Photoluminescence spectroscopy of one-dimensional resonant photonic crystals

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Abstract

We report on a theoretical study of photoluminescence (PL) of the resonant one-dimensional photonic crystals, or the near-Bragg quantum-well structures. The PL spectral intensity is found by including random sources in the equations for the exciton dielectric polarization and introducing the discrete Green’s function. The structures without and with the dielectric contrast are considered. The positions of peaks in the calculated PL spectra are in agreement with the real parts of the exciton–polariton eigenfrequencies. A relation between the PL and absorption spectra is discussed.

Keywords: Exciton polariton; Photoluminescence; Multiple quantum wells; Resonant photonic crystal

1. Introduction

The physics of photonic crystals, i.e. structures with periodically modulated dielectric function allowing for Bragg diffraction of light, is a rapidly developing field. Among one-dimensional (1D) photonic crystals, of particular interest are the so-called resonant Bragg structures with the period $d$ satisfying the Bragg condition $d/\lambda(\omega_0) = 0.5$ at the exciton resonance frequency $\omega_0$, see the book [1] and references therein. Experimentally, photoluminescence (PL) spectra of the resonant Bragg and near-Bragg quantum well (QW) structures were studied in Refs. [2–5]. In this work we propose a theory of secondary radiation of exciton–polaritons in multiple QW (MQW) structures, derive equations for the PL spectral intensity, present the results of numerical calculation and compare the PL and absorption spectra.

2. PL of a single QW (SQW)

In order to derive a general equation for the spectral intensity of PL from MQWs we first present that for a SQW structure. In the simplest kinetic theory, the intensity $I_q$ of electromagnetic wave emitted by a SQW and characterized by the wave vector $\mathbf{q}$ can be written by using Fermi’s golden rule:

$$I_q = \hbar \omega_0 w_q, \quad w_q = \frac{2\pi}{\hbar} f_q |M_{qj}|^2 \delta \left( c \frac{\mathbf{q}}{n_b} \cdot M - E_{qj} \right). \quad (1)$$

Here $\omega_0 = \omega_0/q_n$ is the wave frequency, $n_b = \sqrt{\varepsilon_b}$, $\varepsilon_b$ is the dielectric constant of the barrier material assumed to coincide with the background dielectric constant $\varepsilon_a$ of the QW (the general case of structures with the dielectric contrast $\varepsilon_a \neq \varepsilon_b$ is considered in Section 3), $w_q$ is the probability rate for the photon emission per unit in-plane area, $\mathbf{q}$, the in-plane component of $\mathbf{q}$, $E_{qj} = \hbar \omega_0 + (\hbar^2 k^2/2M)$ is the two-dimensional (2D) exciton excitation energy as a function of the 2D wave vector $\mathbf{k}$, $\omega_0$ and $M$ are, respectively, the exciton resonance frequency and in-plane translational effective mass, $f_k$ is the distribution function defined inside the circle $k \leq q_0$, with $q_0$ being the light wave vector at the exciton resonance frequency $q_0 = n_b \omega_0/c$ and $M_k$ is the matrix element of emission of a photon by an exciton with the wave vector $\mathbf{k} = \mathbf{q}$. Eqs. (1) take into account that only excitons with $k < q_0$ can decay radiatively while excitons with $k > q_0$ are dark, they induce in the adjusting barriers evanescent electromagnetic field.

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which exponentially decays as \( \exp(-\sqrt{k^2 - q_0^2}|z|) \) along the normal \( z \) to the interface plane.

Neglecting exciton scattering within the circle \( k \approx q_0 \) we can write

\[
f_k = \frac{1}{\pi_k} G_k, \quad \frac{1}{\pi_k} = 2(\Gamma_{0k} + \Gamma_k),
\]

where \( \Gamma_{0k} \) and \( \Gamma_k \) are the exciton radiative and nonradiative damping rates, \( G_k \) is the rate of acoustic-phonon-induced transitions from the exciton states with the wave vectors \( k' \) lying outside the circle of radiative states into the radiative state \( k \), namely,

\[
G_k = \frac{2\pi}{\hbar} \sum_{k'Q} |V_{Q}|^2 [(N_Q + 1)\delta(h\omega_Q - E_{k'} - h\Omega_Q)\delta_{k,k+Q} + N_Q\delta(h\omega_Q - E_{k'} - h\Omega_Q)\delta_{k,k-Q}].
\]

Here \( Q \) and \( \Omega_Q \) are the acoustic phonon wave vector and frequency, respectively, \( N_Q \) is the phonon occupation number, \( V_Q \) the matrix element of the exciton–phonon interaction, and both the phonon emission and absorption processes are taken into account.

In order to go beyond validity of the kinetic equation and take into account the effect of finite exciton lifetime on the PL spectral width we can replace the exact \( \delta \)-function in Eq. (1) by the smoothed function

\[
\Delta' \left( \frac{\hbar c}{n_b} q - E_{\text{th}} \right) = \frac{1}{\pi} \cdot \frac{\Gamma_{0k} + \Gamma_k}{|q - (N_Q + 1)\hbar\Omega_Q|^2 + (\Gamma_{0k} + \Gamma_k)^2},
\]

where \( \omega(k) = E_k / \hbar \). Then the radiation intensity can be rewritten as

\[
I_q = \hbar c n_b \pi \frac{|M_q|^2 G_q}{|q - (N_Q + 1)\hbar\Omega_Q|^2 + (\Gamma_{0k} + \Gamma_k)^2}.
\]

We assume the typical kinetic energy \( \hbar^2 k^2 / 2M \) to exceed the exciton radiative damping rate. This allows one to retain the exact \( \delta \)-function for the phonon-induced transition rate \( G_k \) in Eq. (3).

Alternatively, Eq. (5) can be obtained with the help of a phenomenological approach, in which noncoherent polarization responsible for luminescence is modeled by a random source term, \( S_k(\omega) \), in the equation of motion of the exciton-related polarization:

\[
[i\omega(k) - i(\Gamma_{0k} + \Gamma_k)]P_k(\omega) = S_k(\omega).
\]

This equation takes into account the interaction between excitons and the emitted radiation by means of the term \( \Gamma_{0k} \), which represents exciton radiative decay rate. We, however, neglect here a radiative shift of the exciton frequency. From Eq. (6) one immediately obtains

\[
\langle |P_k(\omega)|^2 \rangle = \frac{\langle |S_k(\omega)|^2 \rangle}{[\omega(k) - \omega]^2 + (\Gamma_{0k} + \Gamma_k)^2},
\]

where the angular brackets denote the averaging over realizations of the random source.

In the following we focus on normally emitted light \( (q_1 = 0) \) and fix its particular transverse polarization. This will allow us to use the scalar amplitudes \( P_0 \equiv P_{k=0} \) and \( S_0 \equiv S_{k=0} \) instead of the vectors \( P_k \) and \( S_k \) and to present the electric field of the outgoing light wave in the barrier as \( E_0 e^{i\omega z} \) where the coordinate \( z \) is referred to the QW center.

The amplitude \( E_0(\omega) \) and the exciton polarization \( P_0(\omega) \) are related by [1]

\[
E_0(\omega) = \frac{1}{\xi} P_0(\omega), \quad \xi = \frac{\hbar c}{2\pi n_b a},
\]

where \( a \) is the QW thickness assumed to be small as compared to \( q_0^{-1} \). Then for the intensity of normally emitted light we obtain

\[
I(\omega) = \frac{c n_b}{2\pi} \langle |E_0(\omega)|^2 \rangle = \frac{c n_b}{2\pi} \frac{\langle |S_0(\omega)|^2 \rangle}{(\omega_0 - \omega)^2 + (\Gamma_0 + \Gamma)^2},
\]

where \( \Gamma_0 = \Gamma_{k=0} \) and \( \Gamma = \Gamma_{k=0} \). Comparison between Eq. (5) and Eq. (7) allows us to relate the spectral density of the phenomenological source \( \langle |S_0(\omega)|^2 \rangle \) to microscopic characteristics of the system:

\[
\langle |S_0(\omega)|^2 \rangle = \frac{\hbar c}{2\pi n_b a^2} |M_0|^2 G_0.
\]

3. Exciton–polariton luminescence in finite near-Bragg MQW structures

Now we can derive the general equation for the intensity of PL from a structure containing \( N \) equidistant QWs between semi-infinite barrier layers. In the following the QW width, the width of a barrier separating the nearest QWs and the period are denoted by \( a, b \) and \( d = a + b \), respectively, and the QWs are labeled by the index \( n = 1, \ldots, N \). Taking into account that the random sources \( S_n \) in different QWs are uncorrelated we can write the PL intensity of the normally emitted light as a sum:

\[
I(\omega) = \frac{c n_b}{2\pi} \sum_n \langle |E_n(\omega)|^2 \rangle.
\]

Here \( E_n(\omega) \) is the electric field of the secondary radiation outgoing from the \( n \)th QW, i.e., with the radiation source in the \( n \)th QW, and coming out into one of the semi-infinite barriers, say, the field at the plane shifted by \( d/2 \) from the center of the leftmost QW. One can show that it is given by

\[
E_n(\omega) = \left( \begin{array}{c} \frac{1}{1 - i\tau_{n-1}} + \frac{1}{1 - i\tau_{N-n+1}} \\
\tau_{n-1} \end{array} \right),
\]

where \( \tau_m \) and \( \tau_m^* \) are the amplitude reflection and transmission coefficients, respectively, for a substructure comprised of \( m \) QWs and bounded by the planes shifted by \( \pm d/2 \) from the centers of the leftmost and rightmost QWs. For the particular case \( m = 0 \), i.e., when the bounding planes do not enclose any QW, \( \tau_m \) and \( \tau_m^* \) are equal to 0 and 1, respectively. Using properties of reflection and transmission coefficients determined by their places in a
transfer matrix describing propagation of the field through the structure, this expression for $E^{(n)}$ can be presented in an alternative form

$$
E^{(n)} = \frac{I_N}{t_{N-n}} \left( 1 + r_{N-n} e^{i q d} \right) \frac{i}{\xi} S_n e^{-i q d / 2},
$$  

(11)

where $t_N$ is the transmission coefficient for the entire structure, and the resonant denominator describing excitons dressed by the radiative field (term $S_n/[\omega_0 - \omega - i(\Gamma + \Gamma_0)]$) is replaced by the resonant term corresponding to initial purely mechanical excitons ($S_n/[\omega_0 - \omega - i\Gamma]$). Eq. (11) emphasizes the distinct role of two factors determining the luminescent properties of the structures under consideration: transmission coefficient through the structure $t_N$ reflects the role of the normal photonic modes of the MQW structure, while initial exciton susceptibility determines the efficiency of the initial excitation of the excitons by the noncoherent source.

In structures without the dielectric contrast one can use a simpler approach based on a set of coupled linear equations

$$(\omega_0 - \omega - i\Gamma)P_{n'} + \sum_{n''} A_{n'n''} P_{n''} = S_n \delta_{n'n},$$  

(12)

for the dielectric polarizations $P_{n'}$ ($n' = 1, 2, \ldots, N$) with the random source located in the $n$th QW. Similar to Eq. (8) one has for the mean square

$$
|\langle S_n(\omega) \rangle|^2 = \frac{\varepsilon_h}{2\hbar q_0 a^2} |M_0|^2 G_0(\omega; n),
$$  

(13)

with $G_0(\omega; n)$ given by Eq. (3) where $f_k$ should be replaced by the 2D-exciton distribution function $f_{kn}$ in the $n$th QW. In order to analyze the role of frequency dependence of the generation rate $G_0(\omega; n)$ we consider a sum of two dimensionless integrals,

$$J(\omega) = \beta \int_{E_-}^\infty dE e^{-\beta E} [N(E + h\omega_0 - h\omega) + 1]$$  

$$+ \beta \int_0^{E_-} dE e^{-\beta E} N(h\omega_0 - E - h\omega_0),$$

related to the terms in Eq. (3) due to acoustic-phonon emission and absorption, respectively. The integral $J(\omega)$ can be used for the analysis of $G_0(\omega; n)$ assuming the matrix element of exciton-phonon scattering, $V_Q$, to be independent of the phonon wave vector. However one has to bear in mind that this matrix element vanishes at small $\mathbf{Q}$ and the occupation number $N(E + h\omega_0 - h\omega)$ diverges as $E$ tends to $h(\omega - \omega_0)$. These two complications can approximately be avoided by introducing in the above integrals the limits $E_+ = \max[0, h(\omega - \omega_0) + \delta]$ and $E_- = \max[0, h(\omega - \omega_0) - \delta]$, where $\delta$ is a fixed positive energy. The integration results in

$$J(\omega) = \begin{cases} e^{-\beta(h(\omega - \omega_0))} j_1 & \text{for } \omega - \omega_0 \gg \delta, \\ j_2 & \text{for } \omega_0 - \omega \gg \delta, \end{cases}$$  

(14)

where $j_1, j_2$ are slowly varying functions of $\omega$.

For simplicity, in the following we present the PL spectra calculated neglecting the frequency dependence of the generation rate $G_0(\omega; n)$. One should bear in mind that this dependence can be taken into account by multiplying the spectra by a factor (Eq. (14)) or by a more complicated approximation of the $\omega$-dependent function defined according to Eq. (3). Simultaneously, a frequency dependence of $\Gamma$ has to be taken into consideration according to

$$2\Gamma(\omega) = \frac{2\pi}{\hbar} \sum_{kQ} |F_Q|^2 [\delta_{k_0} - \delta_{k_0 - k_1}]$$  

$$+ (N_Q + 1) \delta(\omega - E_{k_1} - \hbar Q) \delta_{k_1},$$

The electric field $E^{(n)}$ can be related to the source $S_n$ by the discrete Green’s function $G_{n,n}$ of the system as follows

$$E^{(n)} = \left( \omega_0 - \omega - i\Gamma \right) G_{n,n} - \delta_{1n} S_n.$$  

(15)

and can be presented in the form

$$G_{n,n} = P_{n} e^{i K d / n} + P_{n} e^{-i K d / n},$$

where $K$ is the wave vector of an exciton polariton in an infinite QW structure, it satisfies the dispersion equation

$$\cos Kd = \cos qd + \frac{\Gamma_0}{\omega - \omega_0 + i\Gamma} \sin qd.$$  

(16)

$P_0, P$ determine the Green’s function for an infinite QW structure and are given by

$$P = \frac{i\Gamma_0 \sin qd}{(\omega_0 - \omega - i\Gamma)^2 \sin Kd},$$

$$P_0 = P + \frac{1}{\omega_0 - \omega - i\Gamma},$$

$P_{\pm}$ result from the internal reflection of exciton polaritons from the outermost barriers,

$$P_{\pm} = r P \frac{e^{i \Phi_{\pm}} + r e^{i Kd(2N - n - 2)}}{1 - r^2 e^{2i Kd(N-1)}},$$

where

$$r = \frac{1 - e^{-i q - Kd}}{1 - e^{-i q + Kd}},$$

$$\Phi_{\pm} = Kd(n - 2), \quad \Phi_{-} = Kd(2N - n).$$
For structures with the dielectric contrast one can use the equations [6]

\[ r_m = \frac{r_1 \sin(mKd)}{\sin(mKd) - t_1 \sin((m-1)Kd)}, \]
\[ t_m = \frac{t_1 \sin(Kd)}{\sin(mKd) - t_1 \sin((m-1)Kd)}. \]  

(17)

Here \( K \) is a function of \( \omega \) satisfying the dispersion equation for exciton polaritons in an infinite MQW structure, reflection and transmission coefficients from a single QW, \( r_1 \) and \( t_1 \), can be found in Ref. [6]. For the structure containing \( N \) QWs, a semi-infinite back barrier and a front barrier of the thickness \( b' \) between the first-QW left interface and vacuum, the expression for \( E^{(o)} \) is multiplied by the factor \( \tau / (1 - e^{i\rho} p_x) \), where \( \tau = 2n_b/(n_b + 1) \), \( \rho = (n_b - 1)/(n_b + 1) \) and \( \phi = 2b'/(b'2) \omega n_b/c \).

Fig. 1 illustrates the PL spectra calculated for a MQW structure without the dielectric contrast. The structure parameters are indicated in the caption. For simplicity, we set \( \langle |S_d(\omega)|^2 \rangle = 1 \). The arrows indicate positions of real parts of eigenfrequencies for four exciton–polariton modes in the \( N \)-QW structure. The labels \( j = N-2 \), \( N-3 \) and \( N-4 \) correspond to three polaritons with \( \text{Re} \{ \omega \} \) satisfying Eq. (16) with \( K_j = (nj/Nd) \). Fig. 2 shows the effect of the dielectric contrast mismatch on the PL spectral shape. Effects of the detuning from the Bragg condition and the increasing number of QWs are demonstrated in Figs. 3 and 4, respectively.

4. Relation between absorption and PL spectra

We define the absorbance \( A \) in an \( N \)-QW structure with semi-infinite left- and rightmost barriers as \( 1 - |r_N|^2 - |t_N|^2 \). Using the representation of \( r_N \), \( t_N \) in the form of Eqs. (15) and (18) in Ref. [7] we derive the following expression for the absorbance:

\[ A(\omega) = 1 - \frac{1}{2} \prod_{j=1}^{N-2} (\omega_j - \omega_j^2 + (\Gamma_j - \Gamma_j^2)(\Gamma_j + \Gamma_j^2)), \]  

(18)

and \( \omega_j = i(\Gamma_j + \Gamma) \) are the eigenfrequencies of exciton polaritons in the \( N \)-QW structure \( j = 1, 2, \ldots, N \). In the limit of small nonradiative damping the exact Eq. (18) reduces to

\[ A(\omega) = \sum_j \frac{2\Gamma_j}{(\omega - \omega_j)^2 + \Gamma_j^2}. \]  

(19)
of the packet. The flux of photons transmitted from the right-hand medium into vacuum via the multi-layered system equals

\[
\frac{\hbar \omega}{n_b} \frac{C}{N(h\omega)T'}(\omega) \frac{dq_x dq_y dq_z}{(2\pi)^3} = \frac{\hbar \omega N(h\omega)T'(\omega) d\omega dq_x dq_y}{(2\pi)^3},
\]

where \( T'(\omega) \) is the transmittance, i.e., the fraction of photon flux that passes through the system from the right to the left, and the photon dispersion \( \omega = cq/n_b \) in the medium of the refractive index \( n_b \) is taken into account. The PL spectral intensity, \( I_d(\omega) \), emitted by the multi-layered system into vacuum in the state of equilibrium is defined as

\[
I_d(\omega) d\omega \frac{dq_x dq_y}{(2\pi)^3}.
\]

Since in equilibrium the photon flux should be zero we obtain

\[
\hbar \omega N(h\omega)[1 - R(\omega) - T'(\omega)] = I_d(\omega).
\] (21)

Due to the time inversion symmetry \( T'(\omega) \) must coincide with the system transmittance from the left to the right. Therefore, Eq. (21) can be rewritten in the form

\[
I_d(\omega) = \hbar \omega N(h\omega) A(\omega),
\] (22)

relating the absorbance \( A(\omega) \) with the emission spectral intensity \( I_d(\omega) \) in accordance with Kirchhoff's law of thermal radiation. If the PL arises due to the indirect phonon-assisted emission of 2D excitons and the energy distribution of nonequilibrium excitons is characterized by the phonon temperature \( T \), i.e. if \( f_k = \exp[\beta(\mu - E_k)] \), where \( \mu \) is the exciton chemical potential, one can write instead of Eq. (22)

\[
I(\omega) = e^{\beta \hbar \omega} N(h\omega) A(\omega),
\] (23)

where \( I(\omega) \) is the spectral intensity of spontaneous emission of photoexcited multi-layered system. In equilibrium \( \mu = 0 \) and Eq. (23) reduces to Eq. (22). If the frequency interval under consideration is narrow as compared with the thermal energy \( k_b T \) one can set \( I(\omega) \propto A(\omega) \).

Note that an equation relating the PL spectrum with the absorption spectrum multiplied by a Boltzmann factor was obtained in Ref. [9] for exciton–polaritons in a quantum microcavity by introducing the polariton density of states and considering the PL as a direct transmission of cavity polaritons into vacuum. However, this kind of derivation presents difficulties for exciton–polaritons in finite close-to-Bragg MQWs, that are open systems characterized by short polariton radiative lifetimes, in which case the definition of the polariton density of states is problematic [10]. On the other hand, Eq. (22) derived here by using the requirement of zero for the photon flux at thermal equilibrium presupposes no additional assumptions and does not require a definition of polariton density of states. Moreover, Eq. (23) is derived for indirect exciton emission under the single assumption of Boltzmann distribution of photoexcited excitons outside the radiative circle.
The general relation between absorption and emission can be explicitly derived for MQW structure under a simplifying assumption of thin QWs. In this case using definition of absorbance given by Eq. (20) one can derive an expression for $A(\omega)$ in the following form:

$$A(\omega) = \frac{2\Gamma_0 \Gamma}{(\omega - \omega_0)^2 + \Gamma^2} \sum_{n=1}^{N} |E_n|^2,$$

(24)

where $|E_n|^2$ is the magnitude of the field at the center of $n$th QW calculated with scattering boundary conditions. This field, therefore, is different from the field entering Eq. (9) for the emission intensity. In order to relate this expression to the luminescence spectra we have to rewrite it in terms of the reflection and transmission coefficients. It can be shown that if the field behind the right boundary of the structure is $t_N$, then one has at the right boundary of $n$th QW

$$E_n = t_N \left( e^{-iqd/2} + r_{N-n} e^{iqd/2} \right).$$

Substituting this expression into Eq. (24) and comparing the results with Eq. (11) we finally find the following relationship between absorption and emission spectra

$$I(\omega) = \frac{e\hbar}{2\pi c^2} \frac{\langle |S_n|^2 \rangle}{2\scriptstyle{\text{Re}} \Gamma} A(\omega),$$

(25)

where $\langle |S_n|^2 \rangle$ is assumed to be independent of $n$. For a structure with a cap layer of finite thickness, light emission into vacuum and absorption under incidence from vacuum, the relation coefficient in Eq. (25) is multiplied by the refractive index $n_b$. The calculation of the emission spectra for frequency-independent $\langle |S_n|^2 \rangle$, or $G(\omega; n)$, and the absorbance spectra for frequency-independent $\Gamma$ completely confirms this relation.

5. Conclusion

In conclusion, we have developed a theory of PL for multiple QWs and established relation between their PL and absorbance spectra. The PL spectral shape is quite sensitive to the geometrical parameters of the structure, namely, the period $d$, the number of QWs and the cap-layer thickness, as well as the ratio of the exciton radiative and nonradiative damping rates. A quantitative comparison with experiment [4,5] needs a thorough fitting of these parameters and, probably, allowance for inhomogeneous broadening.

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