

Broadening and shift of excitonic resonances in quantum wells with adiabatic interface roughness

N. Atenco-Analco, F. Pérez-Rodríguez
*Instituto de Física, Universidad Autónoma de Puebla,
 Apartado Postal J-48, Puebla, Pue. 72570, México*

N. M. Makarov
*Instituto de Ciencias, Universidad Autónoma de Puebla,
 Priv. 17 Norte No 3417, Puebla, Pue. 72050, México*

The relaxation frequency and shift of exciton levels for a quantum well with rough surfaces are calculated in the adiabatic regime when the correlation radius of roughness is much larger than exciton radius. Unlike the case of a thin film, in a quantum well such characteristics of exciton resonances are attributed to the individual interaction of the electron and hole with the irregular interfaces. With applying the self-consistent Green's function method, we find the dependencies of the resonant broadening and shift upon the parameters of the quantum well and its disordered interfaces. Thanks to the self-consistency, namely to the action of the relaxation frequency and shift on themselves, exciton resonances can be either sharp and asymmetric, or broad and symmetric even in the adiabatic regime. However, within this regime and for realistic values of the correlation radius, the resonances turn out to be typically broad and symmetric.

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1. Introduction

It is commonly believed now that in semiconductor nanostructures the exciton relaxation, which determines the broadening and shift of excitonic resonances, is mainly due to the inherent interface disorder (see, for example, Ref. [1] and references therein). Therefore, the investigation of the exciton-surface scattering is a hot topic of modern optics of quantum-well structures.

An exciton in a bulk (massive) semiconductor is something similar to the hydrogen atom. It is a pair of an electron and a hole coupled by the Coulomb attraction. The motion of its center-of-mass is free while the relative electron-hole motion is quantized by the Coulomb interaction. As a two-particle object, an exciton is defined by the following parameters: typical size a_0 given by the exciton Bohr radius ($a_0 = \hbar^2 \epsilon / e^2 \mu$), the reduced mass of the relative electron-hole motion in the Coulomb potential $\mu = m_e m_h / M$, the electron m_e and hole m_h effective masses, the total mass of the center-of-mass $M = m_e + m_h$, and dielectric constant ϵ of the semiconductor. An important characteristic of exciton is the so-called resonance frequency ω_{res} associated with an excitonic quantum state. Usually, in semiconductors this frequency falls into the visible or infrared spectral range. Therefore, exciton plays a significant role in optical properties of semiconductors. When the light frequency ω becomes equal to the exciton frequency, the excitonic resonance is observed in reflection and absorption spectra of semiconductors.

The effect of surface roughness on spectral and optical properties of excitons has been studied both experimentally and theoretically for over thirty years [1-3]. Theoretical

description of exciton-surface interaction is a rather complex problem because from the very beginning it is a two-particle problem. So, even near a flat surface, exciton dynamics changes drastically due to the finite exciton size. For this reason, optical properties of excitons in massive rough-bounded systems have been mostly studied phenomenologically. These models are based on the reduced Schrödinger equation for the exciton center-of-mass in an effective disordered potential [3].

Now, let us look at the exciton in quantum wells. In this regime, the exciton radius a_0 ($\sim 10 - 100$ angstroms for II-VI and III-V semiconductors) is larger than the average well width d . At the same time, d far exceeds the root-mean-square roughness height $\sigma \sim 1 - 2$ angstroms [1],

$$\sigma \ll d < a_0. \quad (1.1)$$

Due to the right condition, the Coulomb attraction is suppressed in the direction perpendicular to the well. Therefore, only in the plane of the well an electron and a hole can form an exciton whose internal motion is quantized by the two-dimensional Coulomb attraction.

Along the transverse direction (the growth direction of the well), the individual electron and hole motions are quantized by multiple reflections from the walls. So, in analyzing exciton-surface scattering, we should consider the *individual* interactions of the electron and hole with the rough interface instead of considering the scattering of their center-of-mass.

Thus, in quantum wells the excitonic state is substantially different from that in a bulk semiconductor. Therefore, to solve properly the exciton-surface scattering problem for quantum well, we cannot apply old well-developed methods based on the exciton center-of mass model.

For surface-disordered systems with in-plane two-particle Coulomb coupling we have chosen and generalized the self-consistent adiabatic Green's function method. This method is very suitable for four reasons. First, it is based on the original microscopic excitonic Hamiltonian. Second, the exciton-surface scattering frequency and spectrum shift are introduced in a very natural way. Third, this method is self-consistent and, therefore, takes into account the inherent action of the exciton-surface scattering on itself. Only due to the self-consistency we can find the complete line-shape of the excitonic resonance and analyze its dynamics within the whole spectral range. At last, the adiabaticity implies that the surface scattering potential will not vary over the scale a_0 of the relative electron-hole motion in the 2D Coulomb potential as well as over the scale d of their individual transverse motion. This means that the average length of the surface defects R_c should be much greater than the excitonic radius a_0 , which, due to Eq. (1.1), is larger than the well width d ,

$$d < a_0 < R_c. \quad (1.2)$$

The adiabatic model of exciton-surface scattering is widely used in the majority of the theoretical investigations [4,5]. In spite of its restriction of applicability (1.2), this model allows to obtain and analyze a relatively simple equation for the broadening and shift of excitonic resonances in a semiconductor quantum well.

2. Problem formulation. Average green's function

We consider an excitonic quantum well of average width d . The relief $z = \xi(\vec{r})$ of the lower interface is assumed to be randomly rough while the upper one $z = d$ is, for simplicity, flat. In other words, the quantum well is confined within the region

$$\xi(\vec{r}) \leq z \leq d. \quad (2.1)$$

Here z is the transverse to the well plane coordinate. The surface roughness is described by a random function $\xi(\vec{r})$ of the in-plane position vector \vec{r} with standard statistical properties [6]

$$\langle \xi(\vec{r}) \rangle = 0, \quad \langle \xi(\vec{r})\xi(\vec{r}') \rangle = \sigma^2 W(|\vec{r} - \vec{r}'|). \quad (2.2)$$

The angular brackets stand for statistical averaging over realizations of $\xi(\vec{r})$, σ is the root-mean-square (r.m.s.) roughness height, $W(|\vec{r}|)$ is the correlation coefficient with the unit maximal value, $W(0) = 1$, and a scale of decrease R_c (the correlation radius).

Due to the definition (1.1) of a quantum well, the excitonic Hamiltonian can be suitably written in the following form

$$\begin{aligned} \hat{H} = & E_{gap} - \frac{\hbar^2}{2M} \frac{\partial^2}{\partial \vec{R}^2} - \frac{\hbar^2}{2\mu} \frac{\partial^2}{\partial \vec{\rho}^2} - \frac{e^2}{\epsilon\rho} - \frac{\hbar^2}{2m_{e,z}} \frac{\partial^2}{\partial z_e^2} \\ & - \frac{\hbar^2}{2m_{h,z}} \frac{\partial^2}{\partial z_h^2} + U_e(z_e, \vec{r}_e) + U_h(z_h, \vec{r}_h) - i\hbar\nu_0 \end{aligned} \quad (2.3)$$

Here, E_{gap} is the energy gap between the conduction and valence bands of the semiconductor. The second term in Eq. (2.3) describes the kinetic energy of the exciton center-of-mass with in-plane total mass $M = m_{e\parallel} + m_{h\parallel}$ and in-plane radius vector \vec{R} . The third term specifies the kinetic energy of the relative electron-hole motion, which is characterized by the in-plane reduced mass $\mu = m_{e\parallel}m_{h\parallel}/M$ and the internal in-plane vector $\vec{\rho}$. Two-dimensional Coulomb potential is given by the fourth term with ϵ being the dielectric constant of semiconductor. The fifth and sixth terms are responsible for the individual transverse motion of electron or hole, respectively (z_e or z_h are the electron or hole transverse coordinates). The quantities $U_e(z_e, \vec{r}_e)$ and $U_h(z_h, \vec{r}_h)$ are the confining potentials of the quantum well for the electron and the hole. The relations connecting the introduced in-plane vectors are

$$\vec{r}_{e,h} = \vec{R} \pm \mu\vec{\rho}/m_{e\parallel,h\parallel}, \quad \vec{\rho} = \vec{r}_e - \vec{r}_h \quad (2.4)$$

In the Hamiltonian (2.3) we have introduced a homogeneous exciton-bulk damping ν_0 to take into account its effect on the exciton-surface scattering.

In order to analyze properly excitonic states in a surface-disordered quantum well, we shall derive the retarded Green's function of the excitonic Hamiltonian (2.3), which obeys the equation

$$(\hbar\omega - \hat{H})G = \hat{I}. \quad (2.5)$$

Here $\hbar\omega$ is the energy, \hat{I} is the unite-operator, which in the exciton coordinate space is represented by Dirac δ -functions.

For the convenience of subsequent averaging, this differential equation can be reduced to the integral Dyson-type equation that relates the perturbed by the surface disorder Green's function G to the Green's function G_0 for the ideal well with flat interfaces (at $\xi(\vec{r}) = 0$). In line with the general theory of the Green's function [7], the symbolic form of the equation is

$$G = G_0 + G_0(\hat{V}_e + \hat{V}_h)G. \quad (2.6)$$

It contains the sum of the electron-surface \hat{V}_e and hole-surface \hat{V}_h scattering operators, whose kernels are represented as the difference between the confining potentials of the corrugated $z = \xi(\vec{r})$ and ideally flat $z = 0$ interfaces,

$$\begin{aligned}\hat{V}_{e,h} &= U_{e,h} \left[\Theta(\xi(\vec{r}_{e,h}) - z_{e,h}) - \Theta(-z_{e,h}) \right] \\ &= U_{e,h} \delta(z_{e,h}) \xi(\vec{R})\end{aligned}\quad (2.7)$$

Here $\Theta(x)$ is the Heaviside unit-step function, and $U_{e,h}$ is the finite height of the potential barrier for the electron (e) and the hole (h). Note that the roughness profile function $\xi(\vec{r}_{e,h})$ has the variation scale R_c , and hence, within the adiabatic regime (1.2), does not vary over the scale $\rho \sim a_0/2$ of the rapid relative electron-hole motion in the 2D Coulomb potential. Therefore, we have neglected the dependence of this function on the internal vector $\vec{\rho}$ in the expressions (2.7) for the exciton-surface scattering operators V_e and V_h .

Normally, the spatial resolution of an experiment (the size of the light spot) is much larger than the correlation radius R_c of surface defects [8]. Therefore, all observed quantities are statistically averaged over many independent regions of rough surface. This fact allows us to find the averaged Green's function of our problem. We average the exact equation (2.6) by applying the technique proposed in Ref. [9]. As a result, the integral equation for the average Green's function \bar{G} within the self-consistent Born approximation can be symbolically written as

$$\bar{G} = G_0 + G_0 \left\langle (\hat{V}_e + \hat{V}_h) \bar{G} (\hat{V}_e + \hat{V}_h) \right\rangle \bar{G}. \quad (2.8)$$

Note that within the ordinary Born approximation this equation contains G_0 between the interaction operators, while in the self-consistent approach G_0 is replaced by \bar{G} .

In order to solve the Dyson equation (2.8) we should take into account the following facts: (i) Statistical homogeneity and isotropy of the problem with respect to the center-of-mass radius vector \vec{R} , (ii) Adiabaticity of the surface scattering potentials in comparison with the relative electron-hole motion in the 2D Coulomb potential as well as with their individual transverse motion. In this case, we obtain

$$\begin{aligned}\hbar\bar{G} &= \sum_{n=0}^{\infty} \sum_{m=-n}^n \Phi_{nm}(\vec{\rho}) \Phi_{nm}^*(\vec{\rho}') \\ &\sum_{n_e=1}^{N_e} \Psi_{n_e}^{(e)}(z_e) \Psi_{n_e}^{(e)*}(z_e') \sum_{n_h=1}^{N_h} \Psi_{n_h}^{(h)}(z_h) \Psi_{n_h}^{(h)*}(z_h') \\ &\int_{-\infty}^{\infty} \frac{d\vec{k}}{(2\pi)^2} \frac{\exp[i\vec{k}(\vec{R} - \vec{R}')] }{\omega - \omega_{res} - (\hbar k^2 / 2M) + i\nu_0 - M_{n_e n_h}(n)}\end{aligned}\quad (2.9)$$

Here $\Phi_{nm}(\vec{\rho})$ are the eigenfunctions (2D Coulomb modes) for the exciton intrinsic motion in the 2D Coulomb potential with n being radial (energy) and m being azimuth quantum numbers; $\Psi_{n_e}^{(e)}(z_e)$ and $\Psi_{n_h}^{(h)}(z_h)$ are the confinement eigenfunctions with quantum numbers n_e and

n_h for the individual transverse motion of electron (e) and hole (h), the integer N_e (N_h) is the total number of transverse quantum electron (hole) levels in the quantum well of finite depth $U_{e,h}$. The asterisk "*" stands for the complex conjugation. The symbol $\omega_{res} = \omega_{n_e n_h}(n)$ denotes the frequency of exciton eigenstate; the quantity $M_{n_e n_h}(n)$ enters Eq. (2.9) as the self-energy. Because of the self-consistency, the self-energy is governed by the equation that directly follows from the Dyson equation (2.8) and within the adiabatic approximation takes the form

$$\begin{aligned}M_{n_e n_h}(n) &= \frac{\sigma^2 M}{\hbar^3} \left[U_e |\Psi_{n_e}^{(e)}(0)|^2 + U_h |\Psi_{n_h}^{(h)}(0)|^2 \right]^2 \\ &\int_0^{\infty} d\omega_t \frac{\tilde{W}(\sqrt{2M\omega_t/\hbar})}{2\pi \omega - \omega_{res} - \omega_t + i\nu_0 - M_{n_e n_h}(n)}\end{aligned}\quad (2.10)$$

In this equation we have introduced the Fourier transform $\tilde{W}(k)$ of the binary correlator $W(|\vec{r}|)$,

$$\tilde{W}(k) = \int_{-\infty}^{\infty} d\vec{r} \exp(-i\vec{k}\vec{r}) W(\vec{r}). \quad (2.11)$$

As is known, the frequency of the unperturbed excitonic eigenstate $\omega_{res} = \omega_{n_e n_h}(n)$ is degenerated over the discrete azimuth quantum number m . From Eq. (2.10) one can see that the self-energy $M_{n_e n_h}(n)$ also turns out to be independent of m . Moreover, the self-energy does not contain the transitions between different excitonic states as it would take place in the general case. These facts are in full agreement with the general theory of the adiabatic perturbations [10]. Indeed, in the case of an adiabatic potential the discrete quantum numbers are conserved and only continuous quantum numbers may change due to scattering. In the problem of exciton-surface scattering such a quantum number is the in-plane exciton center-of-mass wave vector \vec{k} .

3. Scattering frequency and spectrum shift

As follows from the expression (2.9) for the average Green's function, the surface-induced complex correction to the unperturbed excitonic spectrum $\omega = \omega_{n_e n_h}(n)$ of a quantum well is determined by the self-energy $M_{n_e n_h}(n)$. Its real part is responsible for the real shift $\delta\omega_{res}$ of the excitonic resonance frequency while the imaginary part gives the exciton-surface scattering frequency ν ,

$$\delta\omega_{res} = \text{Re} M_{n_e n_h}(n), \quad \nu = -\text{Im} M_{n_e n_h}(n). \quad (3.1)$$

Therefore, to analyze these quantities we should solve the equation (2.10). One can see that this equation is rather

complicated. Nevertheless, it can be substantially simplified if we choose a step-wise model for the roughness power spectrum,

$$\tilde{W}(k) = 4\pi R_c^2 \Theta(1 - |\vec{k}| R_c). \quad (3.2)$$

Being very simple, the expression (3.2) possesses all general properties of the Fourier transform of the roughness binary correlator $W(|\vec{r}|)$ and consequently, allows us to perform a complete qualitative analysis of the resonance shift $\delta\omega_{res}$ and the scattering frequency ν . After substituting Eq. (3.2) into Eq. (2.10), the integral over ω_l is taken explicitly and we get the relatively simple and easily analyzable equation for the exciton-surface scattering frequency ν and the real spectrum shift $\delta\omega_{res}$,

$$\begin{aligned} -\delta\omega_{res} + i\nu &= \\ &= \frac{\nu_N^2}{\omega_W} \ln \left(1 - \frac{\omega_W}{\omega - \omega_{res} - \delta\omega_{res} + i(\nu_0 + \nu)} \right). \end{aligned} \quad (3.3)$$

Here, the normalization frequency ν_N is defined by

$$\nu_N = \frac{\sigma}{\hbar} \left[U_e |\Psi_{n_e}^{(e)}(0)|^2 + U_h |\Psi_{n_h}^{(h)}(0)|^2 \right]. \quad (3.4)$$

This constant specifies the effect of the finiteness of the potential barrier for a quantum well and the effect of the transverse electron and hole quantization on ν and $\delta\omega_{res}$. The characteristic scaling frequency ω_W of roughness power spectrum is given by

$$\omega_W = \hbar R_c^{-2} / 2M. \quad (3.5)$$

One can see from Eq. (3.3) that the line-shape of the excitonic resonance in the relaxation frequency ν and in the real spectrum shift $\delta\omega_{res}$ is determined by the relation between the scaling frequency ω_W of surface roughness and the total scattering frequency $\nu_0 + \nu$. Namely, the equation (3.3) displays two types of the excitonic resonance.

When the total scattering frequency is much less than the scaling frequency,

$$\nu_0 + \nu \ll \omega_W, \quad (3.6)$$

the excitonic resonance is *sharp and asymmetric*. The resonance shift $\delta\omega_{res}$ and broadening ν are described by the asymptotic

$$\delta\omega_{res} = -\frac{\nu_N^2}{\omega_W} \ln \left[\frac{\omega_W}{\sqrt{(\omega - \omega_{res} - \delta\omega_{res})^2 + (\nu_0 + \nu)^2}} \right] \quad (3.7)$$

$$\nu = \frac{\nu_N^2}{\omega_W} \left[\frac{\pi}{2} + \arctan \left(\frac{\omega - \omega_{res} - \delta\omega_{res}}{\nu_0 + \nu} \right) \right]. \quad (3.8)$$

At the resonance point $\omega = \omega_{res} + \delta\omega_{res}$, the quantity ν is equal to $\pi\nu_N^2 / 2\omega_W$ and reaches its maximal value $\pi\nu_N^2 / \omega_W \ll \nu_N$ to the right of the resonance where the resonance detuning $\omega - \omega_{res} - \delta\omega_{res}$ is positive. Also, the shift $\delta\omega_{res} \approx -2(\nu_N^2 / \omega_W) \ln(\omega_W / \nu_N)$ is negative at the resonance point. Thus, in this case, the resonance turns out to be moved to the left of the point $\omega = \omega_{res}$ by the value $\delta\omega_{res}$ and has an asymmetric line-shape with respect to the resonance point $\omega = \omega_{res} + \delta\omega_{res}$.

The *broad and symmetric* resonance corresponds to the condition when the total scattering frequency is much larger than the scaling frequency,

$$\omega_W \ll \nu_0 + \nu. \quad (3.9)$$

Here the line-shape of the resonance is described by the expressions

$$\delta\omega_{res} = (\omega - \omega_{res}) / 2, \quad (3.10)$$

$$\nu^2 + \delta\omega_{res}^2 = \nu_N^2, \quad (3.11)$$

and, therefore, becomes symmetric with respect to the resonance point $\omega = \omega_{res}$. At this point the surface scattering frequency ν approaches its maximum value $\nu_N \gg \nu_0$ and the spectrum shift $\delta\omega_{res}$ turns out to be zero.

As ω_W decreases (the correlation radius R_c increases) we pass from the sharp to the broad resonance and the resonant point is moved towards its unperturbed value $\omega = \omega_{res}$. Due to the self-consistency, with increasing R_c , not only ω_W is decreased, but also the resonance line-shape is substantially enhanced because of its self-action. That is why the transition from the sharp to the broad resonance is quite rapid. For realistic values of the parameters of a quantum well, the resonance looks broad beginning from $R_c \sim a_0$. Therefore, at the adiabatic surface roughness (1.2) the broad and symmetric resonance is more typical than the sharp and asymmetric one.

4. Numerical results

In this section we present graphs calculated from Eq. (3.3) for the frequency dependencies of the broadening ν and shift $\delta\omega_{res}$ of the ground-state *hh*-exciton resonance ($n_e = 1$, $n_h = 1$, $n = 0$) for a $\text{Zn}_{0.7}\text{Cd}_{0.3}\text{Se}$ quantum well of

thickness $d = 30 \text{ \AA}$ with $\text{ZnS}_{0.06}\text{Se}_{0.94}$ barriers [Fig. 1]. The material parameters used are (see Ref. [11]): a band gap of the well material $E_{gap} = 2.438 \text{ eV}$, a conduction-band offset of 202 meV (with strain), a valence-band offset of 80 meV (with strain), the effective electron mass $m_{e,z} = m_{e\parallel} = 0.14m_0$ (here m_0 is the free electron mass), the in-plane heavy-hole mass $m_{hh,\parallel} = 0.24m_0$, the *z*-axis

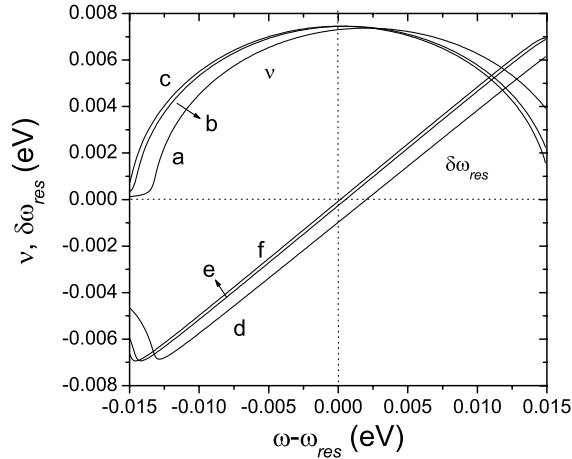


Figure 1. Frequency dependence of the surface-induced inhomogeneous damping ν and shift $\delta\omega_{res}$ for the ground-state hh -exciton resonance of a $Zn_{0.7}Cd_{0.3}Se$ quantum well of thickness $d = 30 \text{ \AA}$ with $ZnS_{0.06}Se_{0.94}$ barriers. Curves were calculated with r.m.s. roughness height $\sigma = 2 \text{ \AA}$ and correlation radius $R_c = 50$ (a,d), 100 (b,e), and 500 \AA (c,f).

heavy-hole mass $m_{hh,z} = 0.84m_0$ (in the calculation all the masses are assumed to be equal for the well and the barrier), $\varepsilon = 8.7$. Other parameters used are: a homogeneous damping factor $\hbar\nu_0 = 0.02 \text{ meV}$, a small r.m.s. roughness height $\sigma = 2 \text{ \AA}$, and a sufficiently large correlation radius $R_c = 50$ (curves a,d), 100 (curves b,e), and 500 angstroms (curves c,f).

Curves a, b, and c of Fig.1 show that the line-shape of ν is quite broad, having a maximum near the resonance frequency ω_{res} , and becomes symmetric as the correlation radius R_c is increased. On the other hand, the shift $\delta\omega_{res}$ vanishes near ω_{res} and has different signs on both sides of the exciton resonance (curves d, e, and f). The calculated frequency dependencies for ν and $\delta\omega_{res}$ at the largest used value of R_c ($R_c = 500$ angstroms, curves c and f) are well described by the asymptotic (3.10) and (3.11) for the case of broad and symmetric resonance (3.9).

5. Summary

Finally, we present a summary of our research.

Applying the self-consistent adiabatic Green's function method, we have derived and analyzed the exciton-surface scattering frequency ν and the real spectrum shift $\delta\omega_{res}$ for an interface-disordered quantum well. The main advantage of this approach is that it allows for revealing the two types of excitonic resonance as well as the transition between them.

The type of resonance and its line-shape are determined by the relation between the total scattering frequency

$\nu_0 + \nu$ and the scaling frequency ω_W . The line-shape is asymmetric in the case of the sharp resonance, and becomes symmetric for the broad resonance. Within the adiabatic regime the broad and symmetric resonance is more common.

The predictions of the adiabatic self-consistent approach were illustrated by calculating the relaxation frequency ν and the shift $\delta\omega_{res}$ for the resonance of the ground-state hh -exciton in $Zn_{0.7}Cd_{0.3}Se$ quantum wells with $ZnS_{0.06}Se_{0.94}$ barriers.

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