* directional component of the current density is given by

$$\begin{bmatrix} \vec{J}^{(2)}(\omega + \Omega, \vec{\beta} + \vec{K}) \end{bmatrix}_{J} = -\frac{e}{m_{0}} \|\vec{A}\| \sum_{I} \left[ (M_{C\nu_{I}})_{i}(M_{\nu_{I}C})_{J} \sum_{\vec{k}} \frac{\rho_{s}^{(1)}c_{\vec{k}}c_{\vec{k}} - \rho_{s}^{(1)}\nu_{I\vec{k}}v_{I\vec{k}}}{\epsilon_{C}(\vec{k}) - \epsilon_{\nu_{I}}(\vec{k}) - \hbar(\omega + \Omega) - i\hbar/\tau_{C\nu_{I}}} - \sum_{J \neq I} (M_{C\nu_{I}})_{i}(M_{\nu_{I}C})_{J} \sum_{\vec{k}} \frac{\rho_{s}^{(1)}c_{\vec{k}}v_{I\vec{k}}}{\epsilon_{C}(\vec{k}) - \epsilon_{\nu_{I}}(\vec{k}) - \hbar(\omega + \Omega) - i\hbar/\tau_{C\nu_{I}}} + (M_{C\nu_{I}})_{i}(M_{\nu_{I}C})_{J} \sum_{\vec{k}} \frac{[f_{C}(\vec{k}) - f_{\nu_{I}}(\vec{k})](H_{s}^{(1)}v_{I},\nu_{I} - H_{s}^{(1)}c_{C})}{[\epsilon_{C}(\vec{k}) - \epsilon_{\nu_{I}}(\vec{k}) - i\hbar\omega - i\hbar/\tau_{C\nu_{I}}]^{2}} + \sum_{J \neq I} (M_{C\nu_{I}})_{i}(M_{\nu_{J}C})_{J} \sum_{\vec{k}} \frac{[f_{C}(\vec{k}) - f_{\nu_{I}}(\vec{k})](H_{s}^{(1)}v_{I},\nu_{I} - H_{s}^{(1)}c_{C})}{[\epsilon_{C}(\vec{k}) - \epsilon_{\nu_{I}}(\vec{k}) - i\hbar\omega - i\hbar/\tau_{C\nu_{I}}]^{2}} \right] .$$
(19)

In the derivation of Eq. (19), we neglected  $\vec{\beta}$  and  $\vec{K}$  compared with  $\vec{k}$ , and neglected  $\Omega$  compared with  $\omega$ , from the approximation (e).

From the definition, the photoelastic constant  $p_{ijkl}$  is given by

$$p_{ijkl} = -\Delta \epsilon_{ij}^{(2)} / \epsilon_{ii} \epsilon_{jj} \epsilon_0 S_{kl} \quad , \tag{20}$$

where  $\Delta \epsilon_{ij}^{(2)}$  is the increment of the tensor component of the dielectric constant induced by the strain  $S_{ij}$ .  $\epsilon_{ii}$ ,  $\epsilon_{jj}$ , and  $\epsilon_0$  are the diagonal tensor components of the specific dielectric constants and the free-space dielectric con-

(18b)

stant, respectively. From Eqs. (13), (14), (16), and (19), we obtain the expressions for  $\Delta \epsilon_{ij}^{(2)}$ . Substituting the results into  $\Delta \epsilon_{ij}^{(2)}$  in Eq. (20), we get the expressions for the increments of the photoelastic constants due to the conduction-to-valence-band transitions

$$\begin{split} \Delta p_{ijkl} &= \frac{(e/m_0)^2}{2\pi^2 \omega^2 \epsilon_0 \epsilon_{ii} \epsilon_{jj}} \left[ \sum_{I} (M_{CV_I})_i (M_{V_IC})_j \int_0^\infty \frac{[\partial f_C(k)/\partial \epsilon_C + \delta_{1/} \partial f_{V_1}(k)/\partial \epsilon_{V_1}] [e(C_{kl}^{(CC)} - C_{kl}^{(V_1V_1)})/2] k^2 dk}{\epsilon_C(k) - \epsilon_{V_I}(k) - \hbar \omega - i\hbar/\tau_{CV_I}} \right] \\ &- \sum_{I} \sum_{I=J} (M_{CV_I})_i (M_{V_JC})_j \int_0^\infty \frac{[f_C(k) - \epsilon_{V_I}(k) - \epsilon_{V_I}(k)] eC_{kl}^{(V_IV_J)} k^2 dk}{[\epsilon_C(k) - \epsilon_{V_I}(k) - \hbar \omega - i\hbar/\tau_{CV_I}] [\epsilon_{V_I}(k) - \epsilon_{V_J}(k) - \hbar \Omega - i\hbar/\tau_{V_IV_J}]} \\ &- \sum_{I} (M_{CV_I})_i (M_{V_IC})_j \int_0^\infty \frac{[f_C(k) - f_{V_I}(k)] e(C_{kl}^{(CC)} - C_{kl}^{(V_IV_J)}) k^2 dk}{[\epsilon_C(k) - \epsilon_{V_I}(k) - \hbar \omega - i\hbar/\tau_{CV_I}]^2} \\ &+ \sum_{I=J} \sum_{I=J} (M_{CV_I})_i (M_{V_IC})_j \int_0^\infty \frac{[f_C(k) - f_{V_I}(k)] e(C_{kl}^{(CC)} - C_{kl}^{(V_IV_J)}) k^2 dk}{[\epsilon_C(k) - \epsilon_{V_I}(k) - \hbar \omega - i\hbar/\tau_{CV_I}]^2} \end{split}$$

where

$$\delta_{1/} = \begin{cases} 1 & \text{for } I = 1 \\ 0 & \text{for } I = 2 \text{ or } 3 \end{cases}$$

Strictly speaking, we must consider the screening effects for the deformation potentials by the free carriers in the conduction and valence bands. But, as discussed in Appendix B, under the population-inversion condition, the deformation potentials are not screened because the semiconductor at high excitation levels is intrinsic and the local charges induced by the low-frequency phonons are perfectly neutralized. Therefore, we can use the bare values for the deformation potentials in Eq. (21). Rather, the deformation potentials may be enhanced by the manybody effects due to high-density electrons under the population-inverted semiconductors. The enhancement of the deformation potentials will be discussed in a separate paper.<sup>23</sup>

We consider the physical meanings of each term in Eq. (21).

(i) The first and second terms in Eq. (21) are due to  $\rho_{sc}^{(2)}$ , i.e., the mixing of  $H_{e}^{(1)}$  and  $\rho_{s}^{(1)}(\Omega)$ .  $\rho_{s}^{(1)}(\Omega)$  is the first-order fluctuation of the electron density due to the phonons. Therefore, these terms represent the modulations of the refractive index and the absorption (or amplification) coefficient for the photons directly induced by the phonons. In the case of unexcitation where  $f_C \approx 0$  and  $f_{V_I} \approx 1$ , these terms are negligibly small, while in the populationinverted semiconductors, these terms make some contributions to the photoelastic constants since  $\partial f_C/\partial \epsilon_C$  and  $\partial f_{V_I}/\partial \epsilon_{V_I}$  have finite values and  $f_{V_I}(k) \neq f_{V_I}(k)$ . These terms have not been considered in the previous papers.<sup>1,14,15</sup>

(ii) The third and fourth terms are due to  $\rho_{es}^{(2)}$ , i.e., the mixing of  $H_s^{(1)}$  and  $\rho_e^{(1)}$ , which correspond to straightforward extensions of the resonant terms in Loudon's theory. The terms involve the effects of the population changes and the intraband relaxations. If we put  $f_C = 0$ ,  $f_V = 1$ , and  $\tau_{H'} = \infty$ , each of the terms reduces to the resonant term in Loudon's theory.

(iii) The first and third terms are due to the intraband transitions of the electrons by the phonons. The second and fourth terms are due to the interband transitions between the valence bands.

(iv) The third and fourth terms include the factor  $f_C(k) - f_{V_I}(k)$ . In low excitation, the factor has a negative value, while in population inversion, the factor has a positive value for  $\epsilon_C(k) - \epsilon_{V_I}(k)$  smaller than  $\epsilon_{C_F} - \epsilon_{V_F}$ . Therefore, the changes of sign of the photoelastic constants at high excitation levels may occur. Some numerical examples of such a phenomenon for GaAs will be described in a separate paper.<sup>23</sup>

(v) As described in the above paragraph, Eq. (21) includes the relaxation terms. As a result, Eq. (21) does not diverge even for the photon energy which is equal to or greater than the band-gap energy of the semiconductor. Also,  $\Delta p_{ijkl}$  in Eq. (21) is a complex quantity. The imaginary part of the photoelastic constant represents the modulation of absorption (or amplification) for the photons. When the photon energy becomes very close to the band-gap energy, the imaginary part of the photoelastic constant may be larger than the real part. The importance of the imaginary part will be explicitly shown by the numerical examples in a separate paper.<sup>23</sup>

(21)

## V. DISCUSSION AND CONCLUSION

We obtained the general expression for the contribution to photoelastic constants from conduction and valence bands ( $M_0$  critical point) taking account of the effect of population inversion. Assumptions (a)-(g) made in Sec. II are sufficiently allowable but we must check the validity of the assumption that we can use the Bloch functions for the electrons as base functions. The use of Bloch functions means that the k selection rule at electronic transitions holds good. On the other hand, it is said that the k selection rule may be broken by carrier-carrier interactions at high excitation levels even in a pure semiconductor.<sup>24</sup> However, from the recent works on semiconductor lasers,  $^{26,27}$  we feel that the k selection rule holds good to some extent at high-temperature region (77 to 300 K). Therefore, Eq. (21) can be used to calculate the photoelastic constants of direct-gap III-V compounds such as GaAs, InP, and their mixed crystals which is highly excited at the high-temperature region. In a separate paper,<sup>23</sup> the explicit expressions for the photoelastic constants of zinc-blende crystals will be derived from Eq. (21) and the numerical examples will be given for the photoelastic constants of GaAs under population-inverted condition together with the discussion on the enhancement of deformation potentials.

Finally, we point out that in the cases of highly doped semiconductors and II-VI compounds, the present theory should be modified. In the former case, we must include the wave functions of impurity levels (or impurity band) in the base functions for density-matrix calculations. In the latter case, the effects of excitons must be taken into account. Also, at intermediate excitation levels where no population inversion is attained, we must take account of the screening of the deformation potentials by free carriers.

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## APPENDIX A: LOCAL EQUILIBRIUM VALUE OF DENSITY MATRIX

In calculating the first-order terms of density matrix due to phonons  $\rho_{s\,l'}^{(1)}$ , we obtain incorrect results if we use the zeroth-order component of density matrix  $\rho_{ll'}^{(0)}$  for the equilibrium value  $\bar{\rho}$ . In such a case, we must introduce the concept of local equilibrium value  $\tilde{\rho}$  since electron densities are spatially modulated by low-frequency phonons. On the analysis of interaction between electrons and phonons in semimetals and semiconductors, Mikoshiba<sup>28</sup> and Spector<sup>29</sup> have introduced the local equilibrium value of electron distribution into a classical Boltzmann equation.

According to their procedures, we replace  $\overline{\rho}$  in the relaxation term of Eq. (1) by  $\tilde{\rho}$ . Therefore, the first-order term  $\rho_{s\,l'}^{(1)}$  should be calculated by

$$i\hbar \left(\frac{\partial \rho_{s}^{(1)}}{\partial t}\right)_{ll'} = \left[H_{0}, \rho_{s}^{(1)}\right]_{ll'} + \left[H_{s}^{(1)}, \rho^{(0)}\right]_{ll'} - \frac{i\hbar}{\tau_{ll'}} \left(\rho_{s}^{(1)}_{ll'} + \rho_{ll'}^{(0)} - \tilde{\rho}_{ll'}\right) \quad . \quad (A1)$$

We determine the local equilibrium value  $\tilde{\rho}_{u'}$  from physical consideration. As shown in Appendix B, the potential fluctuation, due to phonons, for the electrons in the conduction band of a population-inverted semiconductor is given by

$$H_{s}^{(1)}{}_{C_{\vec{k}}+\vec{K}}, C_{\vec{k}} = \frac{1}{2}e(C_{ij}^{(CC)} - C_{ij}^{(V_{1}V_{1})})S_{ij}$$

We can assume that the quasi-Fermi level in the conduction band is spatially constant, since the dielectric relaxation frequency of the electrons is much higher than the frequency of the phonons which can be dealt with as a classical sound wave. Therefore, taking account of only the effect due to the potential modulation, the local equilibrium value of density matrix  $\tilde{\rho}_{C \frac{1}{K}C \frac{1}{K}}$  is written

$$\tilde{\rho}_{C_{\overrightarrow{k}}C_{\overrightarrow{k}}} = \rho_{C_{\overrightarrow{k}}C_{\overrightarrow{k}}}^{(0)} + \frac{\partial \rho_{C_{\overrightarrow{k}}C_{\overrightarrow{k}}}^{(0)}}{\partial \epsilon_{C}} \frac{e(C_{ij}^{(CC)} - C_{ij}^{(\nu_{1}\nu_{1})})S_{ij}}{2}$$
$$= f_{C}(\overrightarrow{k}) + \frac{\partial f_{C}(\overrightarrow{k})}{\partial \epsilon_{C}} \frac{e(C_{ij}^{(CC)} - C_{ij}^{(\nu_{1}\nu_{1})})S_{ij}}{2} \quad .$$
(A2)

Substituting Eq. (A2) into Eq. (A1), we obtain the first-order fluctuation of density matrix

$$\rho_{s}^{(1)}(\Omega)_{C_{\overrightarrow{\mathbf{k}}+\overrightarrow{\mathbf{K}}},C_{\overrightarrow{\mathbf{k}}}} = \frac{\{f_{C}(\overrightarrow{\mathbf{k}}+\overrightarrow{\mathbf{K}}) - f_{C}(\overrightarrow{\mathbf{k}}) - i\hbar[\partial f_{C}(k)/\partial\epsilon_{C}]/\tau_{C}\}\frac{1}{2}e(C_{ij}^{(CC)} - C_{ij}^{(V_{1}V_{1})})S_{ij}}{\epsilon_{C}(\overrightarrow{\mathbf{k}}+\overrightarrow{\mathbf{K}}) - \epsilon_{C}(\overrightarrow{\mathbf{k}}) - \hbar\Omega - i\hbar/\tau_{C}}$$
(A3)

In the long-wavelength limit  $(K \rightarrow 0 \text{ and } \Omega \rightarrow 0)$ , Eq. (A3) reduces to

$$\rho_{s}^{(1)}(\Omega)_{C_{\vec{k}+\vec{k}},C_{\vec{k}}} = \frac{\partial f_{C}(k)}{\partial \epsilon_{C}} \frac{e(C_{ij}^{(CC)} - C_{ij}^{(V_{1}V_{1})})S_{ij}}{2} .$$
(A3')

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Using a similar procedure, we can obtain the first-order fluctuation of density matrix for the electrons in the valence band  $V_1$ :

$$\rho_{s}^{(1)}(\Omega)_{\nu_{1}\vec{k}+\vec{K}}\nu_{1}\vec{k}} = \frac{\{f_{\nu_{1}}(\vec{k}+\vec{K}) - f_{\nu_{1}}(\vec{k}) - i\hbar[\partial f_{\nu_{1}}(\vec{k})/\partial \epsilon_{\nu_{1}}]/\tau_{\nu_{1}}\}\frac{1}{2}e(C_{ij}^{(\nu_{1}\nu_{1})} - C_{ij}^{(CC)})S_{ij}}{\epsilon_{\nu_{1}}(\vec{k}+\vec{K}) - \epsilon_{\nu_{1}}(\vec{k}) - \hbar\Omega - i\hbar/\tau_{\nu_{1}}}$$

$$\approx \frac{\partial f_{\nu_{1}}(\vec{k})}{\partial \epsilon_{\nu_{1}}}\frac{e(C_{ij}^{(\nu_{1}\nu_{1})} - C_{ij}^{(CC)})S_{ij}}{2} \quad .$$
(A4)

For the valence bands  $V_2$  and  $V_3$ , we can put

$$\rho_s^{(1)}(\Omega)_{V_l \vec{k} + \vec{K}, V_l \vec{k}} \simeq 0 \quad (\text{for } l = 2, 3) \quad , \qquad (A5)$$

since there are very few holes in the bands.

In the cases of interband transitions between valence bands, electron densities are not modulated spatially. Therefore, the concept of the local equilibrium value of density matrix is not required in the derivation of Eq. (14) of the text.

# APPENDIX B: SCREENING OF DEFORMATION POTENTIALS IN POPULATION-INVERTED SEMICONDUCTOR

We must consider the screening effects of deformation potentials by free carriers in a populationinverted semiconductor. In principle, we can obtain an exact solution for the problem by solving the simultaneous equations which consist of the equation of motion of density matrix and Maxwell's equations. However, it is very difficult to solve exactly the simultaneous equations. Fortunately, in the population-inverted semiconductor, the deformation potentials are not screened for the following physical reason. When a semiconductor is so highly excited as to be population inverted, the density of the excited carriers is much higher than carrier density at thermal equilibrium. In such a case, the excited semiconductor can be regarded as intrinsic unless the semiconductor is highly doped. Therefore, any local electron charge induced by low-frequency phonons is neutralized perfectly by holes and vice versa. The deformation potentials in the highly excited semiconductor are not screened substantially, since no net charge is induced by the phonons. Mikoshiba<sup>30</sup> analyzed theoretically the interaction of electrons and holes with acoustic waves in intrinsic semiconductors. Following his analysis, we can confirm quantitatively the above speculation on the screening of deformation potentials and examine the effective potentials which act on the electrons and holes. The phonons considered in this paper can be regarded as a lowfrequency acoustic wave; i.e., the phonon frequency ≤10 GHz and

$$Kl_C << 1$$
,  $Kl_V << 1$ ,

where  $l_C$  and  $l_V$  are the mean-free paths of the electrons in the conduction band and of the holes in the valence band, respectively. The densities of the excited electrons and holes are sufficiently high and are nearly equal to each other. For example, in the case of GaAs, the excited carrier density, required to realize population inversion at room temperature, is of the order of  $10^{18}$  cm<sup>-3</sup>. Therefore, the present situation belongs to category (C) in Ref. 30:

$$\frac{3\tau_C}{\tau} << (Kl_C)^2 << 1, \quad \frac{6\omega_C^2}{K^2} >> v_{\text{th},C}^2, \quad \Omega \tau_C << 1 ,$$

where  $\tau$ ,  $\omega_C$ , and  $v_{\text{th},C}$  are the recombination lifetime, the electron-plasma frequency, and the thermal velocity of the electrons, respectively. Similar relations are valid for the holes.

In this case, the ac components of electron-current density  $J_C$  and hole-current density  $J_V$  are given by Eqs. (3.8) and (4.6) in Ref. 30:

$$J_{C} - J_{V} = \frac{-Ne^{2}(\Omega/K)(C_{ij}^{(CC)} - C_{ij}^{(VV)})S_{ij}}{\kappa T} \\ \times \left[1 - \frac{1}{2} \left(\frac{3\tau_{C}/\tau}{K^{2}l_{c}^{2}} + \frac{3\tau_{V}/\tau}{K^{2}l_{V}^{2}}\right)(1 - i\,\Omega\,\tau) + \frac{1}{4} \left(\frac{3\tau_{C}/\tau}{K^{2}l_{c}^{2}} + \frac{3\tau_{V}/\tau}{K^{2}l_{V}^{2}}\right)(1 - i\,\Omega\,\tau)^{2}\right],$$
(B1)

$$J_C + J_V = -\frac{\epsilon K \Omega}{8\pi} (C_{ij}^{(CC)} + C_{ij}^{(VV)}) S_{ij} \\ \times \left[ 1 - \frac{(\Delta + \delta/2) m_C (1 - i \Omega \tau)}{(1 + \delta) \tau_C \tau K^2 \kappa T} \right]$$

where

$$\delta = -\frac{m_C \tau_V}{m_V \tau_C} - 1 \quad ,$$
  
$$\Delta = -\delta \frac{C_{ij}^{(VV)} + (C_{ij}^{(CC)} - C_{ij}^{(VV)})/(2 - 2i\,\Omega\,\tau)}{C_{ij}^{(VV)} + C_{ij}^{(CC)}} \quad ,$$

and the equations are rewritten with the notation used in this paper. Substituting Eq. (B1) into Eq. (2.2) of Ref. 30, we obtain an ac component of electric field as

$$E = \left(\frac{1}{2}iK\right) \left(C_{ij}^{(CC)} + C_{ij}^{(VV)}\right) S_{ij}$$

$$\times \left(1 - \frac{m_C(\Delta + \frac{1}{2}\delta)(1 - i\Omega\tau)}{(1 + \delta)\tau_C K^2 \kappa T\tau}\right) .$$
(B2)

The local field, which acts on the electron, is given by Eq. (2.10) of Ref. 30 as

$$E_{C}' = E - iKC_{ij}^{(CC)}S_{ij} \quad . \tag{B3}$$

By using Eqs. (B2) and (B3) with the relation grad  $\phi = -E$ , i.e.,  $-iK\phi = E$ , we obtain the local potential for the electrons as

$$e \phi_{C}' = \frac{1}{2} e \left[ (C_{ij}^{(CC)} - C_{ij}^{(VV)}) + (C_{ij}^{(CC)} + C_{ij}^{(VV)}) \times \frac{(\Delta + \frac{1}{2}\delta)(1 - i\,\Omega\,\tau)\,m_{C}}{(1 + \delta)\,\tau_{C}K^{2}\kappa T\,\tau} \right] S_{ij} \quad (B4)$$

By the similar procedure, we obtain the local potential for the holes as

$$(-e)\phi_{\nu}' = \frac{1}{2}e\left[ (C_{ij}^{(CC)} - C_{ij}^{(\nu\nu)}) - (C_{ij}^{(CC)} + C_{ij}^{(\nu\nu)}) \right] \times \frac{(\Delta + \frac{1}{2}\delta)(1 - i\,\Omega\,\tau)m_{C}}{(1 + \delta)\tau_{C}K^{2}\kappa T\tau} S_{ij} .$$
(B5)

In usual cases, i.e.,  $\tau_C \sim \tau_V \sim 10^{-13}$  sec,  $\tau \sim 10^{-9}$  sec, and  $T = 77 \sim 300$  K, the second term in the large parentheses of the right-hand sides of Eqs. (B4) and (B5) are much smaller than the first terms. Therefore, the potentials for electrons and holes are approximately given by

$$e \phi'_{C} = \frac{1}{2} e \left( C_{ij}^{(CC)} - C_{ij}^{(VV)} \right) S_{ij} ,$$
  
(-e)  $\phi'_{V} = \frac{1}{2} e \left( C_{ij}^{(CC)} - C_{ij}^{(VV)} \right) S_{ij} .$  (B6)

Thus, the effective deformation potentials for the

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electrons in the conduction and valence bands are given by  $\frac{1}{2}(C_{ij}^{(CC)} - C_{ij}^{(VV)})$  and  $\frac{1}{2}(C_{ij}^{(VV)} - C_{ij}^{(CC)})$ , respectively.

In Ref. 30, it was assumed that the semiconductor has one conduction band and one valence band. Using the above discussion, we can examine the effective deformation potentials in our case where the semiconductor has one conduction band and three valence bands. Almost all holes occupy the highest valence band, i.e., the valence band  $V_1$  in Fig. 1. Therefore, the suffix V in Eq. (B6) should be considered as  $V_1$ . For the valence bands  $V_2$  and  $V_3$ , we can consider that the relative relation between the valence bands  $V_1$  and  $V_2$  (or  $V_3$ ) in energy scale is unchanged compared with that at unexcitation.

Summarizing the above discussions, the effective deformation potentials in population-inverted semiconductor are given by, for the conduction band, valence band  $V_1$ , and valence band  $V_2$  or  $V_3$ , respectively,

$$\frac{1}{2} (C_{ij}^{(CC)} - C_{ij}^{(\nu_1 \nu_1)}) ,$$

$$\frac{1}{2} (C_{ij}^{(\nu_1 \nu_1)} - C_{ij}^{(CC)}) ,$$

$$(C_{ij}^{(\nu_1 \nu_1)} - C_{ij}^{(\nu_1 \nu_1)}) + \frac{1}{2} (C_{ij}^{(\nu_1 \nu_1)} - C_{ij}^{(CC)})$$

$$= C_{ij}^{(\nu_1 \nu_i)} - \frac{1}{2} C_{ij}^{(\nu_1 \nu_1)} - \frac{1}{2} C_{ij}^{(CC)}$$

The intraband deformation potentials are involved in the form of the difference of the deformation potentials for the conduction and valence bands in the third term of Eq. (21). Therefore, even if we replace the deformation potentials in the third term of Eq. (21) with the above effective values, the factor  $(C_{ij}^{(CC)} - C_{ij}^{(V_I V_I)})$  is unchanged. Apparently, the deformation potentials in population-inverted semiconductors can be regarded as being unscreened by free carriers. Also, in the cases of interband transitions, the deformation potentials are not screened, since no net charge is induced.

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