Numerical Study of the Exciton-light Coupling in Quantum Wells

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Abstract— The exciton-light coupling is an important characteristic quantitatively described by the radiative decay rate. Recent experimental studies of the radiative decay rate for high-quality quantum wells show a variance of the experimental data especially for narrow quantum wells. Therefore, the theoretical and numerical studies of the exciton-light coupling as an overlap of the light wave and the exciton wave function are of significant interest. In this paper, we present the radiative decay rates obtained for excitons in the GaAs-based quantum wells of various widths from 1 nm up to 100 nm. The exciton wave function is calculated by the direct numerical solution of the three-dimensional Schrödinger equation. The precise results for the lowest exciton state are obtained by the Hamiltonian discretization using the finite-difference scheme. The numerical results are compared with the experimental data extracted from the reflectance spectroscopy measurements for high-quality GaAs/AlGaAs and InGaAs/GaAs heterostructures.

1. INTRODUCTION

In the last decade, the increased computer performance has provided an exciting opportunity for numerical study of excitons in quantum wells (QWs) as well as of the exciton-light coupling. The analytical description of the exciton states in heterostructures with QWs is difficult due to the degenerate valence band and the impact of the QW potential. An interplay of the QW potential and of the Coulomb electron-hole interaction additionally constrains the possibility for solution of such problems. They can be solved approximately only in the limiting cases of very narrow or very wide QWs ($L < 10\,\mathrm{nm}$ and $L > 150\,\mathrm{nm}$ for GaAs) [1]. Meanwhile, the quality of the heterostructures is constantly growing and new experimental data on exciton-light coupling have become available recently [2, **?**, **?**, **?**]. Therefore, theoretical and numerical studies of the radiative decay rate as the main exciton-light coupling characteristic are of significant interest.

We have developed a method for the precise numerical solution of the Schrödinger equation for a problem of an exciton in the square QW of a finite height [6, 7]. The Schrödinger equation includes the coordinate-dependent mass parameters and takes into account discontinuity of the dielectric constants at the QW interfaces. Due to the cylindrical symmetry of the problem, one can separate three out of the six exciton coordinates. Here, we neglect small effects of the corrugation of the valence band and consider only the excitons with zero in-plane wave vectors, that is typically studied in experiments with the normal incidence and detection of light. Then, the obtained three-dimensional equation is solved by the finite-difference method. As a result, the exciton energies and corresponding wave functions depending on the remaining three coordinates are determined. The latter allowed us to calculate the radiative decay rate of the exciton ground state with a good precision, in particular for narrow QWs.

2. THEORETICAL MODEL

The exciton states in a rectangular QW are defined by the three-dimensional equation derived from the Schrödinger equation for exciton. For description of the degenerate valence band, the diagonal terms of the Luttinger Hamiltonian [8] are used, thus neglecting the corrugation of the valence band. This three-dimensional equation for s-wave exciton states is given as [9]

$$\left(K + V_e^{pot}(z_e) + V_h^{pot}(z_h) + V_e^{self}(z_e) + V_h^{self}(z_h) + V^C(\rho, z_e, z_h)\right)\chi(z_e, z_h, \rho) = E_X \chi(z_e, z_h, \rho), \quad (1)$$

where the kinetic term K reads

$$K = -\frac{\hbar^2}{2}\frac{\partial}{\partial z_e}\frac{1}{m_{ez}(z_e)}\frac{\partial}{\partial z_e} - \frac{\hbar^2}{2}\frac{\partial}{\partial z_h}\frac{1}{m_{hz}(z_h)}\frac{\partial}{\partial z_h} - \frac{\hbar^2}{2}\frac{1}{\mu_{xy}}\left(\frac{\partial^2}{\partial \rho^2} - \frac{1}{\rho}\frac{\partial}{\partial \rho} + \frac{1}{\rho^2}\right).$$

In Eq. (1), indices e and h denote the electron and the hole, respectively. The unknown function χ is related to the three-dimensional wave function ψ as [6, 7] $\psi(z_e, z_h, \rho) = \chi(z_e, z_h, \rho)/\rho$. The potentials $V_{e,h}^{pot}(z_{e,h})$ are the finite rectangular confinement QW potentials. The self-interaction potentials $V_{e,h}^{self}(z_{e,h})$ come from the interaction of carriers with their images appeared due to the dielectric constant mismatch between the QW and barrier materials. The Coulomb potential $V^C(\rho, z_e, z_h)$ also takes into account the dielectric constant mismatch. This mismatch gives rise to the electrostatic perturbation expansions over a small parameter $(\epsilon_w - \epsilon_b)/(\epsilon_w + \epsilon_b)$, where $\epsilon_{w,b}$ are the dielectric constants in the QW and in the barrier [9, 10]. In Eq. (1), the term $\mu_{xy} = m_{exy} m_{hxy}/[m_{exy} + m_{hxy}]$ is the reduced effective mass in the xy-plane.

The exciton-light coupling is usually characterized by the radiative decay rate [1, ?], Γ_0, which describes decay of the electromagnetic field emitted by an exciton ensemble after the pulsed excitation: $E(t) = E(0)\exp(i\omega_0 t - \Gamma_0 t)$. For the exciton in a QW, the radiative decay rate is given by the expression [1, ?]:

$$\Gamma_0 = \frac{2\pi q}{\hbar\epsilon}\left(\frac{e|p_{cv}|}{m_0\omega_0}\right)^2 \left|\int_{-\infty}^{\infty} \psi(z_e = z_h = z, \rho = 0)\exp(iqz)dz\right|^2, \tag{2}$$

where $q = \sqrt{\epsilon}\omega/c$ is the light wave vector, ω_0 is the exciton frequency, $|p_{cv}|$ is the matrix element of the momentum operator between the single-electron conduction- and valence-band states.

3. NUMERICAL METHOD

We numerically solved the boundary value problem for Eq. (1) and accurately obtained the exciton ground state energy E_X and the wave function $\chi(z_e, z_h, \rho)$. The exponential decrease of the exciton wave function at large values of variables allowed us to impose zero boundary conditions for the function χ at the boundary of some rectangular domain. For discretization, we employed the second-order finite-difference approximation [13] of the partial derivatives in Eq. (1) on the equidistant grids over three variables. We use the central second-order finite-difference formula for approximation of the terms with the second partial derivative over z with the discontinuity at the interface [14, ?]:

$$-\frac{\hbar^2}{2\Delta_z^2}\left(\frac{2}{m_{i-1}+m_i}\chi(z_{i-1}) - \left[\frac{2}{m_{i-1}+m_i} + \frac{2}{m_i+m_{i+1}}\right]\chi(z_i) + \frac{2}{m_i+m_{i+1}}\chi(z_{i+1})\right). \tag{3}$$

The grid steps over each variable have been taken to be the same, $\Delta = \Delta_{z_e} = \Delta_{z_h} = \Delta_\rho$. Eq. (3) defines the theoretical uncertainty of the numerical solution of order of Δ^2 as $\Delta \to 0$. The discontinuities of the rectangular confinement potential are treated in a similar way.

The nonzero solution of the homogeneous equation (1) with zero boundary conditions can be found by a diagonalization of the matrix constructed from this equation. A small part of the matrix spectrum was obtained by the Arnoldi algorithm [16]. As a result, we have calculated the lowest eigenvalue of the matrix and the corresponding eigenvector. After the extrapolation to the limit $\Delta = 0$ [6], the accurate asymptotic results are obtained. Thus, we found the exciton states for various widths of QWs. Then, the radiative decay rate was calculated using the trapezoidal rule for integral in Eq. (2).

The calculations were performed for the GaAs/$Al_xGa_{1-x}As$ heterostructures with various concentrations of the solid solutions in barrier layers. Material and energy gap parameters used for solving the eigenvalue problem (1) are based on the data from Refs. [17, ?]. In particular, the difference of the gap energies, as a function of x, is modeled as $\Delta E_g = 1087x + 438x^2$ meV. A ratio of potential barriers is taken to be $V_e/V_h = 65/35$. The Luttinger parameters used in the calculations are $\gamma_1 = 6.85$, $\gamma_2 = 2.10$ for GaAs and $\gamma_1 = 3.76$, $\gamma_2 = 0.82$ for AlAs; the dielectric constants are 12.53 and 10.06, respectively. Masses and dielectric constants for the ternary alloys are obtained by a linear interpolation on x.

4. RESULTS

The exciton-light coupling was studied for the GaAs/$Al_xGa_{1-x}As$ QW heterostructures with Al concentration, $x = 0.30$, and the $In_xGa_{1-x}As$/GaAs structures with small In concentration, $x = 0.02$–0.07. The widths of QW was varied from 1 nm to 100 nm. In Figure 1 one can see the radiative decay rate in energy units, $\hbar\Gamma_0$, of the exciton ground state. It was calculated for the case when the dielectric and mass parameters are taken to be the same in the barrier and in the QW. The radiative decay rate generally decreases as the QW width diminishes. So, the strength of the exciton-light

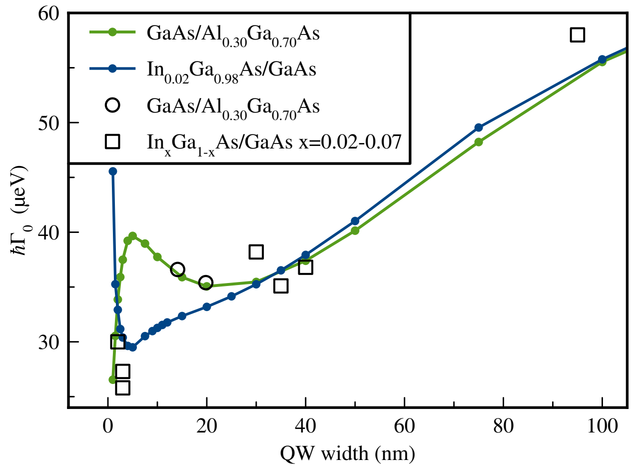

Figure 1. The calculated radiative decay rate of the exciton ground state in GaAs/$Al_{0.30}Ga_{0.70}As$ and $In_{0.02}Ga_{0.98}As$/GaAs QWs. The experimentally measured data are shown by open circles and squares.

interaction reduces and, on the other hand, the exciton lifetime increases. The most long-lived exciton in the GaAs/$Al_{0.30}Ga_{0.70}As$ QW is found to be for QW width of about 20 nm where the minimum of $\hbar\Gamma_0$ is achieved.

The results of our calculations are confirmed by our experimental measurements. For comparison, the high-quality heterostructures with QWs grown by molecular beam epitaxy have been selected to determine their reflectance spectra. The spectra were measured by a simple setup consisting of a white light source, a cryostat, and a spectrometer equipped with a CCD camera. To accurately calibrate the absolute value of reflectance, a monochromatic light of a continuous-wave titanium-sapphire laser was used to measure the reflectance at a spectral point beyond the exciton resonances. Measured reflectance spectra allowed us to extract the radiative decay rate, see Ref. [1, ?] for details. Figure 1 also shows the measured $\hbar\Gamma_0$ data for exciton in GaAs/$Al_{0.30}Ga_{0.70}As$ and $In_xGa_{1-x}As$/GaAs with In concentration $x = 0.02$–0.07 heterostructures. The calculated and measured data are in good agreement, especially for Al-doped heterostructures.

For narrow GaAs/$Al_{0.30}Ga_{0.70}As$ QWs, the situation is more complicated due to a noticeable penetration of the exciton wave function in the barrier layers as well as due to a valuable effect of the charge images. Therefore, simulation of such QWs should properly take into account the discontinuities of the material parameters at the interfaces. Figure 2 shows results of such calculations of $\hbar\Gamma_0$ together with the case when the QW material parameters are chosen to be the same as in the barrier. One can see the dramatic difference in the radiative decay rate behavior. If the material parameters are properly taken into account, the radiative decay rate for narrow QWs becomes of about twice larger than that without accounting: 48 μeV versus 27 μeV. Therefore, the

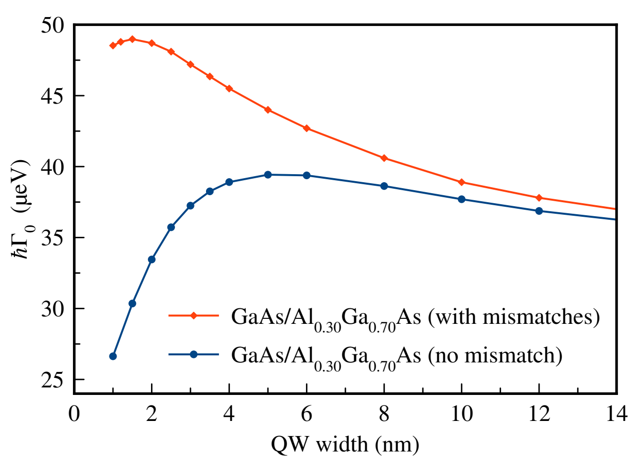

Figure 2. The calculated radiative decay rate of the exciton ground state in narrow GaAs/$Al_{0.30}Ga_{0.70}As$ QWs with an account for the material parameter mismatch as well as without one.

material parameter's mismatch significantly affects $\hbar\Gamma_0$ in narrow QWs. The radiative decay rate for the exciton in $In_xGa_{1-x}As$/GaAs heterostructures with small In concentration $x = 0.02$ are almost not changed even for narrow QWs. This is due to the small mismatch of the mass and dielectric constants.

Our calculations with the mass and dielectric parameter mismatches at the interfaces for narrow GaAs/$Al_{0.30}Ga_{0.70}As$ QWs substantially improve the data given in Ref. [6]. Our numerical results for such QWs are also in agreement with the experimental results obtained by Poltavtsev et al. [2, ?].

5. CONCLUSION

In summary, we have numerically studied the radiative decay rate of excitons in GaAs/$Al_{0.30}Ga_{0.70}As$ and $In_xGa_{1-x}As$/GaAs ($x = 0.02$–0.07) QWs for various QW widths. The calculations were based on the solution of the Schrödinger equation for exciton, taking into account the discontinuities of the material parameters at the QW interfaces. The improved finite-difference numerical scheme has been developed and realized for this problem. The results of calculations for the narrow GaAs/$Al_{0.30}Ga_{0.70}As$ QWs show that the account for the mass and dielectric constant mismatches dramatically changes the radiative decay rate for these QWs. The numerical results are in good agreement with the original experimental data [6].

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