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Collisional broadening and shift of spectral lines in quantum dot lasers

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We estimate homogeneous broadening and shift of optical transition lines in self-assembled quantum dots (SAQD) caused by elastic Coulomb collisions of carriers in wetting layer with carriers in the SAQD. In particular, we demonstrate that the dephasing time for lasing transitions can be $\sim 0.1-1$ ps at carrier densities in wetting layer $\sim 10^{15}\,\mathrm{m}^{-2}$. © *1999 American Institute of Physics.* [S0003-6951(99)01421-7]

Self-assembled quantum dot (SAQD) semiconductor lasers (see Refs. 1 and 2, for instance) attract much attention due to their interesting physics and potential applications. In these lasers, carriers are pumped into barriers around the quantum dots (QDs), and then are captured by the QDs, and relax via 0D energy levels to the low-lying lasing ones. These relaxation processes can strongly affect laser characteristics, and are currently under intense study. While in quantum well (QW) lasers fast carrier capture and relaxation are mediated by carrier-LO phonon interactions, in QD structures carrier relaxation via LO phonon scattering is highly improbable due to the discrete nature of the QD energy levels and the fixed energies of LO phonons. Therefore, observed in SAQD structures carrier relaxation with characteristic times $\sim 1-100$ ps is ascribed, at least, in part to carrier–carrier Coulomb collisions.[3–5]

Some laser behavior and characteristics (mode competition and modulation characteristics, for instance) depend not only on carrier relaxation times in laser structure, but also on the homogeneous linewidth $2\gamma_2$ ($\gamma_2 = 1/T_2$, T_2 is the dephasing time) of lasing transition. T_2 for QDs similarly to bulks and QWs is defined by interactions of QD carriers with surrounding phonons and carriers. The homogeneous linewidth $2\hbar\gamma_2 < 0.5$ meV has been observed in photoluminescence of single QDs at low temperatures, and has been ascribed to carrier-acoustic phonon interactions (see Ref. 6 and references therein). In room temperature QD lasers with carrier injection, many carriers can surround QDs, and their interactions with QD carriers are able to lead to substantial broadening of optical transitions in QDs.

In this letter we estimate theoretically homogeneous broadening and shift of spectral lines in SAQD due to Coulomb collisions of two-dimensional (2D) carriers in wetting layer with QD carriers. The *inelastic* carrier–carrier collisions in QD structures can lead to effective carrier relaxation in QDs,[3–5] and it also implies definite broadening of spectral lines in QD. In this letter we calculate the linewidth broadening due to the *elastic* collisions, and show that these collisions, which do not change occupations of QD levels, can lead to much stronger homogeneous broadening than inelastic collisions.

In our consideration we follow the approach which is utilized successfully in the theory of collisional broadening of atomic spectral lines.[7] Figure 1 illustrates the used model. SAQD of the height H and of the diameter D is located on wetting layer (WL). The 2D density of electrons and holes in WL is N. The carriers impact the QD, changing phase of carrier wave functions in QD. It leads to broadening and shift of QD spectral lines.[7] In the approach, the motion of the carriers in WL is described classically, and the trajectories of motion are assumed to be straight lines. Carrier i ($i=c$ for electrons, and $i=v$ for holes), which moves with the velocity v along trajectory with the impact parameter b (see Fig. 1), changes the transition frequency $\omega=\omega(t)=\omega_o+\Delta\omega_i(t)$ [ω_o is the nonperturbed transition frequency between electron and hole levels in QD, see inset in Fig. 1], and the phase $\eta(t)=\int^t dt\,\Delta\omega_i(t)$ of the QD optical oscillator $f_{\mathrm{osc}}\sim\exp[i\omega_o t+i\eta(t)]$. The frequency deviation $\Delta\omega_i$ is defined by the distance $R(t)$ between the perturbing carrier i and QD (see Fig. 1): $\Delta\omega_i(t)=\Delta\omega_i[R(t)]$. The overall phase change due to the collision,

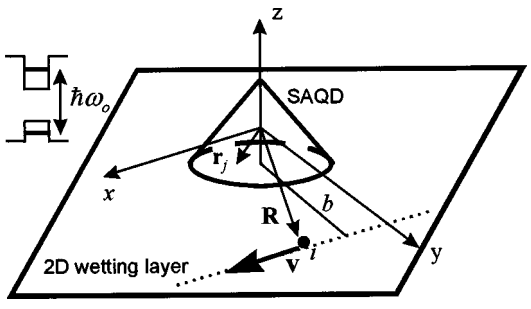


FIG. 1. Illustration of considered SAQD structure. SAQD is a cone of the height H and the diameter D is located on the wetting layer. **R** is the radius vector of the wetting layer carrier i which has the velocity v and collides with QD with the impact parameter b. The term $\mathbf{r}_j$ is the radius vector of the carrier j in QD. Inset shows schematically the band gap diagram of the SAQD. The term $\hbar\omega_o$ is the transition energy between electron and hole ground states in SAQD.

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$$\overline{\eta_i} = \overline{\eta_i}(v,b) = \int_{-\infty}^{+\infty} dt \cdot \Delta\omega_i(t), \tag{1}$$

depends on the impact parameter b and the carrier velocity v. The collisional line shape of the transition is Lorentzian with the maximum at $(\omega_o + \Delta)$ and the width $2\gamma_2$, the shift Δ and the halfwidth γ_2 of spectral line being found by averaging over WL carriers which collide with QD.[7] The collisional shift Δ and the halfwidth γ_2 can be written as $\Delta = \Delta_c + \Delta_v$ and $\gamma_2 = \gamma_{2c} + \gamma_{2v}$, accordingly, where γ_{2c} (γ_{2v}) and Δ_c (Δ_v) are the contributions into the broadening and shift of line, correspondingly, from collisions with electrons (holes)

$$\gamma_{2i} = \int_0^\infty dv f_i(v) v \sigma_b^i(v), \quad \Delta_i = \int_0^\infty dv f_i(v) v \sigma_s^i(v). \tag{2}$$

The term $f_i(v) = N \cdot m_i v / k_B T \cdot \exp(-m_i v^2 / 2k_B T)$ is the Boltzmann distribution over velocities for carriers i with the mass m_i in WL. The 2D cross sections $\sigma_b^i(v)$ and $\sigma_s^i(v)$ are given as

$$\sigma_b^i(v) = 2\int_0^\infty db(1 - \cos\overline{\eta_i}), \quad \sigma_s^i(v) = 2\int_0^\infty db \sin\overline{\eta_i}. \tag{3}$$

Equations (1)–(3) allow one to calculate the linewidth $2\gamma_2$ and the shift Δ at known dependence $\Delta\omega_i = \Delta\omega_i(R)$. The frequency deviation $\Delta\omega_i = (\Delta E_{ic} + \Delta E_{iv})/\hbar$, where $\Delta E_{ic}(R)$ [$\Delta E_{ic}(R)$] is the shift of electron [hole] energy in QD by Coulomb field of carrier i in WL at the distance R from QD (see Fig. 1). The energy shifts $\Delta E_{ij}(R)$ can be evaluated with the perturbation theory in the dipole approximation for the Coulomb energy of the carrier j in QD

$$V(\mathbf{r}_j) = \frac{q_i q_j}{4\pi\epsilon\epsilon_o} \frac{1}{|\mathbf{R} - \mathbf{r}_j|} \approx \frac{q_i q_j}{4\pi\epsilon\epsilon_o}\left(\frac{1}{R} + \frac{\mathbf{R}\mathbf{r}_j}{R^3}\right), \tag{4}$$

where q_j and $\mathbf{r}_j$ are the charge and the radius vector of the carrier j in QD, accordingly (Fig. 1). Below we consider the transition between electron and hole ground states. In the first order perturbation theory we have

$$\Delta E_i^{(1)} = \frac{q_i q}{4\pi\epsilon\epsilon_o} \cdot \frac{\mathbf{R}}{R^3} \cdot \langle|(\mathbf{r}_v - \mathbf{r}_c)|\rangle, \tag{5}$$

where $q = |q_j|$, and $\langle|(\mathbf{r}_v - \mathbf{r}_c)|\rangle$ is the dipole moment of QD in the ground state. If electron and hole wave functions coincide in the ground state, the dipole moment is zero. Due to strain and asymmetry of SAQD structure, strong internal electric field is present in SAQDs, so that electrons and holes in QDs are shifted from each other.[8] In this case the dipole moment is nonzero. Owing to assumed axial symmetry of the QD structure, the dipole moment is directed along the axis z (Fig. 1), and its z component can be written as βH. In general, the coefficient β can be positive or negative, and in strong internal electric fields its magnitude approaches 1. Accordingly

$$\Delta E_i^{(1)} = -\frac{q_i q}{4\pi\epsilon\epsilon_o} \cdot \frac{\beta H^2}{2R^3}. \tag{6}$$

In fact, Eq. (6) describes the linear Stark effect in QD.

The second order perturbation theory gives the quadratic Stark effect in the field of WL carrier

$$\Delta E_i^{(2)} = -K_F \cdot F^2, \tag{7}$$

where $F = q_i/(4\pi\epsilon\epsilon_o R^2)$ is the electric field in the center of QD created by the WL carrier i. Generally, due to anisotropy of SAQD, the coefficient K_F depends on the direction of $\mathbf{R}$. Below we neglect this dependence, and consider K_F as constant.

From Eqs. (6) and (7) we can write for the frequency deviation

$$\Delta\omega_i = -\frac{C_{3i}}{R^3} - \frac{C_{4i}}{R^4} \tag{8}$$

with

$$C_{3i} = \frac{q_i q}{4\pi\epsilon\epsilon_o\hbar} \cdot \frac{\beta H^2}{2}, \quad C_{4i} = \frac{q^2}{16\pi^2\epsilon^2\epsilon_o^2} \cdot \frac{K_F}{\hbar}. \tag{9}$$

The constant C_{4i} is always positive, but the constant C_{3i} can be positive or negative in dependence of the signs of β and the q_i (i.e., electron or hole collides with QD). Below we consider separately the collisional broadening and shift of transition line due to ''linear Stark effect''—the first term in Eq. (8), and ''quadratic Stark effect''—the second term in Eq. (8).

In accordance with Eq. (2), the collisional half width and shift of spectral line can be written as

$$\gamma_2 = A \cdot N, \quad \Delta = B \cdot N, \tag{10}$$

where the broadening coefficient A, and the line shift coefficient B can be written as $A = A_c + A_v$ and $B = B_c + B_v$. The coefficients A_c and B_c (A_v and B_v) describe the contribution of electrons (holes) of WL into the broadening and shift of line, accordingly, and

$$A_i = v_{T_i}\rho_{oi}F(\alpha_i), \quad B_i = v_{T_i}\rho_{oi}G(\alpha_i), \tag{11}$$

where $v_{T_i} = \sqrt{2k_BT/m_i}$ is the thermal velocity of the carriers i. The Weisskopf radius ρ_{oi}[7] is an important parameter characterizing the collisions: if carrier passes near QD at the minimal distance $R_{\min} = (H^2/4 + b^2)^{1/2}$ which is less than the Weisskopf radius ρ_{oi} ($R_{\min} < \rho_{oi}$), the carrier perturbs substantially the phase $\overline{\eta}$ ($\overline{\eta} > 1$), and contributes strongly into the collisional broadening and shift of QD spectral line. Carriers with $R_{\min} > \rho_{oi}$ broaden and shift QD line weakly. The radius ρ_{oi}, the dimensionless parameter $\alpha_i = (H/2\rho_{oi})^2$, and the view of the dimensionless functions F and G are defined by the dependence (8). In particular, $\rho_{oi} = \sqrt{2|C_3|/v_{T_i}}$ for ''linear Stark effect,'' and $\rho_{oi} = (\pi C_4/2v_{T_i})^{1/3}$ for quadratic Stark effect.

We have used the abovementioned formulas to calculate the collisional broadening and shift of spectral lines for InAs/GaAs SAQD. In calculations we assumed $m_c = 0.023$, $m_v = 0.34$ for carrier masses in wetting layer. Figure 2 shows the broadening (solid lines) and shift (dash lines) coefficients for linear Stark effect as a function of the QD height H. β is assumed to be positive. For linear Stark effect, the Weisskopf radius $\rho_{oi} \propto H$, the parameter α_i in Eq. (11) does not depend on H, so that A and B are proportional to H (Fig. 2). In the broadening coefficient A, the contributions from elec-

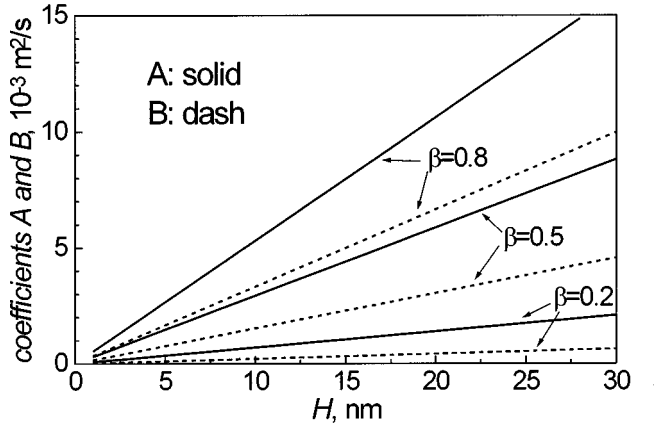


FIG. 2. The collisional broadening coefficient A (solid) and the shift coefficient B (dash) in the case of linear Stark effect as a function of the QD height H at different values of the coefficient β.

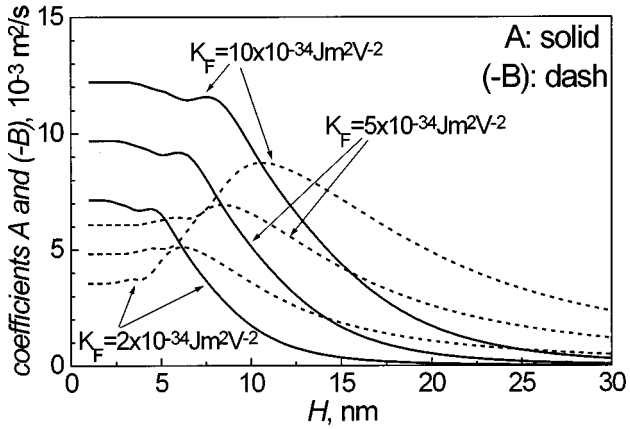


FIG. 3. The collisional broadening coefficient A (solid) and the shift coefficient $(-B)$ (dash) in the case of quadratic Stark effect as a function of the QD height H at different values of the coefficient K_F.

tron and hole collisions with QD are of the same order and are added to each other. Inversely, in the shift coefficient B, the electron and hole collisions contribute with different signs, and are subtracted. Nonzero result for the net shift is due to the difference in electron and hole masses, and accordingly due to the difference in carrier distributions over velocities in the wetting layer. At the used values for the masses, the contribution from electrons is larger than from holes, and this is mainly because the thermal velocity v_{T_i} for electrons is larger than for holes. For $\beta>0$ the net shift is positive (''blueshift''). Figure 2 shows that for $|\beta|>0.2$ one should expect the dephasing time $T_2\sim 0.1-1$ ps at $N=10^{15}\,\mathrm{m}^{-2}$.

In the case of quadratic Stark effect the broadening and shift of line are defined by the coefficient K_F [see Eq. (7)]. One can estimate the coefficient[9] as

$$K_F=q^2\cdot(L_{10}^{v})^2/(E_1^{v}-E_0^{v}).$$

L_{10}^{v} is the dipole moment between the first excited and ground hole levels. In the estimation we take into account only the contribution from transition between two hole levels because the distance $E_1^{v}-E_0^{v}$ between these excited and ground levels for holes is smaller than according distance for electrons. Assuming $E_1^{v}-E_0^{v}\sim 10\,\mathrm{meV}$ and $L_{10}^{v}\sim 5\,\mathrm{nm}$, one can get $K_F\sim 5\times 10^{-34}\,\mathrm{Jm^2\,V^{-2}}$. This estimation is in agreement with the calculation of confined Stark effect in quantum disks.[10] Figure 3 shows the broadening (solid lines) and shift (dash lines) coefficients for quadratic Stark effect as a function of the QD height H at different values for K_F. The shift coefficient B is negative. This ''redshift'' of spectral line due to quadratic Stark effect can be considered as an analog of band gap shrinkage in bulks and QWs caused by Coulomb interactions between carriers.[11] In Fig. 3, A and $(-B)$ decrease at large H. The behavior is caused by reduction of the cross-sections $\sigma_b^i(v)$ and $\sigma_s^i(v)$ with an increase of H: if H becomes larger than the Weisskopf radius ρ_{oi}, carriers in WL move at the distances $R_{\min}>\rho_{oi}$, and perturb QD weakly. Respectively, the collisional broadening and line shift decrease. Strictly speaking, the coefficient K_F depends on the H, and can enlarge with H. But if L_{10}^{v} in Eq. (18) is defined mainly by the QD diameter D, we can assume that K_F does not depend on H. Figure 3 shows that at $N=10^{15}\,\mathrm{m}^{-2}$ one should expect the dephasing time $T_2\sim 0.1-1$ ps.

We have considered the contributions from linear and quadratic Stark effects separately. In real SAQD structures these effects can present simultaneously. Addition of the contributions can have a complicated character, and needs special consideration.

In conclusion, we have demonstrated that elastic collisions of 2D carriers of wetting layers with SAQDs can lead to the dephasing time $T_2\sim 0.1-1$ ps at the carrier densities $N\sim 10^{15}\,\mathrm{m}^{-2}$. The time is much shorter than collisional carrier relaxation times in QDs at the same carrier densities.[5] The situation, when homogeneous broadening is defined rather by elastic dephasing collisions than by inelastic collisions, is typical for atomic gases.[7] The collisional broadening and shift of spectral lines in QDs could be studied with four-wave mixing experiments on SAQD semiconductor optical amplifiers.

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