# Collective Intersubband Excitations in Quantum Wells: Coulomb Interaction versus Subband Dispersion

Dmitri E. Nikonov,[1,*] Ataç Imamoğlu,[1] Leonid V. Butov,[1,2] and Holger Schmidt[1]
[1]*Department of Electrical and Computer Engineering and Center for Quantized Electronic Structures (QUEST), University of California, Santa Barbara, California 93106-9560*
[2]*Institute of Solid State Physics, Russian Academy of Sciences, Chernogolovka, 142432, Russia*
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The dependence of the intersubband absorption line shape on the subband dispersion and the Coulomb interaction between electrons is studied using the Hartree-Fock semiconductor Bloch equations. For subbands with the same effective masses, we show that the absorption/emission line shape is solely determined by the single-electron properties. For very different effective masses, the Coulomb interaction results in a strong redistribution of the oscillator strength. The line shape in this limit is determined by a Fermi-edge collective mode induced by a repelling electron-hole pair. [S0031-9007(97)04616-4]

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The recent demonstration of laser oscillation in intersubband transitions of unipolar quantum wells (QW) [1], observation of Fano interference induced by tunneling in such structures [2,3], and related proposals for semiconductor lasers without inversion [4] make it necessary to revisit the problem of the influence of electron-electron interactions on the line shape of intersubband transitions. The factors determining the shape of the absorption or gain spectrum and the nature of line broadening (inhomogeneous, associated with single particles, or homogeneous, associated with collective modes) are especially important for the laser development.

Electron-electron Coulomb interactions in intersubband transitions can be subdivided into the direct and exchange Coulomb interaction and the depolarization caused by the electron plasma in a specific quantum well structure. It has been shown that these factors determine the nature of intersubband transitions in inelastic light scattering [5]. Another important issue is the dispersion which is expressed as the difference in effective masses of the subbands and is commonly referred to as "nonparabolicity." The influence of the subband dispersion and Coulomb interaction on the intersubband absorption spectra has been considered in [6–8]. It has been shown that the inclusion of only the depolarization terms results in a narrowing of the absorption linewidth.

In this Letter we analyze the relation between the Coulomb interaction and the subband dispersion that provides a new understanding of the collective modes that shape the absorption/gain spectra. Using the semiconductor Bloch equations [9] applied to intersubband transitions, we analytically and numerically demonstrate that (1) in the limit of a small difference of effective masses and in the ideal two-dimensional limit, the influence of exchange terms on the absorption or gain spectrum disappears, which is a generalization of Kohn's theorem for intersubband transitions in arbitrary confinement potentials; (2) in the limit of a large difference of effective masses, the Coulomb interaction results in a strong redistribution of the oscillator strength and thereby makes a wide spectrum associated with the single-particle subband dispersion disappear. The absorption or gain spectrum is a narrow peak determined by a homogeneously broadened collective excitation. The latter effect is explained in terms of a Fermi-edge singularity of a repelling electron-hole pair for two-dimensional systems where exchange terms dominate, and in terms of a negative-mass intersubband exciton when the depolarization terms are dominant.

Consider one-electron states in each subband $\mu$ of the conduction band having a certain momentum $\mathbf{k}$ in the plane of the quantum well (Fig. 1). The Hamiltonian for subbands interacting with electromagnetic field [9] in the rotating frame (see [10]) has the form

$$H = \sum_{\mu,\mathbf{k}} \hbar\Delta_{\mu\mathbf{k}} a^{+}_{\mu\mathbf{k}} a_{\mu\mathbf{k}} - \sum_{\{\mu,\nu\}\mathbf{k}} (\hbar\Omega_{\mu\nu\mathbf{k}} a^{+}_{\mu\mathbf{k}} a_{\nu\mathbf{k}} + \hbar\Omega^{*}_{\mu\nu\mathbf{k}} a^{+}_{\nu\mathbf{k}} a_{\mu\mathbf{k}}) + \frac{1}{2} \sum_{\mu\nu\nu'\mu'\mathbf{k}\mathbf{k}',q} \hbar V^{\mu\nu\nu'\mu'}_{\mathbf{q}} a^{+}_{\mu,\mathbf{k}+\mathbf{q}} a^{+}_{\nu,\mathbf{k}'-\mathbf{q}} a_{\nu',\mathbf{k}'} a_{\mu',\mathbf{k}}\,, \tag{1}$$

where $\Delta_{\mu\mathbf{k}}$ is the detuning of the state $\mu, \mathbf{k}$ from the field, $\Omega_{\mu\nu\mathbf{k}} = \wp_{\mu\nu\mathbf{k}}\mathcal{E}/\hbar$ is the Rabi frequency for an electromagnetic field amplitude $2\mathcal{E}$ applied between an upper subband $\mu$ and a lower subband $\nu$, $\wp_{\mu\nu} = \int f^{*}_{\mu} z f_{\nu}\, dz$ is the dipole matrix element, $f_{\mu}(z)$ is the envelope wave function in the state $\mu$ of the quantum well, and $a_{\mu\mathbf{k}}$ are the electron creation and annihilation operators satisfying $\{a_{\mu\mathbf{k}}, a^{+}_{\nu\mathbf{k}'}\} = \delta_{\mu\nu}\delta_{\mathbf{k}\mathbf{k}'}$. The Coulomb interaction terms for two subbands are nonzero only for $\mu = \mu'$, $\nu = \nu'$ (direct and exchange terms), or for $\mu = \nu' \neq \nu = \mu'$ (depolarization) [5]; the corresponding form factors are

  

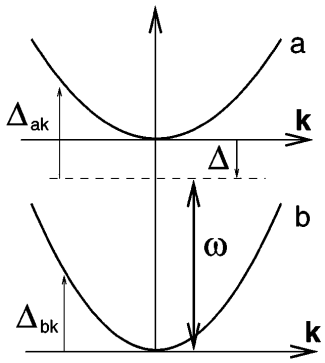


FIG. 1. The scheme of the momentum states of the two subbands $a$ and $b$ coupled by a laser field with the frequency $\omega$. Here $\Delta_{a\mathbf{k}}$ and $\Delta_{b\mathbf{k}}$ are detunings of the states from the field, and $\Delta = -\Delta_{a0}$.

$$V_{\mathbf{q}}^{\mu\nu\nu'\mu'} = V_{\mathbf{q}} \int dz\, dz'\, f_{\mu}(z) f_{\mu'}(z) e^{-q|z-z'|} f_{\nu}(z') f_{\nu'}(z')\,. \tag{2}$$

The Fourier transform of the Coulomb potential in two dimensions is

$$V_{\mathbf{q}} = \frac{e^2}{2\hbar\varepsilon_0\varepsilon_b\varepsilon(\mathbf{q})\,|\mathbf{q}|}\,, \tag{3}$$

where $\varepsilon_b$ is the background dielectric constant. The near-resonant screening [11] is taken into account approximately via the dielectric function $\varepsilon(\mathbf{q})$ given by the static Lindhard formula [9]. All direct terms ($\mathbf{q} = 0$) cancel with the interaction energy of the ions of the crystal lattice; exchange terms do not contain $\mathbf{q} = 0$ contributions. The depolarization is much smaller than the exchange terms for $\mathbf{q} \neq 0$, and, for the present discussion, we retain it only for $\mathbf{q} = 0$. The ratio of depolarization terms to the exchange terms in the equations of motion is seen to have a characteristic value proportional to $k_F a_W$, where $k_F$ is the Fermi momentum and $a_W$ is the effective width of the well given by the integral in (2). The depolarization contribution grows with the electron density and the width of the well. For a truly two-dimensional electron gas, i.e., in the limit of $k_F a_W \ll 1$, the depolarization becomes negligible, and the form factors become $V_{\mathbf{q}}^{\mu\nu\nu'\mu'} = V_{\mathbf{q}}\delta_{\mu\mu'}\delta_{\nu\nu'}$. This limit is realized, e.g., in a GaAs quantum well with transition energies of the order of 0.1 eV for electron densities less than $10^{11}$ cm$^{-2}$.

The Hamiltonian of Eq. (1) along with phenomenological relaxation terms allows us to obtain the equations of motion for the reduced density matrix $\sigma_{\mu\nu\mathbf{k}} = \langle a_{\nu\mathbf{k}}^{+} a_{\mu\mathbf{k}}\rangle$, where $\mathbf{k}$ is the in-plane momentum and the brackets denote a quantum statistical average. The relaxation terms, which we treat in the rate-equation approximation (see [10]), correspond to collisions between electrons with rate $\gamma_{\text{coll}}$, dephasing of the polarizations between subbands with rate $\gamma_{\text{deph}}$, and transitions with spontaneous emission of phonons or photons with rate $w_{ab}$. In the Hartree-Fock approximation, the semiconductor Bloch equations for the polarizations and populations in the two coupled subbands, upper $a$ and lower $b$, are [5]

$$\dot{\sigma}_{aa\mathbf{k}} = i\Omega\sigma_{ba\mathbf{k}} - i\Omega^{*}\sigma_{ab\mathbf{k}} - w_{ab}\sigma_{aa\mathbf{k}} + \dot{\sigma}_{aa\mathbf{k}}|_{\text{coll}} + i\sum_{\mathbf{q}\neq 0} V_{\mathbf{q}}^{abba}(\sigma_{ba\mathbf{k}}\sigma_{ab\mathbf{k}+\mathbf{q}} - \sigma_{ab\mathbf{k}}\sigma_{ba\mathbf{k}+\mathbf{q}}) + iV_0^{abab}\sum_{\mathbf{k}'}(\sigma_{ba\mathbf{k}'}\sigma_{ab\mathbf{k}} - \sigma_{ab\mathbf{k}'}\sigma_{ba\mathbf{k}}) \tag{4}$$

$$\dot{\sigma}_{ab\mathbf{k}} = -\gamma_{ab}\sigma_{ab\mathbf{k}} + i(\Delta - \delta_{\mathbf{k}})\sigma_{ab\mathbf{k}} + i\Omega(\sigma_{bb\mathbf{k}} - \sigma_{aa\mathbf{k}}) + i\sigma_{ab\mathbf{k}}\sum_{\mathbf{q}\neq 0}(V_{\mathbf{q}}^{aaaa}\sigma_{aa\mathbf{k}+\mathbf{q}} - V_{\mathbf{q}}^{bbbb}\sigma_{bb\mathbf{k}+\mathbf{q}}) + i(\sigma_{bb\mathbf{k}} - \sigma_{aa\mathbf{k}})\sum_{\mathbf{q}\neq 0} V_{\mathbf{q}}^{abba}\sigma_{ab\mathbf{k}+\mathbf{q}} - i(\sigma_{bb\mathbf{k}} - \sigma_{aa\mathbf{k}})V_0^{abab}\sum_{\mathbf{k}'\neq\mathbf{k}}\sigma_{ab\mathbf{k}'}\,, \tag{5}$$

where the dephasing rate $\gamma_{ab}$ includes contributions from all relaxation mechanisms, $\Delta = -\Delta_{a0}$ is the detuning of the field from the resonance with the zero-momentum single-particle states, $\delta_{\mathbf{k}} = \hbar\mathbf{k}^2/2m_r$, and the reduced mass is $m_r^{-1} = m_a^{-1} - m_b^{-1}$. We designate the mass of a free electron by $m_0$. The equation for $\dot{\sigma}_{bb\mathbf{k}}$ is similar to that for $\dot{\sigma}_{aa\mathbf{k}}$. Terms with coefficients $V_{\mathbf{q}}^{aaaa}$ and $V_{\mathbf{q}}^{bbbb}$ correspond to the exchange self-energy, $V_{\mathbf{q}}^{abba}$ to the excitonic enhancement, and $V_0^{abab}$ to the depolarization.

Gain or absorption is determined by the sum of the polarizations at all momenta $P_{\mu\nu} = \sum_{\mathbf{k}}\sigma_{\mu\nu\mathbf{k}}$ as follows: $G = -\frac{2\omega|\wp_{ab}|^2}{\hbar\varepsilon_0 cn}\,\text{Im}(\frac{P_{ab}}{\Omega})$, where $\omega$ is the frequency of the field, and $n$ is the index of refraction. Therefore we attempt to write the set of equations containing only total polarizations and populations in a subband. The direct summation of Eqs. (4) and (5) in a manner similar to [12] yields (a similar result is obtained for $P_{bb}$)

$$\dot{P}_{aa} = i\Omega P_{ba} - i\Omega^{*}P_{ab} - w_{ab}P_{aa}\,, \tag{6}$$

$$\dot{P}_{ab} = -(\gamma_{ab} - i\Delta)P_{ab} - iV_0^{abab}(P_{bb} - P_{aa})P_{ab} + i\Omega(P_{bb} - P_{aa}) - iS_{ab}\,, \tag{7}$$

where we defined the inhomogeneous part of the polarization to be $S_{ab} = \sum_{\mathbf{k}}\delta_{\mathbf{k}}\sigma_{ab\mathbf{k}}$. We see that the Coulomb terms completely disappear from the equations for the populations, and are present only in the equation for the polarization as the depolarization shift, and, implicitly, via $S_{ab}$. The result given by Eq. (7) proves an analog of the generalized Kohn's theorem [13] for this case. It states that under the conditions of truly two-dimensional electrons (where the depolarization shift vanishes) and

of parallel subbands ($\delta_{\mathbf{k}} = 0$), the spectrum of gain or absorption does not depend on the Hartree-Fock part of the Coulomb interaction between electrons. Equivalently, the line shape is determined by noninteracting single-particle excitations.

We numerically solve Eqs. (4) and (5) for a small laser field (linear absorption). The case of a small difference of masses in the two subbands corresponds to, e.g., a GaAs/AlGaAs quantum well and a 100 meV transition. Masses of the bands estimated from Kane's model are $m_a = 0.078m_0$, $m_b = 0.069m_0$; $\varepsilon_b = 10.9$, $\gamma_{\text{deph}} = 1$ meV, $\gamma_{\text{coll}} = 1$ meV. The environment temperature is taken to be 12 K, and the electron density $N = 1.25 \times 10^{12}$ cm$^{-2}$. The specific well structure is not essential for the results. In agreement with the above analytical result, the absorption (see Fig. 2) looks like a Lorentzian and is only slightly different due to subband dispersion when calculated with (dashed line) or without (solid line) the exchange terms (which are of the order of several meV in this case). The depolarization terms cause just a shift of the absorption line to higher frequencies (dotted line in Fig. 2) in agreement with (7).

The case of a large difference of masses in the two subbands is realized in a InAs/AlSb quantum well and a 100 meV transition. The parameters different from the previous case are $m_a = 0.039m_0$, $m_b = 0.027m_0$, $\varepsilon_b = 15.7$, $N = 10^{12}$ cm$^{-2}$. In this case (see Fig. 3) the absorption spectrum without the account of the Coulomb interaction essentially replicates the population distribution in the filled lower subband, since the energy density of states is constant. Unlike the previous case, the absorption with the account of the Coulomb interactions is very much different and is sharply peaked with the linewidth determined essentially by relaxation rates. The interpretation of the line shape can be provided via a pair of electrons excited to the upper subband and a hole created thereby in the lower subband. In typical intersubband transitions the mass of the upper subband is greater than that of the lower subband, and therefore the reduced mass of the electron-hole pair is negative. This makes the interaction between the electron and the hole repulsive. The electron and the hole, appearing in the final state of absorption, try to decrease the overlap of their wave functions, which decreases the dipole moment (oscillator strength) of the transition. To form an eigenstate under the interaction, the electron and the hole have to spread over some range of neighboring momentum states. This would be impossible when the neighboring states are occupied and exercise Pauli exclusion. Thus holes around the electron Fermi momentum of the lower subband prevent the decrease of the oscillator strength, while it is decreased at all other momenta.

In case of a small laser field (linear absorption) the equations (5) become a linear set of equations for polarizations at different momenta. The solution for the sum of polarizations can be decomposed into a sum of simple fractions corresponding to collective modes and containing $\Delta + i\gamma_{ab} - \lambda_j$ in the denominator, where $\lambda_j$ is a complex eigenvalue of the matrix. The sum of their numerators is equal to $\sum_{\mathbf{k}}(\sigma_{bb\mathbf{k}} - \sigma_{aa\mathbf{k}})$. The integral of such a Lorentzian is independent of its width and its resonant frequency, and proportional to the numerator. Therefore the integral of the linear absorption coefficient over detuning obeys a specific sum rule

$$\int \mathrm{Im}\left(\frac{P_{ab}(\Delta)}{i\Omega}\right) d\Delta = \pi(P_{bb} - P_{aa}). \qquad (8)$$

The integral in (8) is independent of the interactions between particles [14], and gives a special case of a more general "oscillator strength sum rule," Eq. (5.7.5) in Ref. [15], for frequencies close to the resonance.

Because of this sum rule, the oscillator strength is redistributed by the excitonic enhancement $V^{abba}$ in favor of the collective mode close to the Fermi momentum, and its resonance is shifted to a higher frequency by the exchange self-energy $V^{bbbb}$ (dashed line in Fig. 3). A surprising result is that the electron-hole interaction, despite being repulsive, causes the peak absorption to be greater than the free-electron absorption (see Fig. 3). Such a resonance,

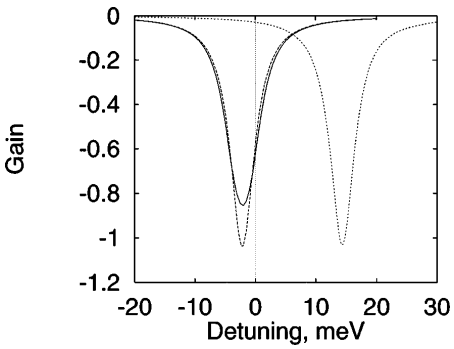


FIG. 2. Gain spectrum for the lower filled subband with a small subband dispersion (corresponding to, e.g., GaAs QW), $m_r = -0.63m_0$, as a function of detuning $\Delta$ without Coulomb interaction (solid line), with only exchange terms (dashed line), and with all terms (dotted line) at $k_F a_W = 0.4$.

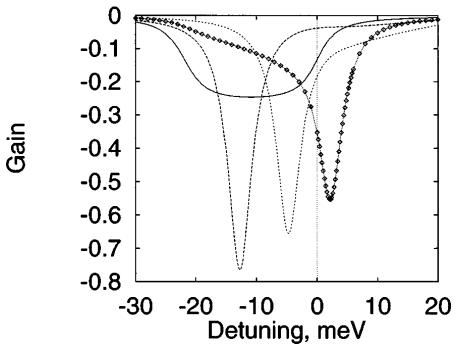


FIG. 3. Gain spectrum for the lower filled subband with a large subband dispersion (corresponding to, e.g., InAs QW), $m_r = -0.093m_0$, as a function of detuning $\Delta$ without Coulomb interaction (solid line), with only exchange terms (dashed line), with only the depolarization (diamond line), and with all terms (dotted line) $k_F a_W = 0.4$.

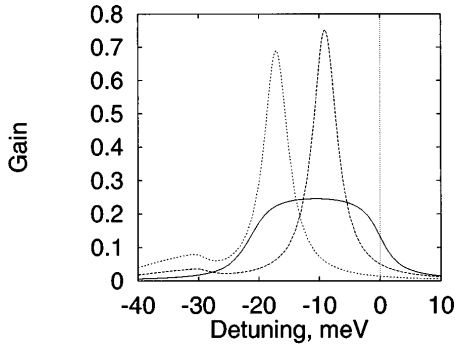


FIG. 4. Gain spectrum for the upper filled subband with a large subband dispersion (corresponding to, e.g., InAs QW), $m_r = +0.093m_0$, as a function of detuning $\Delta$ without Coulomb interaction (solid line), with only exchange terms (dashed line), and with all terms (dotted line) at $k_F a_W = 0.4$.

which we term "repellon," is reminiscent of the Mahan exciton [16]. They both broaden and disappear at higher temperatures. However, our calculations show that the line shape only slightly changes from 0 to 12 K (shown in Fig. 3), i.e., up to the temperatures which are easily accessible experimentally. We emphasize that, contrary to the Mahan exciton, the repellon appears due to repulsive electron-hole interaction.

At large subband dispersion, the depolarization is not reduced to a simple shift. Consider the effect of the depolarization alone, as was done in [6,8]. We find from (5) that this term causes an effectively attractive interaction between an electron and a hole, resulting in a bound state with a positive (above the band edge) energy. Thus depolarization forms an absorption peak associated with the bound state near the higher-frequency ($k \approx 0$) edge of the spectrum (see the diamond line in Fig. 3). It counteracts the excitonic enhancement terms which are repulsive and tend to form a peak near the lower-frequency ($k \approx k_F$) edge of the spectrum. At densities considered, combined action of the exchange and depolarization terms causes the peak to be at some intermediate frequency and to be wider (dotted line in Fig. 3). The depolarization bound state would in fact determine the spectrum at much higher densities, or, alternatively, for much wider wells.

To compare the repellon to usual collective excitations, consider gain in case of a filled upper band and an empty lower band. Then an attractive pair of an electron in the lower band and a hole in the upper band [17], called "antiexciton" with a positive reduced mass will determine the emission line shape. It causes the gain with the account of the Coulomb interactions to have a usual excitonic spectrum (see Fig. 4). In spite of the similarity of the line shapes of the antiexciton and the repellon, the minimum of gain between the antiexciton peak and the tail shows the presence of a bound state separated from the continuum [18].

In conclusion, we show that absorption or gain in intersubband transitions is determined by the interplay between the Coulomb interaction and the subband dispersion. For a small dispersion, the exchange effects do not modify the spectrum while the depolarization terms lead to its blueshift. For a large dispersion, the spectrum is dominated by collective excitations. The excitonic enhancement terms lead to a formation of the resonance (repellon) due to a repelling electron-hole pair near the Fermi edge. The exchange self-energy blueshifts the repellon resonance, and the attractive depolarization terms counteract the formation of the repellon.

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*Electronic address: dmitri@quantum.ece.ucsb.edu

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