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Surface relaxation frequency of ground-state exciton in quantum wells

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Abstract

We derive and analyze the relaxation frequency ν of exciton in a quantum well using the self-consistent Green's function method. The exciton relaxation is caused by the *individual* electron and hole scattering from the randomly-rough well interface. We reveal two types of ground-state exciton resonance and obtain the criteria for the transition from the asymmetric (sharp) resonance to the symmetric (broad) one. The dependence of the excitron–surface relaxation on the microscopic parameters of the interface defects, the average well width d and on the exciton characteristics is analyzed analytically. Specifically, in the case of sharp resonance the frequency $\nu \propto d^{-6}$, whereas for broad resonance $\nu \propto d^{-3}$. Moreover, ν is proportional to the ratio of the total exciton mass M over the squared reduced mass μ^2 ($\nu \propto M/\mu^2$).

Keywords: Excitons; Disordered systems; Quantum wells

1. Introduction

An important characteristic of excitons is their relaxation frequency which determines the broadening of exciton resonance. This frequency is composed of two parts with different physical nature. The first part describes the homogeneous broadening ν_0, while the second one is the so-called inhomogeneous relaxation frequency ν, which arises, for example, from disorder in the semiconductor. In a quantum well with inherent interface disorder, the inhomogeneous frequency ν is principally caused by the exciton–surface scattering and greatly exceeds ν_0. Therefore, the calculation and the study of ν is a relevant problem in the physics of semiconductor quantum well structures.

In our investigation, we suggest a theory of the exciton–surface scattering based on the following three points.

1. The concept of a quantum well implies the exciton radius a_0 associated with the Coulomb interaction to be much larger than the average well width d that, evidently, should far exceed the r.m.s. roughness height ζ,

$$\zeta \ll d \ll a_0. \tag{1}$$

Due to the right condition, the Coulomb interaction is suppressed in the direction perpendicular to the well plane. Consequently, in analyzing surface scattering of exciton we should treat the individual electron–surface and hole–surface interaction separately.
2. In contrast to previous studies, we do not impose any a priori restriction on the roughness correlation length R_c. In particular, the developed approach is valid for an arbitrary relation between the exciton radius a_0 and the correlation length R_c.
3. As we demonstrate here, the appropriate exciton–surface scattering theory should take into account the inherent action of the exciton scattering on itself. Therefore, in order to manage the exciton–surface scattering problem we should apply and generalize to the case of two-particle motion the self-consistent Green's function method [1].

The main result of our theory is a relatively simple equation for the exciton–surface scattering frequency ν, which is easily analyzable and displays explicitly dependence of ν on the parameters of the exciton, interface roughness, and exciton–resonance detuning.

2. Problem statement

We consider a quantum well of average width d, being confined within the region

$$\xi(\vec{r}_{e,h}) \le z_{e,h} \le d, \tag{2}$$

where coordinates $z_{e,h}$ specify a transverse to the well electron or hole motion respectively, $\xi(\vec{r})$ is a random function of the longitudinal (electron or hole) 2D position vector $\vec{r}$

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characterized by

$$\langle \xi(\vec{r}) \rangle = 0, \qquad \langle \xi(\vec{r})\xi(\vec{r}\,') \rangle = \zeta^2 \tilde{W}(|\vec{r} - \vec{r}\,'|). \tag{3}$$

The angle brackets stand for statistical averaging over the ensemble of the functions $\xi(\vec{r})$. The binary coefficient of correlation $\tilde{W}(|\vec{r}|)$ has the unit amplitude $\tilde{W}(0) = 1$, and the typical scale R_c of monotonous decrease. So, our consideration supposes the lower boundary of the well to be randomly rough while the upper one is, for simplicity, flat. However, such a system is physically equivalent to a well with both boundaries being rough, statistically identical, and not intercorrelated. Moreover, our method can be easily generalized to systems with arbitrary statistical properties of both quantum well surfaces.

Under the condition (1) the exciton Hamilton $\hat{H}_{Q2D}$ can be suitably written as

$$\hat{H}_{Q2D} = E_{gap} - \frac{\hbar^2}{2M}\frac{\partial^2}{\partial \vec{R}^2} - \frac{\hbar^2}{2\mu}\frac{\partial^2}{\partial \vec{\rho}^2} - \frac{e^2}{\epsilon_0 \rho} - \frac{\hbar^2}{2m_e}\frac{\partial^2}{\partial z_e^2} - \frac{\hbar^2}{2m_h}\frac{\partial^2}{\partial z_h^2} - i\hbar\nu_0. \tag{4}$$

Here, E_{gap} is the energy gap between the conduction and valence bands, ϵ_0 is the dielectric constant of the excitonic medium, m_e (m_h) is the electron (hole) effective mass. The exciton center-of-mass is described by the total mass $M = m_e + m_h$ and the in-plane 2D radius vector $\vec{R}$. The relative electron–hole motion is specified by the reduced mass $\mu = m_e m_h/M$ and the longitudinal 2D vector $\vec{\rho}(\vec{r}_e = \vec{R} + \mu\vec{\rho}/m_e, \vec{r}_h = \vec{R} - \mu\vec{\rho}/m_h)$. In the Hamiltonian (4) we have introduced a homogeneous exciton–bulk damping ν_0 to take into account its effect on the exciton–surface scattering.

To analyze exciton states in a quantum well, the retarded Green's function $G(\vec{R}, \vec{R}'; \vec{\rho}, \vec{\rho}\,'; z_e, z_e'; z_h, z_h')$ of the Hamiltonian (4) will be derived. We assume that the interband separation inside the well is sufficiently smaller than outside the well so that both well walls are absolutely soft. Therefore, the Green function vanishes at the well interfaces $z_{e,h} = \xi(\vec{r}_{e,h})$ and $z_{e,h} = d$.

The exact integral equation for G can be obtained by making use of Green's theorem for a 6D space. It takes the form of a Dyson-type equation, relates G to the Green function G_0 for the ideal (smooth) well, having $\xi(\vec{r}) \equiv 0$, and contains a sum of the effective electron–surface $\hat{V}_e$ and hole–surface $\hat{V}_h$ scattering operators,

$$\hat{V}_{e,h}\left(\vec{R}, \vec{\rho}, z_e, z_h\right) = \frac{\hbar^2}{2m_{e,h}}\Theta\left[z_{h,e} - \xi\left(\vec{R} \mp \frac{\mu}{m_{h,e}}\vec{\rho}\right)\right] \times \Theta(d - z_{h,e})\delta\left[z_{e,h} - \xi\left(\vec{R} \pm \frac{\mu}{m_{e,h}}\vec{\rho}\right)\right] \times \left[\frac{\partial}{\partial z_{e,h}} - \frac{\partial}{\partial \vec{R}}\xi\left(\vec{R} \pm \frac{\mu}{m_{h,e}}\vec{\rho}\right)\left(\frac{\mu}{m_{h,e}}\frac{\partial}{\partial \vec{R}} \pm \frac{\partial}{\partial \vec{\rho}}\right)\right], \tag{5}$$

where $\Theta(x)$ is the Heaviside unit-step function and $\delta(x)$ is the Dirac delta-function. We have averaged the equation for G by applying the technique proposed in Ref. [2]. The resulting integral equation for the averaged Green's function $\bar{G}$ within the self-consistent Born approximation can be symbolically written as

$$\bar{G} = G_0 + G_0\langle(\hat{V}_e + \hat{V}_h)\bar{G}(\hat{V}_e + \hat{V}_h)\rangle\bar{G}. \tag{6}$$

3. Exciton–surface scattering frequency

We restrict our further consideration to the most actual and widely-analyzed case of the exciton ground state in two-dimensional quantum well when there is only one electron–hole mode of their transverse quantization and the exciton can occupy only the first 2D Coulomb level, i.e.

$$\omega - \omega_0 < \omega_1. \tag{7}$$

The frequency ω_0 of the exciton ground resonance and the distance ω_1 between ω_0 and the frequency of the next exciton resonance are given by

$$\omega_0 = \frac{E_{gap}}{\hbar} - \frac{\hbar}{2\mu}\left(\frac{2}{a_0}\right)^2 + \frac{\hbar}{2\mu}\left(\frac{\pi}{d}\right)^2, \tag{8}$$

$$\omega_1 = \min\left\{\frac{3\hbar}{2\max\{m_e, m_h\}}\left(\frac{\pi}{d}\right)^2; \frac{8}{9}\frac{\hbar}{2\mu}\left(\frac{2}{a_0}\right)^2\right\}. \tag{9}$$

Due to the basic condition (1) the frequency ω_0 is related to the quantized transverse electron–hole motion rather than to the in-plane Coulomb interaction, whereas the latter mainly contributes to ω_1.

Near the exciton ground resonance (7) we can express the perturbed by surface disorder average Green function as

$$\bar{G}\left(|\vec{R} - \vec{R}'|; \vec{\rho}, \vec{\rho}\,'; z_e, z_e'; z_h, z_h'\right) = \left(\frac{2}{d}\right)^2 \sin\left(\frac{\pi z_e}{d}\right) \times \sin\left(\frac{\pi z_e'}{d}\right)\sin\left(\frac{\pi z_h}{d}\right)\sin\left(\frac{\pi z_h'}{d}\right)\left(8/\pi a_0^2\right) \times \exp[-2(\rho + \rho')/a_0]\int_{-\infty}^{\infty}\frac{d\vec{k}_t}{(2\pi)^2} \times \frac{\exp\left[i\vec{k}_t(\vec{R} - \vec{R}')\right]}{\hbar[\omega - \omega_0 - (\hbar k_t^2/2M) + i\nu_0 + i\nu(k_t)]}. \tag{10}$$

Here, $\hbar\omega$ is the energy of the exciton, $a_0 = \hbar^2\epsilon_0/e^2\mu$. The quantity $\nu(k_t)$ denotes the exciton–surface scattering frequency. It satisfies an integral equation that directly follows from Eq. (6). As is known, in the case of normal incidence of light, the exciton–resonance broadening is determined by $\nu(k_t = 0)$. The equation for $\nu \equiv \nu(k_t = 0)$ takes the form

$$\frac{\nu}{\nu_l} = \frac{l}{\pi}\int_0^{\infty} d\omega_t \Delta_{\nu_0+\nu}(\omega_t)Q(\omega_t)\frac{W\left(\sqrt{2M\omega_t/\hbar}\right)}{W(0)}. \tag{11}$$

The integrand of Eq. (11) contains three functions. The sharp resonant function

$$\Delta_{\nu_0+\nu}(\omega_t) = \frac{\nu_0 + \nu}{(\omega - \omega_0 - \omega_t)^2 + (\nu_0 + \nu)^2} \tag{12}$$

with the typical variation scale $\nu_0 + \nu$. The exciton function

$$Q\left(\omega_t\right) = \left\{ \frac{\mu}{m_e}\left[1 + \frac{2M}{\hbar}\left(\frac{\mu}{m_e}\frac{a_0}{4}\right)^2 \omega_t\right]^{-3/2} + \frac{\mu}{m_h}\left[1 + \frac{2M}{\hbar}\left(\frac{\mu}{m_h}\frac{a_0}{4}\right)^2 \omega_t\right]^{-3/2}\right\}^2 \tag{13}$$

which reaches the largest value $Q(0) = 1$ at $\omega_t = 0$ and monotonously decreases with increasing ω_t over the scale

$$\omega_Q = \frac{\hbar}{2M}\left(\frac{\mu}{\min\{m_e, m_h\}}\frac{a_0}{4}\right)^{-2}. \tag{14}$$

Finally, the Fourier transform

$$W(k_t) = \int_{-\infty}^{\infty} d\vec{R} \exp\left(-i\vec{k}_t\vec{R}\right)\tilde{W}(|\vec{R}|) \tag{15}$$

of the correlation coefficient $\tilde{W}(|\vec{R}|)$, as a function of ω_t, has the scale of decrease

$$\omega_W = \hbar R_c^{-2}/2M. \tag{16}$$

The normalization constant ν_l determines the main dependence of the exciton–surface scattering frequency ν on the external parameters of the problem and is defined by

$$\nu_l = B\frac{\hbar(\pi/d)^2}{2\mu}\frac{M}{\mu}\left(\frac{\zeta}{d}\right)^2\left(\frac{\pi R_c}{d}\right)^2. \tag{17}$$

The positive number $B = W(0)/R_c^2$ depends neither on ζ nor on R_c, is determined by the form of the correlator $\tilde{W}(|\vec{R}|)$ and has a value of the order of unity.

Note that only the function (12) in Eq. (11) depends on the resonance detuning $\omega - \omega_0$. Therefore, the line-shape of $\nu(\omega - \omega_0)$ is mainly determined by the relation between the variation scales $\nu_0 + \nu$ and $\min\{\omega_W, \omega_Q\}$ of functions Δ and QW, respectively. At the same time, the ratio of R_c to a_0 and, hence, ω_W to ω_Q can be arbitrary.

4. Analysis and conclusion

The analysis of Eq. (11) leads to the following results.

Let the exciton ground resonance be sharp, i.e.

$$\nu_0 + \nu \ll \min\{\omega_W, \omega_Q\}. \tag{18}$$

In this case, ν is found from the transcendental equation

$$\frac{\nu}{\nu_l} = \frac{1}{2}\left[1 + \frac{2}{\pi}\arctan\left(\frac{\omega - \omega_0}{\nu_0 + \nu}\right)\right], \tag{19}$$

which is valid to the left of the resonance ($\omega - \omega_0 \leq 0$) and within its right vicinity $\left(0 \leq \omega - \omega_0 \ll \min\{\omega_W, \omega_Q\}\right)$. Here, the ratio ν/ν_l is a universal function of the variable $(\omega - \omega_0)/\nu_l$ and the parameter ν_0/ν_l.

On the right of the resonance, when $\omega - \omega_0 \gg \nu_0 + \nu$, the relaxation frequency ν is described by the explicit expression,

$$\frac{\nu}{\nu_l} = \frac{W\left(\sqrt{2M(\omega - \omega_0)/\hbar}\right)}{W(0)} Q(\omega - \omega_0). \tag{20}$$

Within the intermediate region $\nu_0 + \nu \ll \omega - \omega_0 \ll \min\{\omega_W, \omega_Q\}$ where $\nu \approx \nu_l$, the asymptotic (20) coincides with Eq. (19) in the order of value.

As the detuning $\omega - \omega_0$ increases, the surface scattering frequency ν also increases, takes the value $\nu = \nu_l/2$ at the resonance point $\omega = \omega_0$ and then approaches its largest value ν_l. At large frequencies, on the right of the resonance, the value of ν monotonously decreases in accordance with Eq. (20). We emphasize that only in this frequency region $\nu(\omega - \omega_0)$ can be affected by the ratio between the mean length R_c of surface defects and the exciton radius a_0. However, the asymptotic (20) can be realized only when $\min\{\omega_W, \omega_Q\} < \omega_1$ (see Eq. (7)).

Thus, in the case of the sharp resonance (18), the dependence $\nu(\omega - \omega_0)$ is *asymmetric*.

In the opposite case, when the resonance is broad,

$$\min\{\omega_W, \omega_Q\} \ll \nu_0 + \nu, \tag{21}$$

the exciton–surface scattering frequency satisfies the universal cubic equation,

$$\frac{\nu}{\nu_b^2} = \frac{\nu_0 + \nu}{(\omega - \omega_0)^2 + (\nu_0 + \nu)^2}. \tag{22}$$

The normalizing constant ν_b is defined by

$$\nu_b = \left(\frac{\nu_l}{\pi}\int_0^{\infty} d\omega_t Q(\omega_t)\frac{W\left(\sqrt{2M\omega_t/\hbar}\right)}{W(0)}\right)^{1/2}. \tag{23}$$

This frequency is sensitive to the relation between the correlation length R_c and the exciton radius a_0. Within the adiabatic limit $\left(\omega_W \ll \omega_Q\right)$

$$\nu_b \approx 2\frac{\hbar(\pi/d)^2}{2\mu}\frac{\zeta}{d}, \tag{24}$$

while at the non-adiabatic surface interaction $\left(\omega_Q \ll \omega_W\right)$

$$\nu_b \approx \left(\frac{2B}{\pi}\right)^{1/2}\frac{\hbar(\pi/d)^2}{2\mu}\left(1 + \frac{4\mu}{M}\right)^{1/2}\frac{\zeta}{d}\frac{2R_c}{a_0}. \tag{25}$$

The solution $\nu(\omega - \omega_0)$ of Eq. (22) reaches its maximum at the resonance point $\omega = \omega_0$ and *symmetrically* decreases towards both sides of the exciton resonance. The tails of $\nu(\omega - \omega_0)$ crucially depend on the value of the bulk scattering frequency ν_0. When $\nu_0 = 0$, Eq. (22) is reduced to a quadratic whose solution can be presented as the semicircle of radius ν_b with its center at the point ($\nu = 0$, $\omega - \omega_0 = 0$).

The exciton–surface scattering frequency ν depends substantially on both the well width d and the r.m.s. roughness height ζ. In the case of sharp resonance $\nu \propto d^{-6}\zeta^2$, whereas for the broad resonance $\nu \propto d^{-3}\zeta$. It is interesting that the latter dependence of ν on ζ is linear.

Finally, we should emphasize that all our results follow directly from the self-consistent approach applied here to solve the problem. Indeed, within the ordinary Born approximation, Eq. (6) contains G_0 instead of $\bar{G}$ between interaction operators and, hence, the action of ν on itself is not taken into account. As a result, at $\nu_0 = 0$ the resonant function $\Delta_{\nu_0+\nu}(\omega_t)$ is reduced to $\pi\delta(\omega - \omega_0 - \omega_t)$ and Eq. (11) for ν is transformed into the simple explicit expression

$$\frac{\nu}{\nu_1} = \frac{W\left(\sqrt{2M(\omega - \omega_0)/\hbar}\right)}{W(0)} Q(\omega - \omega_0)\Theta(\omega - \omega_0). \qquad (26)$$

In this simpler approximation, the scattering frequency ν turns out to be equal to zero to the left of the exciton resonance and is described by Eq. (20) to its right. Besides, the ordinary Born approximation cannot describe the case (21) of the broad resonance. At the same time, Eq. (26) manifests in the most pronounced form the asymmetry of $\nu(\omega - \omega_0)$ with respect to the resonance point $\omega = \omega_0$. This fact is due to the fundamental quantum origin of that asymmetry, which can be observed within any convenient approach.

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