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# Free-carrier absorption in n-type piezoelectric semiconductor films

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Abstract. Free-carrier absorption in n-type GaAs films has been investigated for the case where the free carriers are confined in a quasi-two-dimensional semiconducting structure with a non-parabolic energy band of electrons. It is assumed that the carriers in semiconductors are scattered by acoustic phonons via firstly the deformation-potential coupling and secondly the piezoelectric coupling. Results show that the free-carrier absorption coefficient depends upon the polarization of the electromagnetic radiation relative to the direction normal to the quasi-two-dimensional structure, the film thickness, the photon frequency and the temperature of the semiconductors. The free-carrier absorption coefficient could be complex owing to the interaction of photons, phonons and conduction electrons in piezoelectric semiconductors. Firstly, when the deformation-potential coupling is dominant, the absorption coefficient increases with decreasing photon frequency and increasing temperature for the radiation field polarized parallel and perpendicular to the layer plane. It is also shown that the absorption coefficient increases with decreasing film thickness. Secondly, when the piezoelectric scattering is dominant, the absorption coefficient increases with decreasing photon frequency and decreasing film thickness for the radiation field polarized parallel and perpendicular to the layer plane. However, the absorption coefficient increases with increasing temperature for the radiation field polarized parallel to the layer plane while, for the radiation field polarized perpendicular to the layer plane, the absorption coefficient increases with decreasing temperature,

# 1. Introduction

There has been a growing interest in the electronic and optical properties of semiconducting layered heterojunctions (Chang et al 1974, Dingle et al 1974, 1975, Esaki and Chang 1974, Seabaugh et al 1989, Chudinov et al 1990), thin films (Mattis and Beni 1978, Wu and Tsai 1983), and inversion layers (Mahan and Hopfield 1965, Ezawa 1971, Padmanabhan and Rothwarf 1989). Because of the confinement of carriers in these quasi-two-dimensional structures, size quantization begins to play an important role in determining the electronic and optical properties of solids. Holonyak et al (1980b) and Vojak et al (1981a, b) have studied both theoretically and experimentally the optical emission from quantum wells because such quantum well devices seem to hold great potential for use in lasers (Holonyak et al 1980a). Tournié et al (1993) have investigated experimentally the optical properties of InAs quantum wells emitting between 0.9 and 2.5  $\mu$ m. The optical absorption can take place via the direct interband transition, the intersubband optical transition, and indirect intraband optical transitions in which carriers absorb or emit a photon while simultaneously scattering off phonons or other imperfections in solids. Such free-carrier absorption accounts for the absorption of the electromagnetic radiation with frequency  $\Omega$  lower than those which give rise to interband transitions in semiconductors, i.e.  $\hbar\Omega < E_g$ , where  $E_g$  is the energy

gap between the conduction and valence bands. In quantizing magnetic fields, the motion of carriers is confined in a plane perpendicular to a DC magnetic field and it was found that the free-carrier absorption coefficient depends upon the polarization of the radiation field relative to the direction of the magnetic field (Rynne and Spector 1981). For III–V compound semiconductors such as GaAs, the interaction of acoustic phonons with carriers is dominated by the deformation-potential coupling and the piezoelectric coupling (Wu 1983). However, the electron-electron interaction is neglected here in the free-carrier absorption phenomenon in III–V semiconductors without considering the hot-electron effect.

In this paper, we investigate the quantum theory of the free-carrier absorption in III– V semiconductors with a non-parabolic energy band of electrons to take account of the quantization of the energy levels of carriers which are confined by their motion in a quasitwo-dimensional structure. In the calculation of the free-carrier absorption coefficient, we make the following assumptions.

(1) For a quasi-two-dimensional non-degenerate electron gas, the distribution function can be represented by the Maxwell-Boltzmann distribution.

(2) The interaction between acoustic phonons and conduction electrons is via the deformation-potential coupling and the piezoelectric coupling in piezoelectric semiconductors.

(3) The energy band of carriers in semiconductors is non-parabolic.

In section 2, we describe electronic states in quasi-two-dimensional semiconducting structures for the non-parabolic band structure. In section 3, the quantum theory of the free-carrier absorption with confined carriers in non-degenerate semiconductors for the deformation-potential coupling is presented. Since we consider the effect of acoustic phonon scattering with conduction electrons and photons, thus the film thickness considered here will be several times those of the usual quantum wells. We discuss two special cases: the radiation field polarized parallel to the layer plane, and the radiation field polarized perpendicular to the layer plane. In section 4, some numerical results of the free-carrier absorption coefficient are presented for n-type GaAs. Finally, a brief discussion is given.

#### 2. Electronic states in a thin layer of semiconductors for non-parabolic bands

The motion of conduction electrons parallel to thin films may be described by plane waves, and those perpendicular to the surface will be described by types of standing wave depending on the structure of the potential. For a square well potential along the z axis with infinitely high barriers at z = 0 and z = d the electron field operator  $\Psi(r)$  is given by (Tamura and Sakuma 1977)

$$\Psi(\mathbf{r}) = \left(\frac{2}{V}\right)^{1/2} \sum_{n=1}^{\infty} \sum_{k} b_{kn} \exp(i\mathbf{k} \cdot \mathbf{x}) \sin\left(\frac{n\pi z}{d}\right)$$
(1)

where the position vector r is given by r = (x, z) = (x, y, z), V = dS is the film volume with a surface area S,  $k = (k_x, k_y)$  is the electron wavevector in the x-y plane, and  $b_{kn}$ and its Hermitian conjugate  $b_{kn}^{\dagger}$  are annihilation and creation operators, respectively, of conduction electrons, satisfying commutative relations of the Fermi type. The  $E_{kn}$  of the conduction electrons for the non-parabolic band structure is given by the relation (Nag 1972, Wu 1983)

$$E_{kn}\left(1+\frac{E_{kn}}{E_g}\right) = \frac{\hbar^2 |k|^2}{2m^*} + \frac{\pi^2 \hbar^2 n^2}{2m^* d^2} \qquad n = 1, 2, 3, \dots$$
(2)

where  $m^*$  is the effective mass of the electron. The energies  $E_{kn}$  of electrons for the non-parabolic band in equation (2) can be written as

$$E_{kn} = -\frac{1}{2}E_{g}\left\{1 - \left[1 + \frac{4}{E_{g}}\left(\frac{\hbar^{2}|k|^{2}}{2m^{*}} + \frac{\pi^{2}\hbar^{2}n^{2}}{2m^{*}d^{2}}\right)\right]^{1/2}\right\} \qquad n = 1, 2, 3, \dots$$
(3)

Since  $(\hbar^2 |k_{\max}|)^2 / 2m^* + (\pi \hbar n)^2 / 2m^* d^2 \simeq k_B T < E_g$  for semiconductors with  $T \leq 300$  K, equation (3) can be expanded as

$$E_{kn} \simeq -\frac{1}{2}E_g + \frac{1}{2}E_g a_n + \hbar |k|^2 / 2m^* a_n \tag{4}$$

with

$$a_n = (1 + 2\pi^2 \hbar^2 n^2 / m^* d^2 E_g)^{1/2}.$$
(5)

The expression for  $E_{kn}$  in equation (5) can be transformed according to Bastard *et al* (1991).

# 3. Quantum theory of free-carrier absorption with confined carriers in non-degenerate semiconductors

The absorption coefficient for the free-carrier absorption can be related to the quantummechanical transition probability for the absorption of photons (Spector 1983):

$$\alpha = \frac{\varepsilon^{1/2}}{n_0 c} \sum_i W_i f_i \tag{6}$$

where  $\varepsilon$  is the dielectric constant of the material,  $n_0$  is the number of photons in the radiation field and  $f_i$  is the free-carrier distribution function. The transition probability can be calculated using the Born second-order golden rule approximation

$$W_{i} = \frac{2\pi}{\hbar} \sum_{f} \left[ |\langle f|M_{+}|i\rangle|^{2} \delta(E_{f} - E_{i} - \hbar\Omega - \hbar\omega_{q}) + |\langle f|M_{-}|i\rangle|^{2} \delta(E_{f} - E_{i} - \hbar\Omega + \hbar\omega_{q}) \right]$$
(7)

where  $E_i$  and  $E_f$  are the initial and final energies, respectively, of electrons,  $\hbar\Omega$  is the photon energy,  $\hbar\omega_q$  is the phonon energy, and  $\langle f|M_{\pm}|i\rangle$  are the transition matrix elements from the initial state to the final state for the interaction between electrons, photons and phonons. These transition matrix elements can be represented by

$$\langle f|M_{\pm}|i\rangle = \sum_{j} \left( \frac{\langle f|H_{\rm rad}|j\rangle\langle j|V_{\rm s}|i\rangle}{E_{j} - E_{i} \mp \hbar\omega_{q}} + \frac{\langle f|V_{\rm s}|j\rangle\langle j|H_{\rm rad}|i\rangle}{E_{j} - E_{i} - \hbar\Omega} \right)$$
(8)

where  $H_{rad}$  is the electron-photon interaction Hamiltonian between conduction electrons and the radiation field, and  $V_s$  is the scattering potential due to the electron-phonon interaction. The sum is over all the intermediate states j of the system.

The electron-photon interaction Hamiltonian is given by

$$H_{\rm rad} = -\frac{e}{m^*} \left(\frac{2\pi\hbar n_0}{\varepsilon\Omega V}\right)^{1/2} \hat{\varepsilon} \cdot p \tag{9}$$

where  $\hat{\varepsilon}$  is the polarization vector of the radiation field and p is the electron momentum in semiconducting films. Its matrix elements for electrons in the same band are given as follows.

(i) When the radiation field is polarized parallel to the layer plane,

$$\langle \mathbf{k}'n'|H_{\rm rad}|\mathbf{k}n\rangle = -\frac{e\hbar}{m^*} \left(\frac{2\pi\hbar n_0}{\varepsilon\Omega V}\right)^{1/2} \hat{\varepsilon} \cdot \mathbf{k}\delta_{n',n}\delta_{k'_x,k_x}\delta_{k'_y,k_y}.$$
 (10)

(ii) When the radiation field is polarized perpendicular to the layer plane,

$$\langle k'n'|H_{\rm rad}|kn\rangle = -\frac{\mathrm{i}e\hbar}{m^*} \left(\frac{2\pi\hbar n_0}{\varepsilon\Omega V}\right)^{1/2} \times \frac{n}{d} \left(\frac{1-\cos[\pi(n'+n)]}{n'+n} + \frac{1-\cos[\pi(n'-n)]}{n'-n}\right) \delta_{k'_x,k_x} \delta_{k'_y,k_y}$$
(11)

with  $n' \neq n$ . The limit of the second term in equation (11) with  $n' \rightarrow n$  will vanish. Thus, when the radiation field is polarized perpendicular to the layer plane, the breakdown of the selection rule occurs at n' = n. This means that one has to sum over all intermediate states when one substitutes equation (11) into equation (8).

The distribution function for a quasi-two-dimensional non-degenerate electron gas can be expressed as

$$f_{kn} = \left(\frac{n_{\rm e}d\hbar^2}{2\pi^2 m^* k_{\rm B}T}\right)^{1/2} \left[\sum_{l=1}^{\infty} a_l \exp\left(-\frac{E_{\rm g}a_l}{2k_{\rm B}T}\right)\right]^{-1} \exp\left(-\frac{E_{\rm g}a_n}{2k_{\rm B}T} - \frac{\hbar^2(k_x^2 + k_y^2)}{2m^* k_{\rm B}T a_n}\right)$$
(12)

where  $n_e$  is the concentration of electrons in the semiconductor.

For III-V semiconductors, there are two dominant electron-phonon scattering mechanisms.

(A) When the deformation-potential coupling is dominant, the matrix element of the electron-phonon interaction is given by (Blatt 1968, Nag 1972)

$$\langle k'n'|V_{s}|kn\rangle = \frac{1}{2} \left(\frac{k_{\rm B}T}{2\rho v_{\rm s}^2 V}\right)^{1/2} E_{\rm d} \delta_{k'_{x},k_{x}+q_{x}} \delta_{k'_{y},k_{y}+q_{y}} [\delta_{q_{z},(\pi/d)(n'-n)} + \delta_{q_{z},-(\pi/d)(n'-n)} - \delta_{q_{z},(\pi/d)(n'+n)} - \delta_{q_{z},-(\pi/d)(n'+n)}]$$
(13)

where  $\rho$  is the density of material,  $v_s$  is the sound velocity, q is the phonon wavevector and  $E_d$  is the deformation potential.

(B) When the piezoelectric coupling is dominant, one may obtain (Nag 1972)

$$\langle k'n'|V_{s}|kn\rangle = \frac{1}{2} \left( \frac{k_{B}T}{2\rho v_{s}^{2} V} \right)^{1/2} \frac{|e|\beta_{p}}{\varepsilon q} \delta_{k'_{s},k_{s}+q_{s}} \delta_{k'_{y},k_{y}+q_{y}} [\delta_{q_{z},(\pi/d)(n'-n)} + \delta_{q_{z},-(\pi/d)(n'-n)} - \delta_{q_{z},(\pi/d)(n'+n)} - \delta_{q_{z},-(\pi/d)(n'+n)}]$$
(14)

where  $\beta_p$  is the appropriate piezoelectric constant.

From equations (6)–(8), (10)–(12) and (13), the free-carrier absorption coefficient for a quasi-two-dimensional electron gas can be obtained as shown in appendix 1. From equations (A1.1) and (A1.7), it can be seen that the free-carrier absorption coefficient could be complex coming from the logarithmic functions due to the interaction between electrons, photons and phonons. Also, from equations (6)–(8), (10)–(12) and (14), the free-carrier absorption coefficient for a quasi-two-dimensional electron gas can be obtained as shown

in appendix 2. From equations (A2.1) and (A2.4), it can also be seen that the absorption coefficient could be complex coming from the logarithmic functions due to the interaction between electrons, photons and phonons. Since the radiation field and phonon field are AC fields, the interaction between the radiation field, the phonon field and conduction carriers could cause a complex expression for the free-carrier absorption coefficient. Thus the free-carrier absorption coefficient can be expressed as  $\alpha = \text{Re}(\alpha) + i \text{Im}(\alpha)$ . The imaginary part Im( $\alpha$ ) of the absorption coefficient contributes to the wavevector propagating with a combined AC field. Consequently, the refractive index of materials could be changed owing to the interaction between AC combined photon-phonon fields and conduction carriers in semiconductors.

#### 4. Numerical analysis and discussion

In this section, a numerical example is performed for an n-type GaAs thin film. The relevant values of physical parameters are taken to be (Wu and Tsai 1983)  $n_e = 1.73 \times 10^{15}$  cm<sup>-3</sup>,  $m^* = 0.07m_0$  ( $m_0$  is the mass of the free electron),  $\rho = 5.32$  g cm<sup>-3</sup>,  $\varepsilon = 12.9$ ,  $E_g = 1.51$  eV,  $E_d = 7$  eV,  $\beta_p = 4.71 \times 10^4$  esu cm<sup>-2</sup> and  $v_s = 3.6 \times 10^5$  cm s<sup>-1</sup>.

#### 4.1. Deformation-potential coupling

In figure 1(a), we plot the free-carrier absorption coefficient  $|\alpha|$  in n-type GaAs films with  $d = 1 \ \mu m$  as a function of the photon frequency for the radiation field polarized parallel to the layer plane for various temperatures. It is shown that  $|\alpha|$  decreases monotonically with increasing photon frequency and increases with increasing temperature. In this case, the real and imaginary parts of  $\alpha$  have the same order of numerical values (Wu 1993). These properties are in qualitative agreement with experimental data for the free-carrier absorption in n-type InAs at room temperature (Fan 1967) and some other experimental work on hot electrons (Uraoka *et al* 1992). However, if a magnetic field is applied, some oscillations with the photon frequency can be observed (Manasreh 1993). In figure 1(b) the free-carrier absorption coefficient  $\alpha$  in n-type GaAs films with  $d = 1 \ \mu m$  is plotted as a function of the photon frequency for the radiation field polarized perpendicular to the layer plane. Since Im( $\alpha$ )  $\simeq 0$ , hence  $\alpha \simeq |\alpha|$  in this case (Wu 1993). It is shown that  $\alpha$  decreases monotonically with increasing photon frequency and increases with increasing temperature.

In figure 2(a), we plot the free-carrier absorption coefficient  $[\alpha]$  in n-type GaAs films as a function of the film thickness with the photon frequency  $\Omega = 28$  THz (or the 10.6  $\mu$ m wavelength of a  $CO_2$  laser) for the radiation field polarized parallel to the layer plane. It is shown that  $|\alpha|$  decreases with increasing film thickness and increases with increasing temperature. In figure 2(b), the free-carrier absorption coefficient is plotted as a function of the film thickness with the photon frequency  $\Omega = 28$  THz (or the 10.6  $\mu$ m wavelength of a CO<sub>2</sub> laser) for the radiation field polarized perpendicular to the layer plane. In this case,  $Im(\alpha) \simeq 0$ ; thus  $\alpha = |\alpha|$  (Wu 1993). It can also be seen that  $\alpha$  decreases with increasing film thickness and increases with increasing temperature. However, the effect of temperature in this case is smaller than that shown in figure  $2(\bar{a})$ . When the photon energy  $\hbar\Omega$  is greater than the separation in energy between the various subbands, the absorption of a photon can occur with the simultaneous emission or absorption of phonons in transitions to the same or other subbands. Thus the free-carrier absorption coefficient decreases considerably. However, as the temperature increases, the phonon energy increases. Consequently, the effect of phonon scattering on conduction electrons becomes significant and the free-carrier absorption coefficient increases with increasing temperature. As the



Figure 1. The free-carrier absorption coefficient in n-type GaAs films due to deformationpotential coupling as a function of the photon frequency with  $d = 1 \ \mu m$  for the radiation field polarized (a) parallel and (b) perpendicular to the layer plane.

film thickness d decreases, the separation between adjacent subbands increases and the phonon-assisted transitions can take place only to states in the same subband.

# 4.2. Piezoelectric coupling

In figure  $3(\alpha)$ , the free-carrier absorption coefficient  $|\alpha|$  in n-type GaAs films is plotted as a function of the photon frequency with  $d = 10 \ \mu m$  for the radiation field polarized parallel to the layer plane. It shows that  $|\alpha|$  decreases monotonically with increasing photon frequency and increases with increasing temperature. For the radiation field polarized perpendicular to the layer plane as shown in figure 3(b), it is shown that the free-carrier absorption coefficient decreases monotonically with increasing photon frequency and decreases with increasing temperature. This is quite different from results for the deformation-potential coupling, and the piezoelectric coupling for the radiation field polarized parallel to the layer plane. In this case,  $Im(\alpha)$  is quite small compared with  $Re(\alpha)$  of the order of  $10^{-7}$ ; thus  $\alpha \simeq \operatorname{Re}(\alpha)$  (Wu 1993). In figure 4(a), we plot the free-carrier absorption coefficient  $|\alpha|$  in n-type GaAs films as a function of the film thickness with the photon frequency  $\Omega = 28$  THz (or the 10.6  $\mu$ m wavelength of a CO<sub>2</sub> laser) for the radiation field polarized parallel to the layer plane. It shows that  $|\alpha|$  decreases monotonically with increasing film thickness and increases with increasing temperature. In figure 4(b), the free-carrier absorption coefficient  $\alpha$  in n-type GaAs films is plotted as a function of the film thickness for the radiation field polarized perpendicular to the layer plane with  $\Omega = 28$  THz (or the 10.6  $\mu$ m wavelength of a CO<sub>2</sub> laser). In this case,  $Im(\alpha)$  is quite small compared with  $Re(\alpha)$ , of the order of  $10^{-6}$  (Wu 1993); thus  $\alpha \simeq \text{Re}(\alpha)$ . It shows that, at lower temperatures,  $\alpha$  decreases and oscillates with increasing film thickness. However, at high temperatures such as T = 300 K,  $\alpha$  decreases rapidly with increasing film thickness in the region of small film thicknesses



Figure 2. The free-carrier absorption coefficient in n-type GaAs films due to deformationpotential coupling as a function of the film thickness with  $\Omega = 28$  THz (or the 10.6  $\mu$ m wavelength of a CO<sub>2</sub> laser) for the radiation field polarized (a) parallel and (b) perpendicular to the layer plane.



Figure 3. The free-carrier absorption coefficient in n-type GaAs films due to piezoelectric coupling as a function of the photon frequency with  $d = 10 \ \mu m$  for the radiation field polarized (a) parallel and (b) perpendicular to the layer plane.



Figure 4. The free-carrier absorption coefficient in n-type GaAs films due to piezoelectric coupling as a function of the film thickness with  $\Omega = 28$  THz (or the 10.6  $\mu$ m wavelength of a CO<sub>2</sub> laser) for the radiation field polarized (*a*) parallel and (*b*) perpendicular to the layer plane.

and then decreases with a small oscillation as the film thickness increases. It can be seen that  $\alpha$  decreases with increasing temperature in the region  $d > 2 \times 10^{-1} \mu m$ . This is quite different from figure 4(a). The oscillatory behaviour arises here because of the phonon-assisted transitions between the subbands of various quantum sizes.

From our numerical results presented here, it is shown that the free-carrier absorption coefficient in n-type GaAs films depends upon the photon frequency, the film thickness and the temperature. The free-carrier absorption coefficient could be complex owing to the interaction between carriers, photons and phonons in semiconductors. However, when the radiation field is polarized perpendicular to the layer plane, the imaginary part of the free-carrier absorption coefficient becomes quite small compared with the real part of the absorption coefficient. Thus  $Im(\alpha) \simeq 0$  and  $\alpha \simeq Re(\alpha)$ . Moreover, since the energy gap of InSb is much smaller than that of GaAs, hence the imaginary part of the absorption coefficient will be reduced to a quite small value for the layer plane (Wu 1993). It can also be seen that the change in the free-carrier absorption coefficient with temperature for a given film thickness appears regular. These results are different from those of the deformation-potential coupling in n-type InSb films in which there are some irregular changes in the free-carrier absorption coefficient with temperature (Wu 1993).

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# Appendix 1. Deformation-potential coupling

(i) When the radiation field is polarized parallel to the layer plane,

$$\alpha = \alpha_{\rm D} \left[ \sum_{l=1}^{\infty} a_l \exp\left(-\frac{a_l}{2\tau}\right) \right]^{-1} \sum_{n_{\rm i}=1}^{\infty} \sum_{n_{\rm i}=1}^{N_{\rm f}} a_{n_{\rm i}} \exp\left(-\frac{a_{n_{\rm i}}}{2\tau}\right) \\ \times \left\{ k_T^2 d^2 a_{n_{\rm i}} \left[ -\frac{1}{4} \ln\left(\frac{U_{\rm if}^+}{U_{\rm if}^-}\right) + \frac{1}{8} \left(\frac{1}{U_{\rm if}^-} - \frac{1}{U_{\rm if}^+}\right) + 4\gamma n_{\rm i} n_{\rm f} \right] \right. \\ \left. + \frac{\pi^2}{\gamma} \left[ \frac{3}{16} \ln\left(\frac{U_{\rm if}^+}{U_{\rm if}^-}\right) - \frac{1}{4}\gamma \sum_{\lambda=-1}^{+1} \left(\lambda (n_{\rm f} + \lambda n_{\rm i})^2 \ln(\frac{4}{3}U_{\rm if}^{\lambda}) \right) \right. \\ \left. + \frac{1}{32} \sum_{\lambda=-1}^{+1} \left(\lambda (U_{\rm if}^{\lambda})^{-1} + \frac{1}{3}\gamma n_{\rm i} n_{\rm f} \right] \right\}$$

$$(A1.1)$$

where

$$\alpha_{\rm D} = \frac{2\pi^4 e^2 n_e k_{\rm B} T E_{\rm d}^2}{\Omega^3 \varepsilon^{1/2} \rho v_{\rm s}^2 m^* c d^3 \hbar^2}.$$
(A1.2)

$$\gamma = (\pi v_{\rm s}/d\Omega)^2 \tag{A1.3}$$

$$k_{\rm B}T = \tau E_{\rm g} = \hbar^2 k_T^2 / 2m^* \tag{A1.4}$$

$$U_{\rm if}^{\pm} = \gamma (n_{\rm f} \pm n_{\rm i})^2 - \frac{1}{4}$$
(A1.5)

and  $N_{\rm f}$  can be determined from

$$n_{\rm i} < N_{\rm f} \leqslant n_{\rm i} + d\Omega/\pi v_{\rm s}. \tag{A1.6}$$

(ii) When the radiation field is polarized perpendicular to the layer plane,

$$\alpha = \alpha_{\rm D} \bigg[ \sum_{l=1}^{\infty} a_l \exp\left(-\frac{a_l}{2\tau}\right) \bigg]^{-1} \sum_{n_i=1}^{\infty} \sum_{n'=1}^{N'} \sum_{n'=1}^{N'} (n')^2 a_{n_i} \\ \times \left(\frac{1 - \cos[\pi(n_{\rm f} + n')]}{n_{\rm f} + n'} + \frac{1 - \cos[\pi(n_{\rm f} - n')]}{n_{\rm f} - n'}\right)^2 \\ \times \exp\left(-\frac{a_{n_i}}{2\tau}\right) \bigg[ \sum_{l=-1}^{+1} ' \ln\left(\frac{V_{\rm if}'^+ + \lambda E_{\rm if}'}{V_{\rm if}'^- + \lambda E_{\rm if}'}\right) \\ + \sum_{\mu=-1}^{+1} ' \sum_{\lambda=-1}^{+1} ' (V_{\rm if}'^{\mu} - \lambda) \exp(\lambda V_{\rm if}'^{\mu} - E_{\rm if}') E_{\rm i}(-\lambda V_{\rm if}'^{\mu} + E_{\rm if}') \bigg]$$
(A1.7)

where

$$V_{\rm if}^{\prime\pm} = \frac{\pi v_{\rm s} h}{dk_{\rm B} T} (n' \pm n_{\rm i}) (a_{n_{\rm f}} + a_{n'}) (a_{n_{\rm f}} - a_{n'})$$
(A1.8)

$$E_{\rm if}' = \frac{E_{\rm g}}{2k_{\rm B}T} \frac{a_{n_{\rm i}}a_{n'} + a_{n_{\rm f}}^2 - a_{n'}^2 - a_{n_{\rm i}}a_{n_{\rm f}}}{a_{n_{\rm f}} - a_{n'}} - \frac{\hbar\Omega a_{n_{\rm f}}}{k_{\rm B}T(a_{n_{\rm f}} - a_{n'})}$$
(A1.9)

and the exponential-integral function  $E_i(x)$  is defined as (Gradshteyn and Ryzhik 1965, Abramowitz and Stegun 1968)

$$E_{i}(x) = -\int_{x}^{\infty} \frac{\exp(-t)}{t} dt = \int_{-\infty}^{x} \frac{\exp(t)}{t} dt.$$
 (A1.10)

N' and  $N_{\rm f}$  are determined from

$$n_{\rm i} < N' < N_{\rm f} \leqslant n_{\rm i} + d\Omega/\pi v_{\rm s}. \tag{A1.11}$$

# Appendix 2. Piezoelectric coupling

(i) When the radiation field is polarized parallel to the layer plane,

$$\alpha = \alpha_{\rm P} \left[ \sum_{l=1}^{\infty} a_l \exp\left(-\frac{a_l}{2\tau}\right) \right]^{-1} \sum_{n_i=1}^{\infty} \sum_{n_r=1}^{N_{\rm f}} a_{n_i} \exp\left(-\frac{a_{n_i}}{2\tau}\right) \\ \times \left\{ \frac{2m^* d^2 \gamma k_{\rm B} T a_{n_i}}{\pi^2 \hbar^2} \left[ 8 \ln\left(\frac{n_{\rm f} + n_{\rm i}}{n_{\rm f} - n_{\rm i}}\right) - 3 \ln\left(\frac{U_{\rm if}^+}{U_{\rm if}^-}\right) - \sum_{\lambda=-1}^{+1} \frac{\lambda}{2} (U_{\rm if}^{\lambda})^{-1} \right] \right] \\ + 2\gamma n_i n_{\rm f} \ln(16U_{\rm if}^+ U_{\rm if}^-) - 8\gamma n_i n_{\rm f} (\frac{1}{2} \ln \gamma + \frac{1}{3}) \\ - 2\sum_{\lambda=-1}^{+1} \frac{\lambda}{\lambda} (n_{\rm f} + \lambda n_{\rm i})^2 \ln(n_{\rm f} + \lambda n_{\rm i}) - 4(\ln 3)\gamma n_{\rm i} n_{\rm f} \\ + \frac{1}{4} [1 + 4\gamma (n_{\rm f}^2 + n_{\rm i}^2)] \ln\left(\frac{U_{\rm if}^+}{U_{\rm if}^-}\right) \right\}$$
(A2.1)

where

$$\alpha_{\rm P} = \frac{2\pi^6 e^4 n_{\rm e} k_{\rm B} T \beta_{\rm P}^2}{\Omega^3 \varepsilon^{5/2} \rho v_{\rm s}^2 m^* c d^3 \hbar^2} \tag{A2.2}$$

and  $N_{\rm f}$  can be determined from

$$n_{\rm i} < N_{\rm f} \leqslant n_{\rm i} + d\Omega/\pi v_{\rm s}. \tag{A2.3}$$

(ii) When the radiation field is polarized perpendicular to the layer plane,

$$\alpha = \alpha_{\rm P} \frac{\gamma \hbar^2 \Omega^2}{k_{\rm B} T \pi^2} \bigg[ \sum_{l=1}^{\infty} a_l \exp\left(-\frac{a_l}{2\tau}\right) \bigg]^{-1} \sum_{n_{\rm f}=1}^{\infty} \sum_{n_{\rm f}=1}^{N_{\rm f}} \sum_{n'=1}^{N'} \frac{(n')^2 a_{n_{\rm f}} a_{n'}^2}{(a_{n_{\rm f}} - a_{n'})^2} \\ \times \bigg[ \frac{1 - \cos[\pi (n_{\rm f} + n')]}{n_{\rm f} + n'} + \frac{1 - \cos[\pi (n_{\rm f} - n')]}{n_{\rm f} - n'} \bigg]^2 \\ \times \exp\left(-\frac{a_{n_{\rm i}}}{2\tau}\right) \bigg\{ 2\ln\left(\frac{n' + n_{\rm i}}{n' - n_{\rm i}}\right) \bigg[ (k_{\rm B} T)^{-1} \exp\left(\frac{K'_{\rm if}}{k_{\rm B} T}\right) E_{\rm i}\left(-\frac{K'_{\rm if}}{k_{\rm B} T}\right) - (K'_{\rm if})^{-1} \bigg] \\ + \sum_{\lambda=-1}^{+1} \sum_{\mu=-1}^{+1} \lambda P[K'_{\rm if}, \mu W'_{\rm if}] + (a_{n_{\rm f}} - a_{n'}) \sum_{\lambda=-1}^{+1} \lambda Q[K'_{\rm if}, W'_{\rm if}] \bigg\}$$
(A2.4)

where

$$K'_{\rm if} = \hbar \Omega a_{n_{\rm f}} (a_{n_{\rm f}} - a_{n'})^{-1} - \frac{1}{2} E_{\rm g} (a_{n_{\rm f}} a_{n'} + a_{n_{\rm f}}^2 - a_{n'}^2 - a_{n_{\rm i}} a_{n_{\rm f}}) (a_{n_{\rm f}} - a_{n'})^{-1}$$
(A2.5)

$$W_{\rm if}^{\prime\pm} = (\pi \hbar v_{\rm s}/d)(n'\pm n_{\rm i})(a_{n_{\rm f}}+a_{n'})(a_{n_{\rm f}}-a_{n'})^{-1} \tag{A2.6}$$

$$\begin{split} P(K, V) &= -k_{\rm B}T \left\{ \left[ 5 - 2\ln\left(\frac{2m^*a_{n_{\rm I}}d^2V}{\pi^2\hbar^2}\right) \right] K^{-1}(K+V)^{-1} \right. \\ &+ \left[ 2 - 2\ln\left(\frac{4m^{*2}a_{n_{\rm I}}^2d^4KV}{\pi^4\hbar^4}\right) \right. \\ &+ \ln\left(\frac{2m^*a_{n_{\rm I}}d^2K}{\pi^2\hbar^2}\right) \ln\left(\frac{2m^*a_{n_{\rm I}}d^2V}{\pi^2\hbar^2}\right) \right] (K+V)^{-2} \\ &+ \left[ \ln\left(\frac{2m^*a_{n_{\rm I}}d^2V}{\pi^2\hbar^2}\right) - 1 \right] \ln\left(\frac{2m^*a_{n_{\rm I}}d^2(K+V)}{\pi^2\hbar^2}\right) K^{-2} \right\} \\ &+ \left[ 1 - \ln\left(\frac{4m^{*2}a_{n_{\rm I}}^2d^4KV}{\pi^4\hbar^4}\right) \right] (K+V)^{-1} \\ &- (2k_{\rm B}TK^{-2} - K^{-1}) \exp\left(\frac{V}{k_{\rm B}T}\right) E_{\rm i}\left(-\frac{V}{k_{\rm B}T}\right) \\ &+ V^{-1} \exp\left(\frac{K}{k_{\rm B}T}\right) E_{\rm i}\left(\frac{K}{k_{\rm B}T}\right) + (k_{\rm B}T)^{-1} \exp\left(\frac{K+V}{k_{\rm B}T}\right) E_{\rm i}\left(-\frac{V}{k_{\rm B}T}\right) \\ &- \left[ 3K^{-1} + V^{-1} - (k_{\rm B}T)^{-1} \ln\left(\frac{4m^{*2}a_{n_{\rm I}}^2d^4KV}{\pi^4\hbar^4}\right) - 2k_{\rm B}TK^{-2} \right] \\ &\times \exp\left(\frac{K+V}{k_{\rm B}T}\right) E_{\rm i}\left(-\frac{K+V}{k_{\rm B}T}\right) \end{split}$$
(A2.7)

and

$$Q(K, V) = V^{-1} \left[ \exp\left(\frac{K+V}{k_{\rm B}T}\right) E_{\rm i} \left(-\frac{K+V}{k_{\rm B}T}\right) - \exp\left(\frac{K-V}{k_{\rm B}T}\right) E_{\rm i} \left(-\frac{K-V}{k_{\rm B}T}\right) \right].$$
(A2.8)

N' and  $N_{\rm f}$  can be determined from

$$n_{\rm i} < N' < N_{\rm f} \leqslant n_{\rm i} + d\Omega/\pi v_{\rm s}. \tag{A2.9}$$

The sums over  $\lambda$  or  $\mu$  with  $\sum'$  indicate  $\lambda$ ,  $\mu = -1$  to +1 without 0. For the notation for  $\lambda$  or  $\mu$ , +1 means '+', and -1 means '-'.

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