Exciton Aharonov–Bohm effect and emission kinetics in nanorings

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The Aharonov–Bohm effect of excitons and their relaxation kinetics are investigated within the model of semiconductor nanorings of zero width. The kinetic equations where calculated deformation potential matrix elements for an acoustic phonon scattering are used as an input are solved for the steady state. Photoluminescence quenching is observed for stronger magnetic fields when electron and hole are spatially separated. The non-radiative decay plays a decisive role.

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1 Introduction

The exciton properties in semiconductor nanorings have gained a lot of interest recently. Attention has been focused mainly on the exciton Aharonov–Bohm effect (X-ABE), i.e. an oscillatory component in the energy of the optically active (bright) states, the persistent current and the magnetization (see Ref. [1] and references therein). When electron and hole are spatially separated (either by confinement or electric field), then the exciton ground state is not optically active for stronger magnetic fields [2–4]. Therefore, the X-ABE in photoluminescence (PL) experiments may be difficult to observe.

In this paper we investigate theoretically the exciton kinetics within the model of a nanoring with zero width. Even though this model has some limitations [1], it still captures the main physical features. Its simplicity enables to proceed with analytical derivations, saves computational time, and gives a qualitative idea of the photoluminescence dependence on the magnetic field. We derive expressions for the exciton–phonon scattering matrix elements and show an example of the photoluminescence quenching with increasing magnetic field under steady state conditions, including a non-radiative decay.

We assume that both the electron and the hole are strongly confined in a narrow quantum well in the z-direction, and further in the lateral direction in the narrow nanoring where the cylindrical symmetry is supposed. This enables to write the exciton wave function in the envelope function formalism in cylindrical coordinates \((r, \phi, z)\) as

\[
\psi_a(r, \phi, z) = \psi_a(\phi, \phi_h) u_a(r_a) v_a(z) v_b(z) ,
\]

where \(v_a(z)\) are \(z\) confinement wave functions centered at \(z = 0\) and \(u_a(r_a)\) are radial confinement wave functions centered around the ring radius \(R_a\) which might be different for electron and hole (type II behavior). The azimuthal boundary conditions have the usual form of

\[
\psi_a(\phi_a, \phi_h) = \psi_a(\phi_a + 2\pi, \phi_h) = \psi_a(\phi_a, \phi_h + 2\pi) .
\]
Omitting the confinement energies, the Hamiltonian for the angle dependent part $\psi_{\alpha}(\phi, \phi')$ reads

$$\hat{H} = \sum_{\alpha=\phi,\phi'} \left[ \frac{1}{2m_cR_\alpha^2} \left( \hbar \left[ \frac{\partial}{\partial \phi} + \frac{1}{2} \frac{\partial}{\partial \Phi} \right] - \frac{eB}{2} R_\alpha^2 \right)^2 \right] + V_C(\Phi, R_c, R_b),$$

(3)

where relative $\phi = \phi - \phi'$ and average angle $\Phi = \frac{1}{2}(\phi + \phi')$ have been introduced. The parameter $a$ in the Coulomb potential

$$V_C(\Phi, R_c, R_b) = \frac{e^2}{4\pi \varepsilon_0 \varepsilon_s \sqrt{a^2 + R_c^2 - 2R_cR_b \cos(\phi)}},$$

(4)

was adjusted in order to reproduce binding energies calculated for a finite width nanoring [1]. $\varepsilon_s$ is the static dielectric constant. Since the Hamiltonian Eq. (3) does not depend explicitly on the coordinate $\Phi$, the wave function can be even further factorized and the part dependent on the relative angle $\phi$ can be expanded as

$$\psi_{\alpha}(\phi, \Phi) = \sum_c c^\alpha_c e^{-i\phi} \frac{\psi_{\alpha_c}(\phi)}{2\pi},$$

(5)

where $m_\alpha$ is the integer quantum number of the total angular motion of state $\alpha$. The application of the boundary conditions Eq. (2) selects the rules for $l$ which depends on $m_\alpha$

$$m_\alpha \text{ even : } l \text{ integer; } m_\alpha \text{ odd : } l \text{ half integer.}$$

(6)

The Hamilton matrix for the ansatz Eq. (5) can be derived straightforwardly

$$\hat{H}_{\alpha\alpha}^{\text{C}} = \frac{\hbar^2}{2\mu_c R_c^2} \left[ (l + \frac{eB}{2\hbar} R_c^2 + \frac{m_\alpha}{2} p)^2 + \frac{\hbar^2}{2(m_cR_c^2 + m_bR_b^2)} \left( \frac{eB(R_c^2 - R_b^2)}{2\hbar} - m \right)^2 \right] + \hat{V}_{\alpha\alpha}^{\text{C}}(R_c, R_b),$$

(7)

where $\mu_c = m_m/m_a + m_b$ is the reduced exciton mass, $R_c$ an effective ring radius for the exciton, and $p$ a phase shift

$$R_c^2 = \frac{R_g^2 R_b^2 (m_a + m_b)}{m_a R_c^2 + m_b R_b^2}; \quad p = \frac{m_b R_c^2 - m_b R_b^2}{m_a R_c^2 + m_b R_b^2}. $$

(8)

The Coulomb potential matrix elements are given by

$$\hat{V}_{\alpha\alpha}^{\text{C}}(R_c, R_b) = \frac{1}{2\pi} \int_0^{2\pi} V_C(\phi, R_c, R_b) \cos(k\phi) d\phi.$$ 

(9)

The dependence of the eigenenergy $E_{\alpha}$ on the magnetic field can be divided into two parts

$$E_{\alpha}(B) = E_{\alpha}(0) + \Delta E^{(1)}_{\alpha}(B) + \Delta E^{(2)}_{\alpha}(B),$$

(10)

where $E^{(1)}_{\alpha}(B)$ is a periodic function of the $B$-field (X-ABE component of the energy) with the period $B_p$ (independent of the quantum number $m_\alpha$) and $\Delta E^{(2)}_{\alpha}(B)$ (the second term in the square brackets of Eq. (7)) is a parabolic function of the $B$-field with zero of the energy shifted to $B_{\alpha,m_\alpha}$

$$B_p = \frac{2\hbar}{e R_c^2}; \quad B_{\alpha,m_\alpha} = m_\alpha \frac{2\hbar}{e R_c^2 - R_b^2}. $$

(11)
The knowledge of the eigenenergies \( E_\alpha \) and corresponding wave functions \( \psi_\alpha \) allows us to calculate the deformation potential scattering between excitons and acoustic phonons. We suppose that excitons are at the final stage of their thermalization after optical excitation and that their density is such low that any exciton–exciton scattering is negligible.

First, we start with the radiative rates, which determine the probability of the exciton annihilation by producing a photon. They can be written in a compact way [6, 7]

\[
\begin{align*}
    r_\alpha &= r_\alpha \left| \int dr \psi'_\alpha(r, r') \right|^2; \\
    r_{\text{sc}} &= \frac{4}{3} \frac{d_{\text{e}} E_{\text{g}} n}{\hbar^3 c^3},
\end{align*}
\]

(12)

where \( d_{\text{e}} \) is the interband dipole matrix element, \( E_{\text{g}} \) is the bandgap plus confinement energy, and \( n \) the refractive index. Using the factorizations Eq. (1) and (5) the radiative rates can be written as

\[
    r_\alpha = r_\alpha \delta_{m_\alpha 0} \sum_i c_i^\alpha \int dz v_\alpha(z) v_\beta(z) \int dr r u_\alpha(r) u_\beta(r) \left| \psi_\alpha \right|^2.
\]

(13)

It should be stressed that only states with \( m_\alpha = 0 \) are optically active (bright). For the sake of the simplicity, we approximate the integrals in the calculation of the radiative rates Eq. (12) as

\[
    \int dz v_\alpha(z) v_\beta(z) = 1; \quad \int dr r u_\alpha(r) u_\beta(r) = e^{-i q R} \sigma_z / \pi^2,
\]

(14)

where \( \sigma_z \) is an effective length describing the overlap of the electron and hole radial functions.

Second, we focus on the calculation of scattering rates between exciton states \( \alpha \) and \( \beta \) due to acoustic phonons which are then given by [5]

\[
\begin{align*}
    \gamma_{\alpha\beta} &= \frac{2 \pi}{\hbar} \sum_q \left| t_{\alpha\beta}^q \right|^2 \left[ (n_s (\hbar \omega_q) + 1) \delta(\omega_\beta - \omega_\alpha - \hbar \omega_q) + n_s (\hbar \omega_q) \delta(\omega_\beta - \omega_\alpha + \hbar \omega_q) \right], \\
    t_{\alpha\beta}^q &= \sqrt{\frac{\hbar \omega_q}{2 \pi^3 \rho_\omega V}} \int dr d_r \Psi'_\alpha(r_r, r_s) \left( D_r \exp(irq) - D_r \exp(irq) \right) \Psi'_\beta(r_r, r_s),
\end{align*}
\]

(15)

(16)

where \( n_s (\hbar \omega_q) \) is the Bose–Einstein occupation function, \( \omega_q = |q| \) the acoustic–phonon dispersion energy, \( s \) the sound velocity, \( \rho \omega_\omega \) the mass density, \( V \) the sample volume, and \( D_r \) \((D_r)\) the deformation potential for electron (hole). The scattering matrix elements \( t_{\alpha\beta}^q \) are calculated in spherical \( q \) coordinates. Using Eq. (1) and (5) gives, up to a phase factor,

\[
    t_{\alpha\beta}^q = \sqrt{\frac{\hbar \omega_q}{2 \pi^3 \rho_\omega V}} \left( S_{\alpha\beta}^q \right) \left( K_r(q_r) K_s(q_s) \left( D_r \right) \left( D_r \right) \right)_{m_\alpha m_\beta = m_\alpha m_\beta},
\]

(17)

introducing state dependent “overlap” sums \( S_{\alpha\beta}^q \), an integral of the Bessel functions \( J_\mu(x) \), and a \( z \)-dependent contribution

\[
    S_{\alpha\beta}^z = \sum_i c_i^\alpha c_i^\beta; \quad I^r_m(q_r) = \int dr r u^r_\alpha(r) J_m(q_r r); \quad K^r_m(q_r) = \int dz v^r_\alpha(z) e^{-i q_r z}.
\]

(18)

In order to simplify the calculation of the scattering rates \( t_{\alpha\beta}^q \), we use the assumptions of a narrow quantum well and of very narrow nanorings with radii \( R \). Then, treating \( z \)-confinement and the radial wave functions as delta functions gives \( K^r_m(q_r) = 1 \) and \( I^r_m(q_r) = J_m(q_r R) \).
Further, the scattering matrix elements $t_{\alpha\beta}^q$ are integrated over $q$ to obtain the scattering rates $\gamma_{\alpha\beta}$ depending on the energy difference $\Delta E = E_{\alpha} - E_{\beta} = \hbar q$:

$$\gamma_{\alpha\beta}(\Delta E) = \gamma_0 q_d(\Delta E) \Delta E^{\gamma}(\text{S}_{\alpha\beta}^q)^2 \left[ 2I_{\alpha\beta}^j + \text{S}_{\alpha\beta}^qI_{\alpha\beta}^{\gamma j} \right] - 2D_\gamma \text{S}_{\alpha\beta}^q \text{S}_{\alpha\beta}^qI_{\alpha\beta}^\gamma \right],$$  

$$\gamma_0 = \frac{2\pi^2}{\hbar^2 S_{\rho M}}; \quad I_{\alpha\beta}^{\gamma j} = \int d\theta' \sin (\theta) J_{\alpha} [q \sin (\theta) R_{\alpha}] J_{\beta} [q \sin (\theta) R_{\beta}].$$

The kinetic equation for the occupation $N_\alpha$ of the state $\alpha$ has the form

$$\dot{N}_\alpha = g_\alpha + \sum_\beta \gamma_{\alpha\beta} N_\beta - \left( r_{\alpha} + d_{\alpha} + \sum_\beta \gamma_{\beta\alpha} \right) N_\alpha,$$

where $g_\alpha$ is a state dependent generation (source) term and $d_{\alpha}$ a phenomenological non-radiative decay rate, representing processes as e.g. exciton annihilation via impurities or escape into the wetting layer. These equations are solved numerically for the steady state $\dot{N}_\alpha = 0$. The absorption $A(E)$ and photoluminescence $P(E)$ spectra have standard form of [9]

$$A(E) = \sum_\alpha r_\alpha \delta(E_{\alpha} - E); \quad P(E) = \sum_\alpha r_\alpha N_\alpha \delta(E_{\alpha} - E).$$

A nanoring with radii $R_1 = 12$ nm and $R_0 = 3$ nm is investigated as a model example. Such a configuration can be achieved e.g. in the type II structure, where the electron is confined in the nanoring and the hole is found in the center of the ring in the potential minimum formed due to the strain [1]. GaAs materials parameters are taken from [8] and give $r_\alpha = 8.9 \times 10^7$ ns$^{-1}$, which produces a radiative rate of the lowest state at $B = 0$ T of

$$r_\alpha = r_\alpha e^{-(R_1 - R_0)^{1/2}/\sigma_0} \left[ \sum_j c_j^\alpha \right]^2 = 7.5 \times 10^{-3} \text{ ns}^{-1},$$

where a radial extension $\sigma_0 = 4$ nm was assumed. The phonon matrix elements are of the following orders

$$\gamma_0 = \gamma_0 D_\gamma^2 = 35.4 \times 10^7 \text{ ns}^{-1} \text{ meV}^{-3},$$

where $D_\gamma = 7$ eV was taken. These numbers clearly show that the exciton–phonon scattering dominates the kinetics since $\Delta E$ is of the order of a few meV.

First, we focus on the X-ABE. The separation of electron and hole reduces their mutual Coulomb interaction and consequently increases the Bohr radius. An increased Bohr radius means that the exciton wave function can sample the whole ring more easily, i.e. it has a non zero value at $\phi = \pi$. Nevertheless, only a very weak onset of the oscillation $\Delta E_{\alpha\beta}^{ij}(B)$ can be observed for the lowest bright state shown in Fig. 1a as the lowest full curve. In order to reveal the oscillations the second derivative of the energy with respect to the magnetic field is calculated. The advantage of this approach is that the energy $\Delta E_{\alpha\beta}^{ij}(B)$ contributes a constant only. Figure 1d reveals the first minimum of the oscillation exactly at $B_0/2 = 15.4$ T. In this way it could be possible, in principle, to verify an oscillatory component $\Delta E_{\alpha\beta}^{ij}(B)$ of the total exciton energy.

However, the ground state is not always optically active if the electron and hole are spatially separated since its quantum number $m_{\alpha}$ abruptly changes with the $B$-field in order to minimize the energy contribution $\Delta E_{\alpha\beta}^{ij}(B)$. This kind of behavior is shown in Fig. 1a where the lowest state with $m_{\alpha} = 1$ ($m_{\beta} = 2$) is shown as a dashed (dotted) curve. The minimum of the energy dependence of the $m_{\alpha} = 1$ state on the $B$-
field is found for $B_{z, f} = 9.7$ T. As expected, the crossing point between the states with $m_\alpha = 0$ and $m_\alpha = 1$ ($m_\alpha = 1$ and $m_\alpha = 2$) is found at around $B_{z, f}/2 (3B_{z, f}/2)$.

In order to estimate quantitatively the change of the PL with magnetic field, we concentrate on the steady state solution of the kinetics Eq. (21). Calculated PL spectra Eq. (22) are shown in Fig. 1b and c. Taking into account the excellent optical yield of nanostructures in general, only a very small non-radiative rate $d_\alpha = 0.013r_\alpha$ was assumed. Figure 1b demonstrates the sharp luminescence quenching with increasing $B$-field for a low lattice temperature of $T = 4$ K shortly after the ground state gets dark (at $B_{z, f}/2$). The PL-quenching is much weaker if the temperature is increased up to $T = 40$ K as shown in Fig. 1c where even the second lowest bright state can be observed (magnified by factor of 10).

Summing the kinetic Eq. (21) over all states $\alpha$, a conservation law for the exciton number in the steady state is found

$$\sum_\alpha g_\alpha = \sum_\alpha (r_\alpha + d_\alpha)N_\alpha$$

(25)

Fig. 2 Occupation of the lowest optically active state $N_0$ (solid) and of the lowest optically non-active state $N_1$ with $m_\alpha = 1$ (dashed) for the lattice temperatures a) $T = 4$ K and b) $T = 40$ K. The dotted line is the equilibrium occupation of $N_0^{eq}$ (see text).
Then, if the non-radiative rate is zero ($d_\alpha = 0$) the number of excitons which are (optically) generated and then decay radiatively is identical. Consequently the spectrally integrated PL intensity (see Eq. (22))

$$\int dE \ P(E) = \text{const}_B,$$

is constant with magnetic field. Thus the PL quenching is a clear indication of nonradiative processes ($d_\alpha \neq 0$).

In order to understand the behavior of the PL with $B$-field deeper, the occupation of the lowest bright state (with $m_\alpha = 0$), called 0, and the lowest state with $m_\alpha = 1$, called 1, are plotted as function of the $B$-field in Fig. 2. The low temperature case shows a pronounced behavior with magnetic field. At the zero $B$-field the state 0 is strongly occupied and this does not change up to the crossing point of the energies and occupations of both states. On the contrary, the occupation of the state 1 increases linearly (in $\log_{10}$ plot) since its energy decreases. As the occupation of the state 0 starts to decrease linearly at around $B = 6$ T the PL starts to quench. The case of higher temperature shows similar behavior qualitatively but the change in the occupation of state 0 and 1 are smoother and smaller compared to the low temperature case. Thermal equilibrium between both states would result in $N_0^{eq} = N_0 \exp (- (E_0 - E_1)/k_B T)$ ($k_B$ being the Boltzmann constant, $E_0$ and $E_1$ eigenenergies of both states). As shown by dotted curves in Fig. 2, this is well preserved up to the $B = 14$ T when the ground state changes its quantum number to $m_\alpha = 2$.

In the low temperature case thermal equilibrium is found only among the first few states unlike for the higher temperature where it is achieved for almost all states.

This analysis would suggest to measure at higher temperatures to diminish the PL-quenching. However, an experimental disadvantage for such a choice is the strongly reduced signal to noise ratio. Since our goal is not only to observe the energy of the lowest bright state with $B$-field but to calculate the second derivative from the data, noise-free high quality data are desired as well.

To conclude, we have used deformation potential scattering with exciton kinetic equations for a zero width nanoring to demonstrate the influence of the electron–hole spatial separation on the photoluminescence signal. The luminescence quenching is observed only if non-radiative decay rates, even though small, are introduced. The quenching gets less abrupt with increasing the effective lattice temperature.

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