Theory of the spectral line shape and gain in quantum wells with intersubband transitions

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We investigate the spectral line shape of radiative intersubband transitions in a quantum well as determined by two factors: the electron scattering rate from states of given energy and the mass difference between the two subbands involved. The interplay between these factors leads to an essentially non-Lorentzian form of the spectral line. We develop an analytic theory of the line shape and calculate the dependence of the intersubband optical gain in a quantum well on both the population inversion and the temperature. Under typical conditions, the effect of electron temperature on the gain is similar to that of the lattice temperature, which points to the importance of hot carrier effects in understanding the behavior of intersubband lasers. © *1996 American Institute of Physics.* [S0003-6951(96)00916-4]

The quantum cascade laser (QCL)[1–3] based on intersubband transitions in a quantum well (QW) is a serious contender for various applications in the midinfrared range ($\lambda \gtrsim 4$ μm). Because the QW subbands are nearly parallel, the QCLs are commonly considered by analogy with a two-level laser, where the temperature dependence of output characteristics is controlled by the *width* of the spontaneous emission line, the latter having the usual Lorentzian shape.[4] The subband nonparabolicity is then taken into account as a refinement similar to inhomogeneous broadening.

As shown below, this approach is valid only at very low electron temperatures T_e. However, in a practical QCL, even at low temperatures T, the effective T_e may be rather high. This results in a significant broadening of interband transitions by phonon emission and other collisions, which leads to the situation where the spectral power at each wavelength is due to transitions from a broad range of initial states. Therefore, a consistent treatment of the line shape problem requires that both the collision broadening and the nonparabolicity effects are taken into account on equal footing from the outset. In this letter we develop a first-principles temperature-dependent model for the intersubband line shape and the resulting optical gain in a QW, which is consistent with the experimental data available. The model has been successfully used[5] to predict the behavior of QCL as controlled by both T and T_e.

We shall consider a QW with infinitely high walls—ignoring the question of how electrons are pumped into the upper subband and how they are removed from the QW. The ratio of electron populations n_1/n_2 in the two subbands will thus be regarded as a parameter. Of all the scattering processes we shall include only the dominant interaction with polar optical phonons, and neglect impurity scattering and electron-electron interaction. These simplifying assumptions enable a consistent analytic treatment of both the nonparabolicity and the transverse relaxation due to scattering; moreover, the evaluation of gain is reduced to one quadrature.

The nonparabolicity in a QW is estimated from the Kane model,[6] including the interaction between six valence subbands and two degenerate conduction bands and neglecting the spin-orbit interaction. For a QW bounded by infinite walls, equations of this model (written in terms of envelope functions) admit an exact solution, which gives the dispersion relations in both subbands in the form:

$$E_n(k)=\frac{E_G}{2}\left[1+\frac{4E_n^{(0)}}{E_G}+\frac{2\hbar^2k^2}{m_eE_G}\right]^{1/2}-\frac{E_G}{2}, \tag{1}$$

where m_e is the Kane effective mass at the conduction band bottom, E_G is the semiconductor band gap, and $E_n^{(0)} \equiv \pi^2\hbar^2n^2/2m_ea^2$, $n=1,2$, are the QW energy levels in the parabolic approximation. Expanding in k, we find the effective masses at the bottom of the two subbands

$$m_n=m_e\left(1+\frac{2E_n(0)}{E_G}\right). \tag{2}$$

For an InGaAs QW with $E_2(0)-E_1(0)\equiv\hbar\Omega_0=0.3$ eV, the effective mass ratio is rather large, $m_2/m_1=1.5$. The nonparabolic subband structure is illustrated in Fig. 1, which also shows electronic transitions, both radiative and nonradiative.

The line shape and the gain are determined from the dynamic conductivity related to the intersubband transition, following the well-known techniques.[7] First we solve the density matrix equations for a two level system in the presence of an electromagnetic wave at the optical frequency Ω:

$$\hbar\Omega\rho_{12}-[H,\rho]_{12}+i\hbar\gamma\rho_{12}=eF[z,\rho]_{12}, \tag{3a}$$

$$\hbar\Omega\rho_{21}-[H,\rho]_{21}+i\hbar\gamma\rho_{21}=eF[z,\rho]_{21}, \tag{3b}$$

where F is the electric field. The Hamiltonian H entering Eqs. (3) for the off-diagonal elements of the density matrix is itself diagonal, but includes the nonparabolicity. Equations (3) describe a particular ''vertical'' transition

$$\hbar\Omega(\epsilon)=\hbar\Omega_0+\epsilon_2-\epsilon_1, \tag{4}$$

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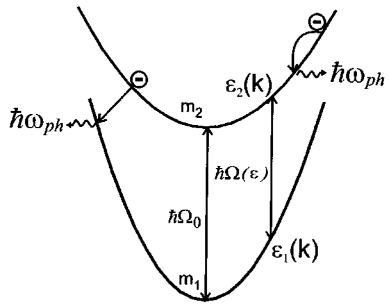

FIG. 1. The subband diagram in an infinite quantum well. The direct intersubband radiative transition energy $\hbar\Omega$ depends on the wave vector of the initial state. The largest separation between the subbands $E_2(k)$ and $E_1(k)$ is denoted by $\hbar\Omega_0$. Also shown are inter- and intra-subband transitions with emission of a polar optic phonon $\hbar\omega_{ph}$.

where $\epsilon_n(k)\equiv E_n(k)-E_n(0)$, $n=1,2$. The transition can be designated by $\epsilon\equiv\epsilon_2$, since for a given k (neglecting the photon momentum) the energy ϵ_2 determines ϵ_1 and vice versa (see Fig. 1). The damping term $i\hbar\gamma\rho_{12}$ describes the transverse phase relaxation, where $\gamma=\gamma(\epsilon)$ results mainly from the intrasubband optical phonon scattering.

The intersubband dipole moment $\langle z\rangle=z_{12}\rho_{21}+z_{21}\rho_{12}$, corresponding to transition (4) equals

$$\langle z\rangle=eF|z_{12}|^2(\rho_{22}-\rho_{11})\left(\frac{1}{\Omega+\Omega(\epsilon)+i\gamma}+\frac{1}{-\Omega+\Omega(\epsilon)-i\gamma}\right), \tag{5}$$

where $\rho_{22}=f_2(\epsilon_2)$ and $\rho_{11}=f_1(\epsilon_1)$ are occupation probabilities in each subband.

Integrating (5) over all possible transitions, we obtain the susceptibility, $\chi\cdot F=(1/2\pi^2)\int d^2ke\langle z\rangle$, and the dynamic conductivity, $\sigma_{zz}=\Omega\ \mathrm{Im}\ \chi$. When averaging over an optical period, we find the power absorption (or generation) per unit QW area, $W_{st}=\sigma_{zz}F^2/2$,

$$W_{st}=\frac{16e^2|z_{12}|^2m_2\Omega^2N_q}{\hbar\kappa_\infty V}\times\int_0^\infty\frac{d(\epsilon/\hbar)\,\gamma(\epsilon)\Omega\Omega(\epsilon)[f_2(\epsilon)-f_1(\epsilon_1)]}{\{[\Omega(\epsilon)]^2-(\Omega^2-\gamma^2)\}^2+4\gamma^2\Omega^2}, \tag{6}$$

where κ_∞ is the dielectric permittivity at optical frequencies and V the QW volume. In accordance with the quantum mechanical correspondence principle, we have replaced the energy density of radiation by the number of photons per unit volume as follows: $(\kappa_\infty/8\pi)F^2=\hbar\Omega N_q/V$. Equation (6) describes stimulated processes (absorption and emission) due to the interaction with a photon mode q. Spontaneous emission into the same mode is found from Einstein's relation.

An expression for the optical gain $g(\Omega)$ is found from Eq. (6) and the usual relation $(\hbar\Omega cN_q/V\sqrt{\kappa_\infty})g=W_{st}/a$ between W_{st} and g, where a is the QW width and c the speed of light. In the natural limit $\gamma(\epsilon)\ll\Omega$ this expression reduces to the following form:

$$g=\frac{4\pi e^2|z_{12}|^2m_2\Omega}{\hbar^3ac\sqrt{\kappa_\infty}}\int_0^\infty d\epsilon\mathscr{T}_\epsilon(\Omega)[f_2(\epsilon)-f_1(\epsilon_1)], \tag{7}$$

where $\mathscr{T}_\epsilon(\Omega)$ is the line shape function:

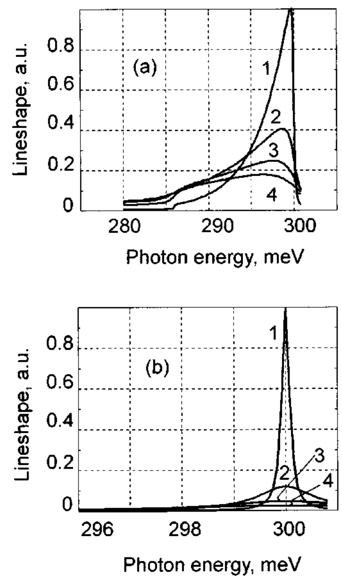

FIG. 2. Calculated spontaneous emission spectra at $T_e=T$ ranging from 100 to 400 K (curves 1–4, respectively). (a) Assumed material parameters: $E_G=1$ eV, $m_e=0.04m_0$, and $a=76$ Å, resulting in $E_1=138$ eV, $E_2=438$ eV, and $m_1=1.28m_e$, $m_2=1.88m_e$. (b) Same spectra calculated in the parabolic model, $m_2=m_1$.

$$\mathscr{T}_\epsilon(\Omega)\equiv\frac{\gamma(\epsilon)/\pi}{[\Omega-\Omega(\epsilon)]^2+[\gamma(\epsilon)]^2}. \tag{8}$$

Spontaneous emission line in the same limit is:

$$W_{sp}=\frac{4\pi e^2|z_{12}|^2m_2\Omega^2}{\hbar^2\kappa_\infty V}\int_0^\infty d\epsilon\mathscr{T}_\epsilon(\Omega)f_2(\epsilon)[1-f_1(\epsilon_1)]. \tag{9}$$

The validity of Eqs. (6)–(9) is not restricted to any particular scattering mechanism, responsible for the transverse phase relaxation. In order to proceed with exemplary calculations, we need to make certain assumptions about $\gamma(\epsilon)$. We assume that γ is dominated by the interaction with polar optical phonons.[8–10] Only intrasubband scattering need be considered, since in a narrow QW intersubband processes are much slower due to the large momentum transfer involved.[10] For a sufficiently narrow QW of any shape, one finds[8,9]

$$\gamma(\epsilon)=\frac{\pi}{2}\frac{e^2}{\hbar}\left(\frac{1}{\kappa_0}-\frac{1}{\kappa_\infty}\right)q_{ph}\times\begin{cases}N_{ph}\\(N_{ph}+1)\,\theta(\epsilon-\hbar\omega_{ph}),\end{cases} \tag{10}$$

where the top line corresponds to absorption and the bottom line to emission of optical phonons, N_{ph} is the phonon Planck function, $\theta(\epsilon)$ is a step function, and $q_{ph}=\sqrt{2m_e\omega_{ph}/\hbar}$. The steplike nature of $\gamma(\epsilon)$ is important in the line shape formation. However, the ultimate sharpness of the step, peculiar to the QW case, is not essential: results obtained with the 3D scattering rate function are similar.

In the numerical examples presented below, the ratio $\xi=n_1/n_2$ between the electron populations will be regarded as an independent parameter, essentially governed by the kinetics of intersubband transitions and electron removal from the QW. Electron distributions in the two subbands will be assumed in a quasiequilibrium form, characterized by Fermi functions, corresponding to an electron temperature T_e.

Figure 2(a) shows the calculated spectral line shapes for several different T_e. Even though it is assumed that the elec-

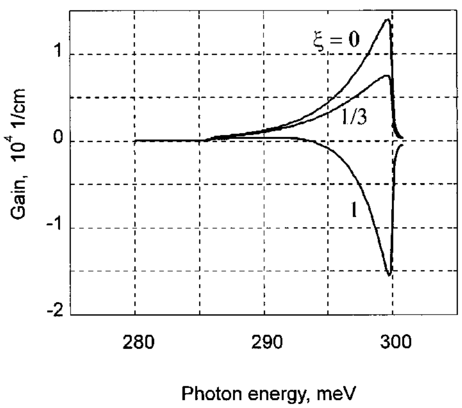

FIG. 3. Calculated intersubband gain spectra for material parameters as in Fig. 2(a) and $n_2 = 10^{11}$ cm^{-2} for different values of the population inversion parameter $\xi = n_1/n_2$.

tron and the lattice temperatures coincide, we stress that calculations with a constant lattice temperature $T=100$ K and only T_e varying give practically identical results. When T_e strongly departs from T, the line shape is mainly determined by T_e, since the only effect of $T \ll T_e$ is through the factor N_{ph} in Eq. (10). This is a noteworthy result: it shows that carrier heating is as important for the operation of an intersubband laser as is the ambient temperature.

For comparison, Fig. 2(b) shows the spectra calculated in the parabolic model $m_1 = m_2$. We see that the nonparabolicity has a dramatic effect on the line shape. Not only does it broaden the spectra considerably in the long wavelength direction, but most importantly, the maximum spectral intensity is no longer determined by the linewidth.

The gain spectra calculated from Eq. (7) are presented in Fig. 3 for parameters similar to those in Fig. 2 with $n_2 = 10^{11}$ cm^{-2} and the population inversion parameter $\xi \equiv n_1/n_2 = 1/3$. We see that the gain can be positive even in the absence of inversion between the two subbands, i.e., for $n_1 \gtrsim n_2$ [see Fig. 3 (for $\xi=1$)]. In the model[11] where both subbands are characterized by the same T_e this effect occurs due to nonparabolicity: at a sufficiently high wave vector k the occupation probability of state $\epsilon_2(k)$ in the upper subband is higher than that of state $\epsilon_1(k)$, even though the lower subband has higher overall population.

The characteristic kink seen in Figs. 2 and 3 in the long wave portion of the spectra, occurs at $\Omega_{ph} = \Omega(\epsilon = \hbar\omega_{ph}) \approx \Omega_0 - (m_2/m_1 - 1)\omega_{ph}$ and reflects the fact that transitions corresponding to $\epsilon > \hbar\omega_{ph}$ suffer a steplike increase in the broadening by optical phonon emission. Accordingly, the spectra are depressed at $\Omega < \Omega_{ph}$ and at the same time enhanced in the vicinity of Ω_{ph} on the $\Omega > \Omega_{ph}$ side. Because of this, at high T_e the gain spectra exhibit a second maximum near Ω_{ph} which may lead to a shift in the lasing frequency with increasing current.[5]

In the entire range of temperatures presented in Fig. 3 the peak value of the gain is suppressed compared to the parabolic case by at least an order of magnitude. The calculated peak gain is shown in Fig. 4(a) as a function of T_e for different values of ξ. It should be kept in mind that the peak in gain occurs at different wavelengths for different temperatures, shifting to longer wavelengths with higher T_e. The

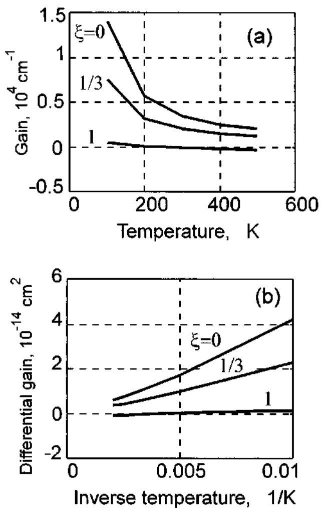

FIG. 4. Temperature dependence of (a) the peak gain $g_{\max}$ and (b) the differential gain g_n' for different values of the population inversion parameter $\xi = n_1/n_2$.

highest gain achievable at given n_2 and T_e corresponds to $\xi=0$; in the present case it is about 1.5×10^4 cm^{-1}.

Equations (7) and (8) yield a convenient expression for the differential gain g_n'. Temperature dependence of g_n' is shown in Fig. 4(b). Plotted against $1/T_e$ the function g_n' is rather well approximated by a straight line in a wide range of n_1/n_2. The high values of g_n' suggest that intersubband lasers will have a superior high-frequency performance.

In summary, we have developed a theory of the intersubband optical gain which takes into account the transverse degrees of freedom of QW electrons. In essence, we have replaced the conventional two-level model of intersubband transitions by a two-band model, which includes from the first principles such effects as energy-dependent scattering and the subband nonparabolicity. We have shown that inclusion of these effects leads to a qualitative change in the line shape of the intersubband resonance. This implies that the electron heating effects are of paramount importance in understanding the behavior of intersubband lasers.

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[11] This is a reasonable model for the experimental conditions reported in Refs. 1–3, where the average carrier concentration per period ($\gtrsim 10^{17}$ cm^{-3}) is sufficiently high to establish a uniform T_e throughout the entire period (~400 Å). A very interesting situation arises in the opposite limit of low carrier concentrations, where the rate of electron-electron collisions is too low to establish Maxwellian distribution functions in the two subbands. In this limit (Ref. 5), the existence of a positive gain persists to concentrations far from an overall inversion and does not rely on the nonparabolicity.