

Effect of macroscopic irregularities of the heteroboundaries on the optical susceptibilities of quantum structures

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The effect of technological fluctuations of the positions of the heteroboundaries on the optical susceptibilities of quantum structures was investigated. It was shown that these fluctuations cause a temperature-dependent shift of the resonance frequencies in the absorption spectrum on intersubband transitions. It was demonstrated for the example of a solitary quantum well that in a system which is symmetric on the average there arises a second-order optical response which is proportional to the difference of the rms deviations of the position of the opposite boundaries of the structure. © 1995 American Institute of Physics.

One of the main methods for investigating quantum nanostructures is measurement of the optical characteristics of artificially layered compounds grown by means of the molecular-beam-epitaxy technology. Moreover, the possibility of producing a structure with a prescribed symmetry of the wave functions and arrangement of the energy levels also makes it possible to construct systems with the required optical properties, which is extremely important for applications. This is why so much attention is being devoted to the optics of heterostructures and why so many theoretical and experimental studies have been performed in this field. In most studies, however, it is assumed that the heteroboundaries are ideal, and that the widths of the quantum wells and barriers remain constant in a direction perpendicular to the direction of growth. Nonetheless, even the most perfect samples contain random growth defects in the form of steps which cause the widths of the layers to vary, as a minimum, by one or two atomic layers.¹

In the present paper we shall study the average effect of fluctuations of the position of the heteroboundaries on the optical properties of quantum structures. We shall restrict the analysis to the linear response (the absorption coefficient) and the second-order nonlinear response (for definiteness, the optical rectification coefficient) at frequencies corresponding to interband transitions. This range corresponds to the infrared region of the spectrum.

1. First, we calculate the linear absorption coefficient. We shall show below that the technological fluctuations in the position of the interfaces lead not only to an obvious broadening of the spectral lines, but also to a temperature-dependent shift of the resonance frequencies in the absorption spectrum. As the simplest example, we shall study an isolated, two-level quantum well with average thickness L and barrier height V (Fig. 1a). The cross section of this structure in the (x, z) plane, where x is the direction of growth, is shown schematically in Fig. 1b. The physical nature of the above-mentioned shift of

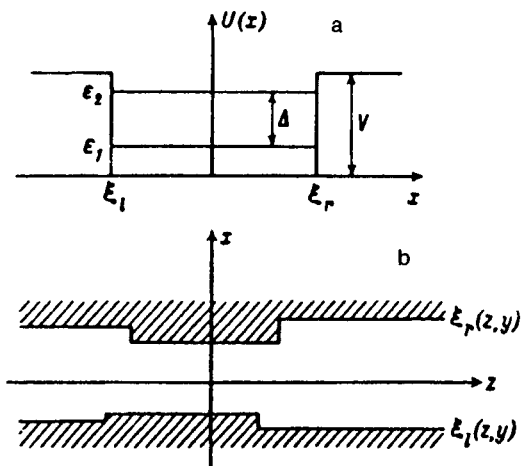


FIG. 1. a — Local potential well of a two-level quantum well. b — Schematic diagram of a section of the structure by the (x,z) plane (the barrier regions are hatched).

the resonance frequency is attributable to the nonuniform distribution of charge carriers along a quantum well with variable thickness. At low temperatures the particle concentration in the wide regions is higher than the average concentration along the well. Upon averaging, these regions therefore make a large contribution to the absorption coefficient. At the same time, the splitting Δ between the quantum-size subbands in the wide regions is less than the average splitting. Therefore, the resonance in the average absorption coefficient will be shifted downward in frequency relative to the position of the corresponding average width of the quantum well. As the temperature is raised, the particle distribution along the structure becomes more uniform as a result of the temperature broadening of the distribution function. The shift of the resonance frequency will therefore decrease. In carefully grown structures the characteristic size of the uniform regions (regions of constant width) is quite large (from hundreds of angstroms up to several microns).¹ Nonuniformities of such a scale can be regarded as macroscopic and the quasiclassical approximation can be used to calculate the optical susceptibilities. Specifically, the calculation of the absorption coefficient $\alpha(\omega)$ reduces to averaging the quantity $\alpha_{loc}(\omega)$, which depends on the local values of the coordinates of the right-hand ξ_r and left-hand ξ_l boundaries (see Fig. 1a). Averaging should be performed with the distribution functions $P(\delta\xi_{r(l)})$ of the deviations $\delta\xi_r$ and $\delta\xi_l$ of the right- and left-hand boundaries, respectively, from their average values $\bar{\xi}_r$ and $\bar{\xi}_l$. We assume that the distribution of the fluctuations is Gaussian:

$$P(\delta\xi_j) = \frac{1}{\sqrt{2\pi\sigma_j^2}} \exp\left\{-\frac{\delta\xi_j^2}{2\sigma_j^2}\right\}; \quad \sigma_j^2 = \langle \delta\xi_j^2 \rangle, \quad (1)$$

where j corresponds to the index of the right-hand ($j=r$) or left-hand ($j=l$) boundaries. The local width of the quantum well is determined in the obvious manner

$$L = \bar{L} + \delta L, \quad \bar{L} = \bar{\xi}_r - \bar{\xi}_l; \quad \delta L = \delta \xi_r - \delta \xi_l. \quad (2)$$

The local absorption coefficient $\alpha_{\text{loc}}(\omega)$ is proportional to the imaginary part of the polarizability $\text{Im}\chi_{\text{loc}}^{(1)}(\omega)$, which depends on the local width L (2) of the quantum well. Near a resonance corresponding to a transition from the ground state 1 into the excited state 2

$$\alpha_{\text{loc}}(\omega) \propto \text{Im} \chi_{\text{loc}}^{(1)}(\omega) = e^2 |x_{12}|^2 n_{\text{loc}} \frac{\gamma_{\text{ph}}}{(\omega - \Delta)^2 - \gamma_{\text{ph}}^2}, \quad (3)$$

where x_{12} is the matrix element of the coordinate operator with respect to the wave functions of the ground state and the excited state, n_{loc} is the local particle concentration in the bottom subband in the given region of the quantum well, γ_{ph} is the natural (phonon) width of the resonance, and $\Delta = \varepsilon_2 - \varepsilon_1$ is the local splitting between the quantum-size subbands. In Eq. (3) the frequency ω is expressed in energy units and it is assumed that $T \ll \Delta$. We obtain the absorption coefficient of the structure as a whole by averaging expression (3) with the distribution function (1). For $\delta L/L \ll 1$, the dependence of the quantities n_{loc} and Δ on δL makes the main contribution to the deviation of $\alpha_{\text{loc}}(\omega)$ from $\alpha(\omega)$. For the structure at hand, the increase δL leads to a proportional decrease in the splitting Δ between the subbands compared with the average value $\bar{\Delta}$ because of the shift in the energy levels ε_1 and ε_2 :

$$\Delta = \bar{\Delta} - \delta \Delta, \quad \delta \Delta = \delta \varepsilon_2 - \delta \varepsilon_1 = \alpha_{21} \delta L. \quad (4)$$

Here we have introduced the notation

$$\alpha_{21} = \alpha_2 - \alpha_1, \quad \alpha_i = \left| \frac{\partial \varepsilon_i}{\partial L} \right|. \quad (5)$$

An expression for the coefficient α_i in the case of a square quantum well can be easily obtained directly from the solution of the Schrödinger equation

$$\alpha_i = \frac{2\varepsilon_i}{L} \left[1 - \frac{2\hbar}{L\sqrt{2m(V - \varepsilon_i)}} \right]^{-1}.$$

The quantity n_{loc} for a nondegenerate electron gas is an exponential function of δL :

$$n_{\text{loc}} = \frac{mT}{2\pi} e^{\mu/T} \exp \frac{\alpha_1 \delta L}{T}. \quad (6)$$

The chemical potential μ is determined from the conservation of the total number of particles in the structure:

$$n = \langle n_{\text{loc}} \rangle = \frac{mT}{2\pi} e^{\mu/T} \exp \frac{\alpha_1^2 \sigma_L^2}{2T^2}; \quad \sigma_L^2 = \langle \delta L^2 \rangle = \sigma_r^2 + \sigma_l^2. \quad (7)$$

The brackets in Eq. (7) indicate averaging over $\delta \xi_r$ and $\delta \xi_l$ with the distribution function (1). It follows from Eq. (7) that fluctuations in the width of the quantum well lead to a shift of the chemical potential by the amount $\delta \mu = -\alpha_1^2 \sigma_L^2 / 2T^2$. To simplify the calculations, we assume that the line width is determined mainly by fluctuations of the thickness of the quantum well, i.e., we assume that

$$\sigma_{\Delta} = \sqrt{\langle \delta \Delta^2 \rangle} \equiv \alpha_{21} \sigma_L \gg \gamma_{ph}. \quad (8)$$

This condition holds well for sufficiently narrow quantum wells, where a small change in the width has a large effect on the shift of the energy levels. The determination of the absorption coefficient reduces to calculating the following average:

$$\alpha(\omega) \propto \frac{1}{\pi} e^2 |x_{12}|^2 \langle n_{loc} \delta(\omega - \Delta) \rangle. \quad (9)$$

Substituting into Eq. (18) n_{loc} [Eq. (6)] and Δ [Eq. (10)] and performing the elementary integration with the function (1), we obtain at the final result

$$\alpha(\omega) \propto \frac{e^2}{\pi} |x_{12}|^2 \frac{n}{\sqrt{2\pi\sigma_{\Delta}^2}} \exp\left\{-\frac{1}{2\sigma_{\Delta}^2} \left(\omega - \bar{\Delta} + \frac{\alpha_1}{\alpha_{21}} \frac{\sigma_{\Delta}^2}{T}\right)^2\right\}. \quad (10)$$

The rms deviation σ_{Δ} of the splitting between the subbands is defined in Eq. (8). Equation (10) clearly demonstrates the shift of the resonance frequency with the fluctuations in the width of the quantum well.

2. As an example of the nonlinear optical susceptibility we shall study optical rectification — the appearance of a static polarization P under the action of an alternating electric field $E = E_0 \cos(\omega t)$. The induced polarization is proportional to the squared amplitude of the field: $P = \chi_0^{(2)}(\omega) E_0^2$. The local susceptibility $\chi_{0,loc}^{(2)}(\omega)$ for a two-level system is determined by the expression (see, for example, Ref. 2)

$$\chi_{0,loc}^{(2)}(\omega) = \frac{2e^3 \tau}{\pi \hbar} M(\mathbf{p}) f_{loc}(\mathbf{p}) \delta(\omega - \Delta), \quad M(\mathbf{p}) = |x_{12}|^2 (x_{11} - x_{22}), \quad (11)$$

where τ is the damping (phonon) time of the diagonal element of the density matrix, and $f_{loc}(\mathbf{p})$ is the particle distribution function in the lower subband. We note that Eq. (11) is valid if condition (8) is satisfied. The structure of the matrix elements in Eq. (11) indicates the nature of the effect: The static polarization arises as the center of gravity of an electron shifts as a result of the transition from state 1 to state 2. It is well known that the second-order optical susceptibility is different from zero only in systems in which the spatial parity is violated. For example, in Ref. 2 it was demonstrated that an anomalously large (compared with the bulk material) second-order response appears in a stepped quantum well or in an asymmetric system of two coupled quantum wells. We shall show below that if the rms deviations of the left- and right-hand boundaries are different, then fluctuations in the position of the interfaces lead to the appearance of a nonzero optical rectification coefficient even in a structure that is symmetric on the average. The latter condition is obvious for real structures, since different heteroboundaries are grown under different technological conditions. The assertion does not contain any inconsistencies, since the locally spatial parity, despite the symmetry of the structure on the average, is still destroyed, which accounts for the second-order nonzero susceptibility.

In the zeroth-order quasiclassical approximation, which was employed above, the expression for $\chi_{0,loc}^{(2)}(\omega)$ [Eq. (11)] vanishes identically. This situation is in some sense analogous to the case of the absorption of light by a disordered semiconductor with parallel curved bands.³ The next order corresponds to a calculation of the local susceptibility, with allowance for the two neighboring regions of widening and narrowing of a

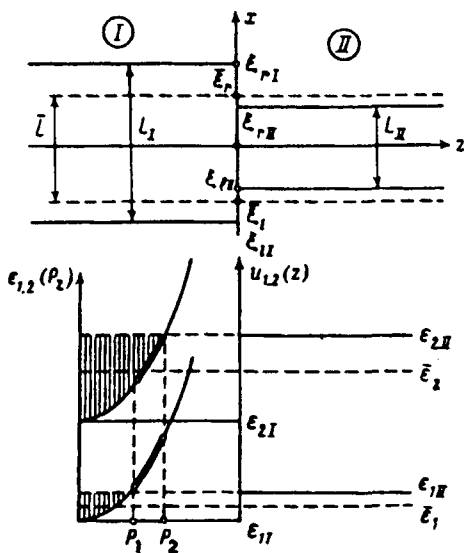


FIG. 2. Section by the (x, z) plane, the effective potential $u_{1,2}(z)$ in the z direction, and the p_z -dependence of the energy in the ground (1) and excited (2) subbands $e_{1,2}(p_z)$ for a system of two quantum wells of different width in contact with one another. The vertical dashed lines mark the states which are localized in the wide region. The thick line separates the region of states where transitions result in a shift of the center of gravity of the electron.

quantum well, followed by averaging of the result over the positions of the boundaries of the regions which are in contact with one another. We denote these regions as I and II and all quantities referring to them by the corresponding subscripts. For definiteness, let the quantum well be wider in region I than in region II, as shown in Fig. 2. In the case $\delta L/L \ll 1$ the eigenfunctions $\psi_i(\mathbf{r})$ ($i = 1, 2$) of such a system can be written in the form

$$\psi_i(\mathbf{r}) = e^{ip_y y} \chi_{i,p_z}(z) \{ \varphi_{i,I}(x) \theta(-z) + \varphi_{i,II}(x) \theta(z) \}, \quad (12)$$

where $\varphi_{i,I(II)}(x)$ are the eigenfunctions of a one-dimensional quantum well with the local width $L = L_I(L_{II})$, and $\chi_{i,p_z}(z)$ are the solutions of the one-dimensional Schrödinger equation for each subband in the z direction with a step potential $u_i(z)$ illustrated in Fig. 2. The value of the potential $u_i(z)$ for $z > 0$ corresponds to the energy level $\epsilon_{i,II}$ in a quantum well of width L_{II} and for $z < 0$ it corresponds to the level $\epsilon_{i,I}$ in a quantum well of width L_I (see Fig. 2). The wave functions of the i th subband (12) are localized in the wide region I if

$$p_z < p_i = \sqrt{2m(\epsilon_{i,II} - \epsilon_{i,I})} \equiv \sqrt{2m\alpha_i(\delta L_I - \delta L_{II})} \quad (13)$$

and delocalized into two regions if $p_z > p_i$. In Fig. 2 the regions of the states which are localized in the wide part are indicated by dashed lines. For delocalized states ($p_z > p_i$) the center of gravity of an electron (the diagonal matrix element of the coordinate operator) coincides with the arithmetic-mean center of regions I and II. Specifically,

$$x_{ii} = \frac{1}{2}(x_I + x_{II}), \quad p_z > p_i, \quad (14)$$

where $x_{I(II)} = (\xi_{r,I(II)} + \xi_{l,I(II)})/2$ is the center of the quantum wells I or II. For localized states ($p_z < p_i$) x_{ii} is the center of the quantum well I:

$$x_{ii} = x_I, \quad p_z < p_i. \quad (15)$$

Since $p_2 > p_1$, the quantity $x_{11} - x_{22}$ appearing in the expression for the local susceptibility (11) is different from zero only for transitions from the delocalized states of subband 1 into localized states of subband 2 (the region $p_2 > p_z > p_1$) and is determined by the expression [taking into account Eqs. (27) and (15)]

$$x_{11} - x_{22} = \frac{1}{2}(x_{II} - x_I). \quad (16)$$

In the same region, in the leading order in $\delta L/L$, the dipole matrix element is $x_{12} \approx x_{12}^{(0)}/2$ where $x_{12}^{(0)}$ was calculated using the wave functions of an ideal structure. Therefore, the combination of matrix elements M appearing in Eq. (11) is determined by the expression

$$M(\mathbf{p}) = \begin{cases} \frac{1}{8} |x_{12}^{(0)}|^2 (x_{II} - x_I), & p_2 > p_z > p_1, \\ 0, & p_z \notin [p_1, p_2]. \end{cases} \quad (17)$$

The total susceptibility $\chi_0^{(2)}(\omega)$ is obtained by calculating the average

$$\begin{aligned} \chi_0^{(2)}(\omega) &= \frac{2e^3}{8\pi} \tau |x_{12}^{(0)}|^2 e^{\mu/T} Q, \\ Q &= \left\langle (x_{II} - x_I) \int_{-\infty}^{\infty} \frac{dp_y}{2\pi} \int_{p_1}^{p_2} \frac{dp_z}{2\pi} \exp\left\{-\frac{\mathbf{p}}{2mT} + \frac{\alpha_1 \delta L_I}{T}\right\} \right. \\ &\quad \left. \times \delta(\omega - \bar{\Delta} + \alpha_{21} \delta L_I) \theta(\delta L_I - \delta L_{II}) \right\rangle. \end{aligned} \quad (18)$$

The averaging in expression (18) is performed with the distribution function (1) over $\delta\xi_{r,I}$, $\delta\xi_{r,II}$, $\delta\xi_{l,I}$, and $\delta\xi_{l,II}$. It is convenient to calculate the integral in Eq. (18) after switching from integration over fluctuations of the right- and left-hand boundaries $\delta\xi_{r,I(II)}$ and $\delta\xi_{l,I(II)}$ to integrations over fluctuations of the width $\delta L_{I(II)}$ of the well and fluctuations of the position of the center $x_{I(II)}$ of the well. As a result, we obtain the following expression for the average susceptibility describing optical rectification:

$$\chi_0^{(2)}(\omega) = \frac{2e^3}{4\pi} \tau |x_{12}^{(0)}|^2 \frac{\sigma_r^2 - \sigma_l^2}{\sigma_L} \frac{\exp\left\{-\frac{1}{2\sigma_\Delta^2} \left(\omega - \bar{\Delta} + \frac{\alpha_1 \sigma_\Delta^2}{\alpha_{21} T}\right)^2\right\}}{\sqrt{2\pi\sigma_\Delta^2}} F\left(\frac{\omega - \bar{\Delta}}{\sigma_\Delta}\right), \quad (19)$$

where the function $F(\Omega)$ is determined by the integral

$$F(\Omega) = \int_0^\infty \frac{xdx}{\sqrt{2\pi}} e^{\frac{1}{2}(x+\Omega)^2} \left[\operatorname{erf}\left(\sqrt{\frac{2\alpha_1\sigma_\Delta}{\alpha_{21}T}} x\right) - \operatorname{erf}\left(\sqrt{\frac{2\alpha_2\sigma_\Delta}{\alpha_{21}T}} x\right) \right]. \quad (20)$$

In Eq. (20) the function $\operatorname{erf}(t)$ is the standard error function. It is evident from expression (19) that a static polarization arises in the field of the electromagnetic wave because of

the difference in the standard deviations of the positions of the right- and left-hand boundaries of the quantum well. The induced-polarization vector \mathbf{P} is directed in the direction of the interface with the stronger fluctuations.

In summary, the joint measurement of the linear and nonlinear responses of a quantum structure offers the opportunity to experimentally study not only fluctuations of the width of the structure, but also to directly analyze the relative magnitudes of the rms deviations of the opposite boundaries of the layer.

In conclusion, we note that the appearance of a temperature-dependent shift, as demonstrated above, of the resonance frequency in the absorption spectrum at frequencies of intersubband transitions [see Eq. (10)] should also be manifested at a frequency corresponding to interband transitions (in both the absorption and luminescence spectra). This question will be investigated in detail in a separate paper.

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