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Multiple peak structure of intersubband absorption in heterostructures with closely spaced levels

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The shape of far-infrared absorption spectra due to resonant transitions of electrons between two-dimensional subbands is studied. A quantum kinetic theory is applied to describe both the Coulomb interaction in the Hartree-Fock approximation and elastic scattering of electrons. A multipeak structure of the absorption spectra is found as a result of the nonlocal nature of the exchange part of the Coulomb interaction. [S0163-1829(99)03335-4]

Although the resonant intersubband transitions excited by infrared radiation in heterostructures with two-dimensional (2D) electron gas are the subject of intensive studies over the last decades,[1,2] there is a lack of a detailed consideration of the shape of absorption peak. In the case of GaAs-based structures, where the electronic spectra in the ground-state (1) and excited (2) subbands are satisfactorily described by the parabolic laws with equal effective masses, it is usually assumed that the peak has either Lorentzian or Gaussian shape. The Gaussian shape is relevant when the broadening is dominated by the large-scale inhomogeneities,[3] while the Lorentzian shape may be realized when the intersubband level splitting energy $\hbar\omega_{21}$ is greater than the optical phonon energy $\hbar\omega_0$, and the peak broadening is determined by a substantial broadening of the excited subband due to a spontaneous emission of the optical phonons by the electrons.[4]

The problem appears to be more complicated for heterostructures with closely spaced levels (Fig. 1), when $\omega_{21} < \omega_0$ and the intersubband transitions are excited in far-infrared spectral region. In this case, the peak shape should be governed mostly by Coulomb interaction effects[5] and elastic scattering of electrons by imperfections of the heterostructure. Recently, the intersubband transitions in the far-infrared region have been directly investigated in double quantum wells as well as in wide-square quantum wells under excitation by far-infrared gas laser or free-electron laser.[6]

The influence of Coulomb interaction on intersubband transitions in 2D systems has been studied in several papers uncovering its different manifestations. The depolarization shift of the spectrum due to dynamical resonant screening of the infrared field by electrons has been considered first by Allen *et al.* in the mean-field (Hartree) approximation.[7] Then, Ando[8] studied the exchange contribution to the depolarization effect and found that it leads to a reduction of the depolarization shift, while the shape of the absorption line remains unchanged. This consideration followed Lundquist's scheme[9] based upon introduction of local exchange-correlation potential and such an approach, as well as its improved form,[10] does not take into account the nonlocal properties of the exchange interaction. Next, Bandara *et al.* have studied the exchange-induced nonparabolicity[11] of electron spectra. This effect results in energy-dependent renormalization of the level splitting and, in principle, could produce the broadening of the absorption line up to several meV. However, it was shown[12] that the depolarization effect strongly narrows the absorption line broadened due to energy dependence of the level splitting. In subsequent papers,[13–15] a full Hartree-Fock scheme taking into account both depolarization terms and exchange renormalization of the splitting energy has been employed. Nevertheless, the shape of infrared absorption spectra calculated for narrow quantum wells did not show a considerable difference in comparison to the case when only depolarization effect is accounted. The reasons for this have been revealed in Ref. 15, whose authors show that in frames of the two-dimensional approximation for Coulomb interaction, both exchange terms, being summed up over the electron states, exactly compensate each other and vanish. It means that the absorption, as an integral value, does not depend on the exchange interaction if the level splitting energy and dephasing times are energy independent.

The heterostructures with closely spaced levels give new birth to the problem of the influence of exchange interaction on the absorption spectra in far-infrared region. Due to considerably larger transverse dimensions of the 2D states therein (typically 20-30 nm), the two-dimensional approximation for Coulomb interaction is no longer satisfactory, and nonlocal nature of the exchange interaction, as we show in this paper, leads to a qualitatively new phenomenon: a *multiple peak structure* of the absorption. Below we analyze the characteristic features of the absorption in heterostructures with closely spaced levels using the resonant approximation for high-frequency contribution to the one-particle density matrix of electrons. In contrast to the method of semiconductor Bloch equations with phenomenological broadening used in Ref. 15, our method allows to consider both Coulomb interaction and scattering effects on the equal ground. In the

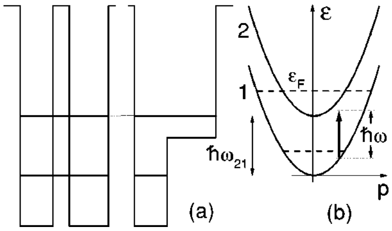

FIG. 1. Band diagrams (a) and electron spectra (b) for two examples of heterostructures with closely spaced levels: double quantum wells and step quantum wells (other examples are wide square and parabolic quantum wells). The level splitting energy $\hbar\omega_{21}$ and optical quantum energy $\hbar\omega$ are shown. The dashed lines correspond to Fermi levels in strongly and weakly doped structures.

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equations below we put $\hbar = 1$.

Consider a two-level 2D system excited by the far-infrared radiation whose electric field $E_\perp e^{-i\omega t}$ is directed perpendicular to the 2D plane. The induced transverse current $j_\perp e^{-i\omega t}$ is expressed through the nondiagonal part of the density matrix δf_{21} according to $j_\perp = e\rho_{2D} v_{12} \int_0^\infty d\varepsilon\, \delta f_{21}(\varepsilon)$, where $v_{12} = -i\omega_{21} \int dz\, \psi_{1z} z \psi_{2z}$ is the intersubband matrix element of the transverse velocity operator, ψ_{1z} and ψ_{2z} are the wave functions of electrons in the subbands, ρ_{2D} is the 2D density of states, and $\varepsilon = p^2/2m$ is the kinetic energy for in-plane motion. Due to the assumed in-plane isotropy of the problem, the density matrix depends on this energy only. The relative absorption ξ, introduced as a ratio of the absorbed power $\mathrm{Re}(E_\perp j_\perp)$ to the Poynting vector for radiation through the structure, is determined by

$$\xi = \frac{e^2}{c\sqrt{\kappa}} \frac{4m|v_{12}|^2}{\omega_{21}} \mathrm{Im} \int_0^\infty d\varepsilon\, \varphi(\varepsilon), \tag{1}$$

where κ is the dielectric permittivity, which is supposed to be constant across the structure, and normalized intersubband polarization φ is introduced according to $\delta f_{21} = -(ie/\omega_{21}) E_\perp v_{21} \varphi$. The frequency dependence of the absorption is determined in a rather complex way because $\varphi(\varepsilon)$ is governed by a kinetic equation

$$\begin{aligned} &[\omega - \omega_{21}(\varepsilon) + i\Gamma(\varepsilon)]\varphi(\varepsilon) + i\mathcal{D}(\varphi|\varepsilon) + \mathcal{K}(\varphi|\varepsilon) \\ &\quad = f_2(\varepsilon) - f_1(\varepsilon). \end{aligned} \tag{2}$$

Here, $\omega_{21}(\varepsilon)$ is the energy of the intersubband transition, which is renormalized (shifted and becomes energy dependent) due to both the elastic scattering (imaginary part of the collision integral) and Coulomb effects, $\Gamma(\varepsilon)$ is the broadening energy coming from the real part of the collision integral, $\mathcal{D}(\varphi|\varepsilon) = \gamma(\varepsilon)[\varphi(\varepsilon + \delta\omega) + \varphi(\varepsilon - \delta\omega) - 2\varphi(\varepsilon)]$, (here $\delta\omega = \omega - \omega_{21}$ and γ is a characteristic energy comparable to Γ) is a finite-difference operator also coming from the real part of the collision integral. In the vicinity of the intersubband resonance $|\omega - \omega_{21}| \ll \omega$ the finite-difference operator gives only a small contribution as compared to $\Gamma(\varepsilon)$ term and will be neglected in the following. Next, $\mathcal{K}(\varphi|\varepsilon) = \int d\varepsilon' L(\varepsilon, \varepsilon')\varphi(\varepsilon')$ is the integral operator incorporating both the Coulomb effects and elastic scattering effects (incoming contribution of the imaginary part of the collision integral). The right-hand side of Eq. (2) is expressed through the equilibrium Fermi distribution functions $f_i(\varepsilon)$ for the electrons in subbands $i = 1$ and $i = 2$. Below we consider the low-temperature conditions, when the electron gas is degenerate. The right-hand part is nonzero and equal to -1 inside an interval of energies from $p_{F2}^2/2m$ to $p_{F1}^2/2m$, or from 0 to $p_{F1}^2/2m$, if only subband 1 is populated (here p_{F1} and p_{F2} are the Fermi momenta of electrons in the subbands). Equation (2) has been derived from the general form of quantum kinetic equation.[16]

Before presenting our results for spectral dependence of ξ obtained in the numerical analysis of Eq. (2), we point out two important qualitative features of this equation. Even if we omit, for a moment, the integral operator, it is still evident that Eq. (2) describes a non-Lorentzian peak because of the energy dependence of both the energy of intersubband transition and broadening energy. Another important qualitative feature of Eq. (2) is a possibility of multipeak solutions for the absorption. This possibility is directly related to the presence of the integral operator. To demonstrate this, let us consider a simplified variant of Eq. (2)

$$[\omega - \omega_{21} + i\Gamma]\varphi(\varepsilon) + \int d\varepsilon' \mathcal{L}(\varepsilon, \varepsilon')\varphi(\varepsilon') = f_2(\varepsilon) - f_1(\varepsilon), \tag{3}$$

where the energy dependence of ω_{21} and Γ is neglected, and the integral operator is described by a model degenerate kernel $\mathcal{L}(\varepsilon, \varepsilon') = \Sigma_{k=1}^m g_k(\varepsilon) g_k(\varepsilon')$. In this case Eq. (3) is reduced to a set of $m+1$ linear equations for $s_0 = \int d\varepsilon \varphi(\varepsilon)$ and $s_k = \int d\varepsilon g_k(\varepsilon)\varphi(\varepsilon)$. The determinant for such a set is a polynomial of the order $m+1$. It means that the solution s_0 can have up to $m+1$ poles close to real axis. Therefore, the relative absorption, which is proportional to imaginary part of s_0, can have up to $m+1$ peaks provided that the broadening Γ is weak and these peaks can be resolved. In order to test the possibility of the multipeak absorption for the realistic integral operator, we solve Eq. (2) numerically as described below.

We assume that the elastic scattering is weak enough and the main contributions to $\omega_{21}(\varepsilon)$ and $\mathcal{K}(\varphi|\varepsilon)$ appear from Coulomb interaction terms. Then, for $\omega_{21}(\varepsilon)$ we obtain

$$\omega_{21}(\varepsilon) = \omega_{21} - \omega_H + \omega_F(\varepsilon), \tag{4}$$

where ω_H is the Hartree shift of the splitting energy. This term, due to its independence on the kinetic energy, can be initially included to definition of ω_{21} and, therefore, dropped out from the equations. The next term describes the exchange renormalization of the splitting energy

$$\begin{aligned} \omega_F(\varepsilon) = \frac{e^2}{\kappa} \Bigg\{ &\int_0^{p_{F1}-p} dq [M_{1111}(q) - M_{1212}(q)] \\ &+ \pi^{-1} \int_{p_{F1}-p}^{p_{F1}+p} dq [M_{1111}(q) \\ &- M_{1212}(q)] \cos^{-1}\left(\frac{p^2 + q^2 - p_{F1}^2}{2pq} \right) \\ &- \pi^{-1} \int_{p-p_{F2}}^{p+p_{F2}} dq [M_{2222}(q) \\ &- M_{2121}(q)] \cos^{-1}\left(\frac{p^2 + q^2 - p_{F2}^2}{2pq} \right) \Bigg\}, \end{aligned} \tag{5}$$

where $p \equiv \sqrt{2m\varepsilon}$ and $M_{abcd}(q) = \int dz \int dz' e^{-q|z-z'|} \psi_{az} \psi_{bz'} \psi_{cz'} \psi_{dz}$. The last term in Eq. (5) must be skipped if only the ground-state subband is populated.

The integral operator is given as

$$\begin{aligned} \mathcal{K}(\varphi|\varepsilon) = [f_2(\varepsilon) - f_1(\varepsilon)] &\int d\varepsilon' \\ &\times \left\{ L_H - \frac{1}{a_B} \int_0^\pi \frac{d\phi}{\pi} \frac{M_{1221}[p_{\varepsilon\varepsilon'}(\phi)]}{p_{\varepsilon\varepsilon'}(\phi)} \right\} \varphi(\varepsilon'), \end{aligned} \tag{6}$$

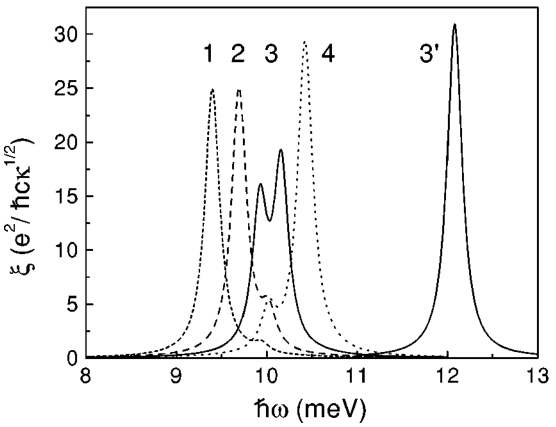

FIG. 2. Frequency dependence of the intersubband absorption for a double quantum well structure in tunneling resonance conditions. The calculation is done at $\hbar\omega_{21}=6.5\,\mathrm{meV}$, $\hbar/\tau=0.4\,\mathrm{meV}$ ($\Gamma=0.1\,\mathrm{meV}$) and $\varepsilon_F=5$, 5.5, 6, and 6.5 meV, curves 1–4, respectively, the Fermi energy ε_F is counted from the bottom of the ground-state subband. The curve 3′ shows the shape of absorption line for $\varepsilon_F=6$ meV if the exchange interaction is neglected.

where a_B is the Bohr radius, and $p_{\varepsilon\varepsilon'}(\phi)=[2m(\varepsilon+\varepsilon'-2\sqrt{\varepsilon\varepsilon'}\cos\phi)]^{1/2}$. The energy-independent part $L_H=-(2/a_B)\int dz\int dz'|z-z'|\psi_{2z}\psi_{1z'}\psi_{2z'}\psi_{1z}$ of the kernel describes Hartree depolarization.[7] The second, energy-dependent part is the exchange contribution, which is responsible for the nonlocality of Eq. (2) leading to the multiple-peak structure of the optical transitions as demonstrated below.

The calculations are done for a symmetric (equal well widths) double quantum well heterostructure. The distance between the centers of the wells is chosen to be equal to the Bohr radius, and material parameters of GaAs are used. We also apply the approximation of short-range scattering potential, which renders Γ independent on the energy at $\varepsilon<\omega$ and equal to $(\hbar/\tau)[1-(3/4)(\Delta_{SAS}/\hbar\omega_{21})^2]$, where τ is the scattering time, and Δ_{SAS} is the minimum level-splitting energy. A numerical solution of Eq. (2) is obtained in a direct way using the Gaussian method.

Figure 2 shows frequency dependence of ξ, normalized to $e^2/(\hbar c\sqrt{\kappa})$, at several values of the Fermi energy corresponding to sheet electron densities 1.4, 1.55, 1.7, and 1.83 $\times 10^{11}\,\mathrm{cm}^{-2}$. The calculation is done for tunneling resonance conditions $\hbar\omega_{21}=\Delta_{SAS}$, at $\hbar/\tau=0.4\,\mathrm{meV}$.[17] Due to the Hartree depolarization effect, the absorption lines are shifted to higher energies. The exchange effect decreases this shift (compare curves 3 and 3′) and causes the splitting of the lines in two peaks. We can see either a weak satellite on the left or right shoulder of the main peak, or two closely spaced peaks with nearly equal heights. The favorable conditions for the peak doubling are realized when the Fermi energy is slightly below the bottom of the excited subband. Varying the parameters in reasonable limits, we did not find more than two peaks. However, when we introduced a pronounced energy dependence of the level splitting in Eq. (2), we have seen three peaks. This is not surprising, since such an energy dependence increases the importance of the exchange terms.[15]

From the experimental point of view, it is convenient to vary the frequency of transitions rather than the frequency of infrared radiation. For this reason, we did a calculation of ξ versus the splitting energy parameter Δ, which is proportional to a voltage V applied to the gates enclosing the double quantum well. The intersubband splitting energy is expressed through Δ as $\hbar\omega_{21}=\sqrt{\Delta^2+\Delta_{SAS}^2}$. The results are presented in Fig. 3. Regarding the importance and high sensitivity of differential spectroscopic measurements, we plot also derivatives $d\xi/d\Delta$. The dependence $\xi(\Delta)$ has a two-peak shape for small enough Δ. With an increase of Δ, the broadening energy Γ also increases, and the peaks become unresolved. However, the differential spectrum for this case clearly indicates the presence of two-peak structure of absorption.

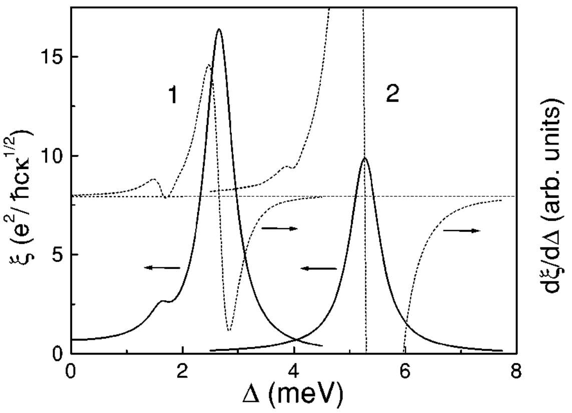

FIG. 3. Level-splitting dependences of the absorption and their first derivatives (scaled and shifted up for clarity) for a double quantum well with $\hbar/\tau=0.4\,\mathrm{meV}$, $\Delta_{SAS}=6\,\mathrm{meV}$, $n_1=1.6\times 10^{11}\,\mathrm{cm}^{-2}$ ($n_2=0$), and $\hbar\omega=10\,\mathrm{meV}$ (curve 1) and $n_1=1.8\times 10^{11}\,\mathrm{cm}^{-2}$ and $\hbar\omega=12\,\mathrm{meV}$ (curve 2).

The Hartree-Fock approach does not account for Coulomb correlation effects of the higher order and, therefore, is open for improvements. A development of a consistent theory is, however, a very complex problem. Some efforts have been made to extend the nonlocal theory beyond the Hartree-Fock approach by an introduction of the screened exchange potentials.[18,19] An application of the results to a calculation of the intersubband transition energy in double quantum wells gives a good agreement with the experiment (see Plaut *et al.* in Ref. 6). However, the theories[18,19] are not rigorous because they treat the screening itself in the random phase approximation, i.e., without the exchange contributions. On the other hand, any nonlocal theory of Coulomb interaction would lead us to the integral equations for intersubband polarization, and the phenomena described in this letter could exist.

In conclusion, we have found a qualitative modification of the resonant intersubband transition spectra: the multipeak absorption due to exchange contribution to the dynamical screening (depolarization) of the far-infrared radiation field by electrons. The doubling of the peak can be physically interpreted as a result of an additional particlelike contribution into intersubband polarization. Such a picture is analogous to description of the exciton contribution into interband polarization because the equations for interband (see Ref. 20) and intersubband [see Eqs. (2) or (11) from Ref. 13] polarizations have the same form. From the mathematical point of view, the appearance of the multipeak structure of absorption spectrum is connected with the integral operator standing in the kinetic equation for the nondiagonal part of the density matrix. Therefore, possible physical mechanisms leading to this effect are not necessarily limited to Coulomb interaction: any mechanism, which gives the integral operator in the kinetic equation can be responsible for the multipeak absorption. This simple mathematical fact is the most unusual point of our study. We stress that the heterostructures with closely spaced levels demonstrate strong peculiarities of the absorption, and hope that the presented results will stimulate spectroscopical investigations in far-infrared spectral region.

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