Effect of a random adiabtic potential on the optical properties of two-dimensional excitons

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We investigate the absorption spectrum and the distribution of radiative decay times for twodimensional excitons in semiconductor quantum wells, as affected by a random adiabatic in-plane excitonic potential. We limit our discussion to the case of a white-noise potential. Such a potential could arise as a result of fluctuations in the composition of an alloy semiconductor, or alternatively, fluctuations in quantum-well thickness. We find, in general, that the shortest radiative decay time is directly proportional to the inhomogeneous broadening of the exciton line in absorption, which in turn is proportional to the correlation parameter of the random potential. We calculate the magnitude of the correlation parameter for several cases of potential interest. The theory is in qualitative agreement with available experimental data.

From the earliest days in the history of undoped semiconductor quantum wells,¹ exciton absorption and recombination spectra have been used to estimate or verify the quantum well growth parameters. The energy at the peak of the spectrum was immediately understood to be that of an exciton and thus related to the onedimensional (1D) "particle in a box" confinement energies of the electron and the hole. Somewhat later² it was also recognized that the exciton linewidth could be used to estimate fluctuations in the width of the box, and the linewidth quickly became a standard measure of the interface quality: narrow linewidths imply uniform interfaces, and good quantum wells.

The Stokes shift, or the difference between the exciton peak energies for absorption and emission, has also been recognized to be important.³ While the peak of absorption can be thought to depend simply on the intrinsic mean quantum-well thickness, the emission peak must be explained as a more complicated average over the actual partially thermalized exciton population. Even the concept of mean quantum-well thickness is not yet fully understood, as pointed out by Warwick, et al.,4 who called attention to the fact that an actual interface likely has a complicated spectrum of fluctuations. It is the purpose of this paper to make a theoretical contribution to the understanding of both absorption and emission spectra and their relation to the interfacial structure. In doing so, we use simplified, single-parameter, models of interfacial disorder to obtain densities of states and transition rates for two-dimensional weakly localized excitons.

We took our original inspiration from the seminal work of Feldmann *et al.*,⁵ who measured exciton recombination rates in a variety of samples and then related them to the corresponding homogeneous linewidths. They further saw a connection between the linewidth and a coherence volume (coherence area in the 2D case) over which the center-of-mass wave function is "coherent." We find this connection to be somewhat mysterious. In this paper we develop an alternative description of the coherence volume, related closely to the localization length. The spectrum of localization lengths, in turn, can be related to the spectrum of fluctuations in the exciton's potential energy as it moves in the plane of the quantum well. Finally, this random potential is analyzed in terms of interfacial and alloy disorder.

The idea that the exciton recombination rate should depend on the center-of-mass coherence volume goes back at least to the work of Rashba and Gurgenishvili,⁶ who discussed the giant oscillator strength that could be found for an exciton weakly bound to a point defect: the weaker the binding, the larger the (center-of-mass) wave function, and hence the larger the oscillator strength. Numerous authors have made use of this idea to develop understanding of the optical properties of quantum-well and quantum-wire excitons. For example, Hanamura 7 and later Andreani, Tassone, and Bassani⁸ showed how the 1D confinement of a 2D free exciton would allow it to couple to a 1D density of photon states and recombine radiatively with a well defined lifetime. This idea has been extended by Citrin⁹ and by Srinivas et al.,¹⁰ to include dephasing processes, removing the limitation of total excitonic freedom. Others have sought to develop statistical models of exciton trapping or classical localization,^{3,11} and have shown the important effect of in-plane localization on excitonic nonlinearities.^{12,13} The lifetimes of quantum-wire (1D) excitons have also been explored theoretically by Citrin.^{14,15} The experimental investigation of the lifetimes of excitons in a variety of 2D and 1D structures has been the focus of a number of publications^{5,10,16-20} and the relation of these to the present work will be discussed later.

For our description of the exciton's random 2D poten-

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tial, we invoke a model of statistical interfacial roughness, which lends itself to the well-developed theory of localization. In our model, there is no long-range correlation of the interface structure, so we can make no contact with those^{4,16,21-24} who invoke atomically smooth "grown islands" with lateral dimensions exceeding the exciton Bohr radius (10–15 nm). We are not aware of any statistical techniques for generalizing our theory to allow some long-range correlation.

The rate of radiative recombination is described by the following expression:²⁵

$$\frac{1}{\tau_r} = \frac{4}{3} \frac{\omega n}{\hbar} \frac{e^2}{m_0^2 c^3} \sum_{\mu} \left| \langle i | \hat{p}_{\mu} | f \rangle \right|^2 \,, \tag{1}$$

where n is the refractive index, ω and c are the frequency and velocity of light. For the heavy-hole exciton, summing over μ gives

$$\sum_{\mu} |\langle i | \hat{p}_{\mu} | f \rangle|^{2} = K \left[\left| \left\langle S | \hat{p}_{x} | \frac{(X + iY)}{\sqrt{2}} \right\rangle \right|^{2} + \left| \left\langle S | \hat{p}_{y} | \frac{(X + iY)}{\sqrt{2}} \right\rangle \right|^{2} \right]$$
$$= K P^{2} , \qquad (2)$$

where $P = \langle S | \hat{p}_x | X \rangle$ is the Kane matrix element, and K is the square of the overlap integral of electron and hole wave functions:

$$K = \left| \int d^3 r_e \, d^3 r_h \, \psi(\mathbf{r}_e, \mathbf{r}_h) \, \delta(\mathbf{r}_e - \mathbf{r}_h) \right|^2 \,. \tag{3}$$

As a result, the rate of radiative recombination can be written in the following form:

$$\frac{1}{\tau_r} = \frac{K}{\tau_0} , \qquad (4)$$

where $\frac{1}{\tau_0} = \frac{4}{3 \times 137} \omega n \frac{P^2}{m_0^2 c^2}$ is of the order of 1 ns for all semiconductors.²⁶ The overlap integral in Eq. (3) expresses the strong sensitivity of the radiative process to the correlation of electron and hole motion.

We shall consider only the case of a two-dimensional exciton for which the wave function can be written as the following product:

$$\psi(\mathbf{r}_e, \mathbf{r}_h) = \Psi(\mathbf{R}) \,\varphi_{2\mathrm{D}}(\rho) \,\chi_e(z_e) \,\chi_h(z_h) \,, \qquad (5)$$

where $\rho_{e,h}$ and $z_{e,h}$ are the electron and hole positions along and in the perpendicular to the plane of the quantum well, respectively, $\rho = \rho_e - \rho_h$, $\mathbf{R} = (m_e \rho_e + m_{\parallel} \rho_h)/(m_e + m_{\parallel})$ is the coordinate describing the position (in-plane) of the exciton's center of mass, and m_e and m_{\parallel} are the respective electron and hole masses. $\varphi_{2D}(\rho)$ is the function describing the relative positions of the electron and the hole in the plane of the quantum well (QW). In the strictly 2D case, φ_{2D} has the form:

$$\varphi_{2\rm D}(\rho) = \frac{4}{a\sqrt{2\pi}} e^{-2\rho/a} ,$$
 (6)

where $a = \kappa \hbar^2 / \mu e^2$ is the exciton Bohr radius, $1/\mu = 1/m_e + 1/m_{\parallel}$ is a reduced effective mass. In the quasitwo-dimensional case, a can be considered as a variational parameter.²⁷ $\Psi(\mathbf{R})$ describes the exciton's center-of-mass motion in the plane of the quantum well. $\chi_e(z_e)$ and $\chi_h(z_h)$ are the wave functions for electron and hole motion in the direction of the QW axis. For an electron or hole in the absence of the other, this function is that of a particle in a one-dimensional rectangular quantum well.²⁸ For our purposes here, we recognize that these functions appear only in the integral $\int dz \chi_e(z) \chi_h(z)$, which is of order unity and in any case is constant, in a given structure, for all excitons under consideration.

Substituting Eq. (5) into Eq. (3) one gets the following expression for the rate of radiative recombination of the *i*th exciton state:

$$\frac{1}{\tau_r(i)} = \frac{|\varphi(0)|^2 K_z}{\tau_0} \left| \int d^2 R \Psi_i(\mathbf{R}) \right|^2 , \qquad (7)$$

where $K_z = |\int dz \chi_e(z) \chi_h(z)|^2$. One can see that the rate of exciton radiative recombination depends strongly on the exciton's center-of-mass wave function through the factor $|\int d^2 R \Psi_i(\mathbf{R})|^2$. This factor has the dimension of an area, and expresses precisely the idea of the "coherence area" of the radiating exciton. The radiative recombination rate is enhanced by this area divided by the effective area of the exciton $1/|\varphi(0)|^2$.

From now on we will study this function in nonideal quantum wells, in which the exciton line is inhomogeneously broadened. We will use the adiabatic approximation: we assume that the energies of electron and hole quantization (within the well), as well as the binding energy of the two-dimensional exciton are larger than the inhomogeneous broadening. In this case, the exciton's center-of-mass wave function is described by the Schrödinger equation with the adiabatic random potential $V(\mathbf{R})$:

$$\left[\frac{\hat{p}^2}{2M} + V(\mathbf{R})\right] \Psi(\mathbf{R}) = \varepsilon \Psi(\mathbf{R}) , \qquad (8)$$

where $M = m_e + m_{\parallel}$ is the total mass of the exciton.

The exciton's radiative lifetime depends on the exciton state i and we can reasonably limit our interest to the average decay rate (at energy ε),

$$\frac{1}{\tau_{r}(\varepsilon)} = \frac{\sum_{i} \frac{1}{\tau_{r}(i)} \,\delta(\varepsilon_{i} - \varepsilon)}{\sum_{i} \delta(\varepsilon_{i} - \varepsilon)} \,, \tag{9}$$

which can be rewritten in the following form:

$$\frac{1}{\tau_r(\varepsilon)} = \frac{|\varphi(0)|^2 K_z}{\tau_0} \frac{\sum_i \left| \int dR^2 \Psi_i(\mathbf{R}) \right|^2 \delta(\varepsilon_i - \varepsilon)}{\sum_i \delta(\varepsilon_i - \varepsilon)} \\ = \frac{|\varphi(0)|^2 K_z}{\tau_0} \frac{\mathbf{A}(\varepsilon)}{\rho(\varepsilon)} , \qquad (10)$$

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where $\rho(\varepsilon)$ is the density of exciton states, and the function $\mathbf{A}(\varepsilon)$ is directly proportional to the exciton absorption coefficient. It is easy to show that $\mathbf{A}(\varepsilon)$ satisfies the following condition:²⁹

$$\int \mathbf{A}(\varepsilon) \, d\varepsilon = 1 \; . \tag{11}$$

To evaluate $\mathbf{A}(\varepsilon)$, we turn our attention to the potential function $V(\mathbf{R})$. We will assume that it is a whitenoise random potential, satisfying the following correlation relation:

$$\langle V(\mathbf{R})V(\mathbf{R}')\rangle = \Gamma \,\delta(\mathbf{R} - \mathbf{R}') \;.$$
 (12)

The value of Γ will be obtained later for several different cases. In the case of the white-noise potential, the function $\mathbf{A}(\varepsilon)$ has been discussed previously by Raikh and Efros.³⁰ We outline their conclusions briefly.

In an ideal quantum well, direct optical excitation of free excitons is possible only at a single isolated frequency, because of the requirement of the wave vector or momentum conservation. Absorption of light over a broad range of energies is made possible by scattering or localization of the exciton in the potential $V(\mathbf{R})$.

At large energy ε , the excitonic wave functions are nearly plane waves, and one can use perturbation theory to calculate $\mathbf{A}(\varepsilon)$. The result is³⁰

$$\mathbf{A}(\varepsilon) = \frac{1}{S} \int \frac{d^2k}{(2\pi)^2} \frac{\left\langle \left| \int V(\mathbf{R}) \, e^{i\mathbf{k}\mathbf{R}} \, d^2R \right|^2 \right\rangle}{\varepsilon_k^2} \, \delta(\varepsilon - \varepsilon_k) \,, \tag{13}$$

where S is a normalization constant, $\varepsilon_{\mathbf{k}} = (\hbar k)^2/2M$ is the kinetic energy of the exciton, **k** is the exciton's wave vector in the plane of the quantum well. Using Eq. (12), we get from Eq. (13):

$$\mathbf{A}(\varepsilon) = rac{W}{\varepsilon^2} \quad ext{for} \quad \varepsilon \gg W \quad , agenum{(14)}$$

where $W = M\Gamma/2\pi\hbar^2$ is the appropriate energy scale for discussing localization of a particle of mass M.

In the opposite extreme, i.e., the low-energy absorption tail ($\varepsilon < 0, |\varepsilon| \gg W$), the absorption coefficient was found by the optimal fluctuation technique. Here, the absorption is determined by those optimal fluctuations that are capable of localizing excitons most deeply. The procedure then gives

$$\mathbf{A}(\varepsilon) = \frac{0.24}{|\varepsilon|} \left(\frac{|\varepsilon|}{W}\right)^{0.57} e^{-0.93|\varepsilon|/W} \quad \text{for} \quad |\varepsilon| \gg W \;.$$
(15)

Joining of Eq. (12) and Eq. (13) and using the proper normalization of $\mathbf{A}(\varepsilon)$ leads to the following expressions:³⁰

$$|\tau|^{-0.43} e^{-0.93|\tau|}, \qquad \tau < -1.5$$

$$\mathbf{A}(\varepsilon) = \frac{1}{W} \begin{cases} \frac{1}{(\tau - 0.67)^2 + 5.28} \frac{1}{2} \\ \times \left(1 + \frac{1}{\sqrt{1 + \frac{0.86}{(\tau + 1.5)^2}}} \right), \ \tau > -1.5 \end{cases}$$
(16)

where $\tau = \varepsilon/W$. The function $\mathbf{A}(\varepsilon)$ is presented in Fig. 1, multiplied by W and by 5, the first in order to make it dimensionless and universal, and the second simply to make it reach a peak value near unity. Recall that $\mathbf{A}(\varepsilon)$ represents the shape of the exciton absorption line, whose half width Δ can be found from the figure: $\Delta \approx 3.7 W = 0.59 M\Gamma/\hbar^{2.30}$ Since in the figure the energy scale is also normalized by the parameter W, we see, as expected, that the absorption spectrum becomes broader and correspondingly weaker at the peak as the disorder, W, increases, so that its integrated strength remains constant.

The problem of defining the zero of energy is not trivial, but has been shown by Thouless and Elzain^{31} to have an acceptable solution. For our purposes and for comparison with experiments, the energy scale can be set by the position and the width of the absorption spectrum, in comparison with $\mathbf{A}(\varepsilon)$.

In Fig. 1, we also show the dependence of the dimensionless density of states $\rho(\varepsilon)/\rho_0$ calculated by Thouless and Elzain for a white-noise potential. $\rho_0 = M/2\pi\hbar^2$ is the 2D density of states for free, nondegenerate par-

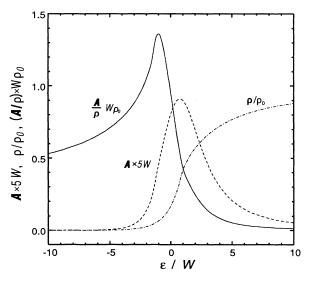


FIG. 1. Spectra of absorption (**A**), radiative recombination rate (\mathbf{A}/ρ), and density of states (ρ), for 2D excitons in a white-noise random adiabatic potential with strength parameter W. The energy scale and the curves are all normalized by the same parameter W, so each function fits any random potential. The integrated absorption strength is independent of the broadening parameter, and the minimum radiative lifetime (found at the peak of the \mathbf{A}/ρ curve) is directly proportional to W. The normalizing parameter for the density of states is the 2D free-particle density of states $M/2\pi\hbar^2$.

ticles of mass M. Finally, we plot also the function $\mathbf{A}(\varepsilon)/\rho(\varepsilon)$ made dimensionless by multiplying by $W\rho_0$. This function, according to Eq. (10), represents the distribution of the radiative rates $1/\tau_r(\varepsilon)$, normalized by $\tau_0/(K_z S_{\mathrm{loc}}|\varphi(0)|^2)$, where $S_{\mathrm{loc}} = (1/W)(2\pi\hbar^2/M) = (1/\Gamma)(2\pi\hbar^2/M)^2$. S_{loc} is a characteristic area determined by the above combination of particle mass and depth of potential fluctuations.

We call particular attention to the fact that the recombination rate appears to be reduced by disorder. From the height of the peak of this curve, we see that fastest rate of recombination in this distribution is described by the following expression:

$$\frac{1}{\tau_{\min}} \approx 1.36 \, \frac{K_z |\varphi(0)|^2}{\tau_0} \frac{(2\pi\hbar^2)^2}{\Gamma M^2} \\\approx 31.5 \, \frac{K_z |\varphi(0)|^2}{\tau_0} \frac{\hbar^2}{\Delta M}. \tag{17}$$

One can see that the minimum time of recombination is directly proportional to the width of the exciton line Δ . Feldmann *et al.*,⁵ use, by assumption, a similar relationship, but with the recombination time proportional to the *homogeneous* linewidth, rather than as in our theory, the inhomogeneous linewidth. Each is an inversion of the usual inverse relationship between lifetime and width in the case of lifetime broadening.

On each side of the peak the recombination rate decreases: for low energies because of the shrinking of the wave function $\Psi(\mathbf{R})$ for more strongly localized states, and for high energies as the states approximate more exactly those of freely propagating nonradiative excitons.

Next we consider the question of how the energy dependence of the radiative lifetime might affect the shape and the time dependence of the recombination spectrum. The rate of recombination at a given energy will depend not only on the radiative lifetime, but also on the rate at which excitons jump between their localized states, emitting and absorbing phonons. Two temperature regimes can be identified.

At low temperature, the distribution of excitons in the tail of localized states will be far from equilibrium. This is a result of the fact that radiative rates will be overwhelmingly faster than thermalization rates. The rate of exciton tunneling between localized states decreases drastically in the tail of the distribution because the concentration $N(\varepsilon)$ of states to which the exciton with localization energy $|\varepsilon|$ can jump decreases exponentially with $|\varepsilon|$, according to

$$N(\varepsilon) = \int_{-\infty}^{-|\varepsilon|} \rho(\varepsilon) \, d\varepsilon = 0.18 \, W \rho_0 \, e^{-0.93 \, |\varepsilon|/W}.$$
(18)

As a result the average tunneling distance grows and makes further thermalization very improbable.

The position of the maximum ε_m of the luminescence line can be taken to be approximately equal to the energy at which the thermalization time is equal to the recombination time. Excitons from shallow states can live long enough to reach this energy, but those below ε_m will probably decay radiatively before further thermalization. To find the balance between these competing processes and to determine the resultant spectrum requires a detailed understanding of the thermal relaxation of the localized excitons. An excellent calculation of siteto-site transfer rates for localized excitons has been made by Takagahara,³² who evaluated the phonon-assisted exciton transition rates within the adiabatic approximation. He included both the tunneling mechanism and the long-range Coulomb mechanism in his calculations. We have not yet attempted to convolute his work with ours. A recent effort to calculate a distribution function for nonequilibrium localized excitons used only an energy dependent density of states, ignoring the energy dependence of radiative lifetime and localization radius.³³

Direct experimental access to the radiative lifetime of a localized exciton appears impossible, except perhaps in the case of direct optical excitation in the low energy tail of absorption. An alternative has very recently appeared, using a near-field microscopic technique to isolate the sites at which excitons are localized.³⁴

At high temperature (kT larger than the exciton)linewidth W, the process of thermalization is much faster than that of radiation. In this case, there will be a Boltzmann distribution of exciton energies, and the decay times at all energies will be determined by the average decay time $\tau(T)$. Neglecting the possibility of nonradiative decay, this time is determined by

$$\frac{1}{\tau(T)} = \frac{\int 1/\tau_r(\varepsilon) \exp(-\varepsilon/kT)\rho(\varepsilon) d\varepsilon}{\int \exp(-\varepsilon/kT) \rho(\varepsilon) d\varepsilon}$$
$$= \frac{|\varphi(0)|^2 K_z}{\tau_0} \frac{\int \mathbf{A}(\varepsilon) \exp(-\varepsilon/kT) d\varepsilon}{\int \exp(-\varepsilon/kT)\rho(\varepsilon) d\varepsilon}.$$
(19)

 $\tau(T)$ increases with temperature simply because the higher-energy, nearly free excitons, have longer radiative lifetimes. In the limit $kT \gg W$, one can obtain

$$\tau(T) = \frac{\tau_0 \,\rho_0 \,kT}{|\varphi(0)|^2 K_z} = \frac{\tau_0 \,kTM}{2\pi\hbar^2 \,|\varphi(0)|^2 \,K_z},\tag{20}$$

and the radiative lifetime has no dependence on the degree of disorder. This expression is identical to the result obtained for free exciton emission by Andreani *et al.*,⁸ when we take into account the "triplet" exciton state, whose radiative lifetime is effectively infinite.

Numerical calculation of Eq. (19) shows that Eq. (20) remains valid with 10% accuracy if kT > 1.4 W, i.e., if $kT > 0.38 \Delta$ where Δ is the linewidth (inhomogeneous) of the exciton's absorption spectrum. One can see that at the low temperature limit for Eq. (20) (for $kT = 0.38 \Delta$), the lifetime is only 1.9 times longer than the shortest radiative lifetime given by Eq. (17) and it is also proportional to the width of the exciton line. As a result the fastest exciton radiative decay time that can be observed at any temperature is in the range of τ_{\min} to $1.9 \tau_{\min}$ and is limited by the exciton's inhomogeneous broadening. Equation (20) may have a very narrow or even nonexistent range of validity if the exciton linewidth is comparable with the exciton binding energy.

Most of the experimental investigations¹⁷⁻²⁰ of the decay time as a function of QW thickness and temperature really show instead its dependence on the interface quality.^{17–20} There is little quantitative agreement between these data and calculations done on the basis of an ideal quantum-well model. One can find a connection of the decay time with inhomogeneous broadening in the data of Sauer et al.¹⁸ for ${\rm Ga_{0.47}In_{0.53}As/Ga_{\it x}In_{1-\it x}As_{\it y}P_{1-\it y}}$ QW's. Two types of samples with different inhomogeneous broadening were investigated there. In the QW's with the same thickness, the decay time was shorter for the sample where inhomogeneous broadening was smaller. This is in qualitative agreement with our results. Nevertheless more experimental investigations of this effect should be done for comparison with the theory.

In the Appendix which follows, we evaluate the correlation parameter for several cases of interest. From these we have evaluated the shortest radiative lifetimes, for quantum-well thicknesses of 2nm, where the alloy fluctuation have their strongest influence. The results are as follows: for Ga_{0.47}In_{0.53}As/InP, The results are as follows: for $Ga_{0.4}TII_{0.53}AS/III^{2}$, $\tau_{\rm min} = 1.2 \times 10^{-11} \, {\rm s}$; for $CdTe/Cd_{0.8}Mn_{0.2}Te$, $3.4 \times 10^{-13} \, {\rm s}$; for $Ga_{0.7}In_{0.3}As/GaAs$, $7.4 \times 10^{-12} \, {\rm s}$; and for $GaAs/Al_{0.3}Ga_{0.7}As$, $2.2 \times 10^{-13} \, {\rm s}$. These theoretical decay times are extremely short, and are predicted for QW's for which the only randomness in the exciton's adiabatic potential comes from alloy fluctuations. These times could be observed in ideal quantum wells, in the absence of quantum-well-thickness fluctuations. Realistic modeling of well-thickness fluctuations is beyond the scope of this paper. We have investigated the effect of atomic-scale white-noise fluctuations at the interfaces and shown that they produce negligible contributions to the correlation parameter Γ . Extension of the model to include atomic clusters (short-range correlations) indicates that such correlations increase considerably the parameter Γ , and hence the distributions of energies and lifetimes. However, we see no way to extend our model to include the long-range order that appears to be present in quantum wells with large uniform areas referred to as islands. This is a serious limitation to the applicability of our technique, as a preponderance of actual quantumwell samples show evidence of long-range islandlike correlations. Within the confines of a large island, however, our theory could account for broadening and lifetime effects.

In conclusion, the decay time of the 2D exciton luminescence depends not only on the exciton's *internal* properties such as binding energy, the overlap integral between electron and hole wave function, and Kane matrix elements, but also on the exciton's *external* motion in the QW plane. In our model with a white noise random potential, the resulting spectrum of localized exciton states gave us a correlation between the inhomogeneous broadening of the absorption line and the radiative decay time. We found the minimum recombination time to be directly proportional to the width of the exciton line. This is, as noted above, in qualitative agreement with some recent experimental data.

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APPENDIX

In this section, we will show how to evaluate the correlation parameter Γ for two cases of potential interest, namely, those in which the quantum well is an alloy and the barrier is a simple binary III-V semiconductor (I) and in which the roles of alloy and binary semiconductors are reversed (II). In these cases the "well width fluctuations" are simple consequences of the fluctuations of chemical concentrations in the alloys.

Case I. The quantum well is a semiconductor alloy and the barrier is a normal binary semiconductor. In this case the random potential acting on electrons, $V_e(\mathbf{r})$, and holes, $V_h(\mathbf{r})$, can be written in the form

$$V_{e,h}(\mathbf{r}) = \begin{cases} \alpha_{e,h}\xi(\mathbf{r})/N , & |z| < L/2; \\ 0 , & |z| > L/2; \end{cases}$$
(A1)

where $\xi(\mathbf{r})$ is the random fluctuation of local concentration of atoms in the semiconductor alloy $A_x B_{1-x}$ from the average value xN, where N is the concentration of lattice cites:

$$\langle \xi(\mathbf{r})\xi(\mathbf{r}')\rangle = Nx(1-x)\,\delta(\mathbf{r}-\mathbf{r}')$$
 (A2)

 $\alpha_e = dE_c/dx$ and $\alpha_h = dE_v/dx$ characterize the rate of the shift of conduction and valence bands with composition x. Using the adiabatic approximation, we can obtain the random potential $V(\mathbf{R})$ acting on the exciton center of mass:

$$V(\mathbf{R}) = \int dz \, d^2 \rho \, |\chi_e(z)|^2 \, |\varphi_{2\mathrm{D}}(\rho)|^2 \, V_e\left(\mathbf{R} + \frac{m_{\parallel}}{M}\rho, z\right) \\ + \int dz \, d^2 \, \rho \, |\chi_h(z)|^2 \, |\varphi_{2\mathrm{D}}(\rho)|^2 \, V_h\left(\mathbf{R} - \frac{m_e}{M}\rho, z\right).$$
(A3)

Using Eq. (A1) and neglecting $(m_{e,\parallel}/M)\rho$ in comparison with R, an assumption whose validity is implied in the adiabatic approximation, we can obtain an expression for Γ in Eq. (14):

$$\Gamma = \frac{(1-x)x}{N} \left(\alpha_e^2 \int_{-L/2}^{L/2} dz \, \chi_e^4(z) + \alpha_h^2 \int_{-L/2}^{L/2} dz \, \chi_h^4(z) \right). \tag{A4}$$

In Fig. 2, we present the calculated dependence of Γ on *L* for quantum wells of $\ln_x \operatorname{Ga}_{1-x} \operatorname{As}$ within barriers of GaAs and InP. For the strained $\operatorname{Ga}_x \operatorname{In}_{1-x} \operatorname{As}/\operatorname{InP}$ system, we use the conduction band offset $\Delta E_c = (464 - 408 x)$ meV (Ref. 35) in the range 0.4 < x < 0.6 and the experimental effective masses $m_{e,w}/m_0 = 0.033 + 0.027 x$

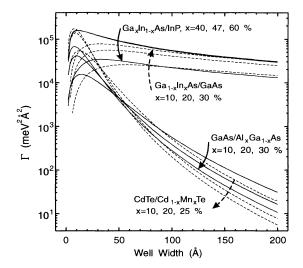


FIG. 2. Evaluation of the correlation parameter Γ plotted as a function of quantum-well width for a variety of interesting material combinations, as indicated. The arrow indicates the direction of the increasing composition parameter x. Γ is directly proportional to the broadening parameter via the 2D free-particle density of states. As discussed in the text, the major differences are between systems with alloy in the well and alloy in the barrier. In the latter case, for moderately wide wells, the fluctuations have relatively little effect on the random potential. In both cases, the sensitivity peaks for narrow wells, and then weakens for very narrow wells as the excitons are no longer seriously confined.

in the well³⁶ and $m_{e,b} = 0.077 m_0$ in the barrier.³⁷ For the valence band, we use $\Delta E_v = (449 - 445.5 x) \text{ meV}$, $m_{h,w}/m_0 = 0.41 + 0.04 x$ in the range $0.4 \le x \le 0.47$, and $\Delta E_v = (287.8 + 110.5 x) \text{ meV}.^{35} m_{h,w}/m_0 = 0.025 + 0.057 x$ for $0.47 < x \le 0.6.^{35}$ The mass in the barrier is $m_{hh,v}/m_0 = 0.60.^{37}$ The lattice constant is given by $a = 5.869 \text{ Å}.^{37}$

In the Ga_{1-x}In_xAs/GaAs system the corresponding values are given by the band gap difference $\Delta E_g = (1144 x - 255 x^2) \text{ meV},^{38}$ the relative conduction band offset $\Delta E_c/\Delta E_g = 0.60$, and the masses $m_e/m_0 = 0.065 - 0.070 x,^{38}$ and $m_v/m_0 = 0.45 - 0.93 x + 0.83 x^2.^{38}$ The lattice constant is $a = 5.653 \text{ Å}.^{37}$

Here we see that for reasonably wide quantum wells the broadening parameter is only weakly (as 1/L) dependent on the width. In thinner quantum wells Γ drops rapidly because the electron and hole wave functions expand into the barrier regions, and the alloy disorder affects only the part of the wave function within the well. With a lower barrier, the crossover occurs at a larger well width, which explains the variation with alloy parameter x.

Case II: The quantum well is a binary semiconductor and the barrier is a semiconductor alloy. In this case the random potentials acting on electrons and holes have the following form:

$$V_{e,h}(\mathbf{r}) = \begin{cases} \alpha_{e,h} \, \xi(\mathbf{r})/N \ , \ |z| > L/2; \\ 0 \ , \qquad |z| < L/2; \end{cases}$$
(A5)

where $\alpha_{e,h}$ characterize the dependences of conduction and valence band edges of the barrier on the composition x. Now the random potential acting on exciton depends on penetration of electron and hole wave functions into the barrier, and the correlation constant Γ has the following form:

$$\Gamma = \frac{2(1-x)x}{N} \left(\alpha_e^2 \int_{L/2}^{\infty} dz \, \chi_e^4(z) + \alpha_h^2 \int_{L/2}^{\infty} dz \, \chi_h^4(z) \right) \,. \tag{A6}$$

In Fig. 2, we present the dependence of Γ on L for quantum wells of GaAs within $Al_xGa_{1-x}As$ barriers and of CdTe within Cd_xMg_{1-x} Te barriers. In $GaAs/Al_xGa_{1-x}As$ the bandstructure is given by $\Delta E_g = 1230 x \text{ meV},^{37} \Delta E_c/\Delta E_g = 0.65$, and the masses are given by $m_e/m_0 = 0.065 + 0.084 x$. For the heavy holes, we use $m_h/m_0 = 0.51 - 0.10 x.^{37}$

In CdTe/Cd_{1-x}Mn_xTe we use for the band offset $\Delta E_g = 1592 x \text{ meV}$,³⁹ $\Delta E_c/\Delta E_g = 0.7$ and the masses $m_{e,w}/m_0 = m_{e,b}/m_0 = 0.1$, and $m_{h,w}/m_0 = m_{h,b}/m_0 = 0.49$.³⁹ The lattice constant is a = 6.481 Å.³⁷ In these cases, the alloy is in the barrier, and so has a minimal effect on excitons in moderately wide wells. With decreasing width, however, the electron and hole feel the alloy fluctuations more seriously, and Γ rises. Eventually, in very thin wells, the expanded wave functions average over many layers of the alloy, and Γ again decreases.

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- ¹ R. Dingle, W. Wiegmann, and C. H. Henry, Phys. Rev. Lett. **33**, 827 (1974).
- ² C. Weisbuch, R. Dingle, A. C. Gossard, and W. Wiegmann, Solid State Commun. **38**, 709 (1981).
- ³ F. Yang, M. Wilkinson, E. J. Austin, and K. P. O'Donnell, Phys. Rev. Lett. **70**, 323 (1993).
- ⁴ C. A. Warwick, W. Y. Jan, A. Ourmazd, and T. D. Harris, Appl. Phys. Lett. **56**, 2666 (1990).
- ⁵ J. Feldmann, G. Peter, E. O. Göbel, P. Dawson, K. Moore,

C. T. Foxon, and R. J. Elliott, Phys. Rev. Lett. 59, 2337 (1987).

- ⁶ E. I. Rashba and G. E. Gurgenishvili, Fiz. Tverd. Tela 4, 1029 (1962) [Sov. Phys. Solid State 4, 759 (1962)].
- ⁷ E. Hanamura, Phys. Rev. B 38, 1228 (1988).
- ⁸ L. C. Andreani, F. Tassone, and F. Bassani, Solid State Commun. 77, 641 (1991).
- ⁹ D. S. Citrin, Solid State Commun. 84, 281 (1992).
- ¹⁰ V. Srinivas, J. Hryniewicz, Y. J. Chen, and Colin E. C. Wood, Phys. Rev. B 46, 10193 (1992).
- ¹¹ G. Bastard, C. Delalande, M. H. Meynadier, P. M. Frijlink, and M. Voos, Phys. Rev. B 29, 7042 (1984).
- ¹² T. Takagahara and E. Hanamura, Phys. Rev. Lett. 56, 2533

- ¹³ R. Zimmermann, Phys. Status Solidi B **173**, 129 (1992).
- ¹⁴ D. S. Citrin, Phys. Rev. Lett. **69**, 3393 (1992).
- ¹⁵ D. S. Citrin, Solid State Commun. **92**, 851 (1994).
- ¹⁶ K. Fujiwara, H. Katahama, K. Kanamoto, R. Cingolani, and K. Ploog, Phys. Rev. B 43, 13978 (1991).
- ¹⁷ A. Polhman, R. Hellmann, E. O. Göbel, D. R. Yakovlev, W. Ossau, A. Waag, R. N. Bicknell-Tassius, and G. Landwehr, Appl. Phys. Lett. **61**, 2929 (1992).
- ¹⁸ R. Sauer, S. Nilsson, P. Roentgen, W. Heuberger, V. Graf, A. Hagleiter, and R. Spycher, Phys. Rev. B 46, 9525 (1992).
- ¹⁹ A. Hoffmann, H. Siegle, L. Eckey, B. Lummer, P. Thurian, R. Heitz, B. K. Meyer, C. Wetzel, D. M. Hofmann, V. Härle, F. Scholtz, and A. Kohl, Superlatt. Microstruct. 15, 303 (1994).
- ²⁰ D. Gershoni, M. Katz, W. Wegscheider, L. N. Pfeiffer, R. A. Logan, and K. West, Phys. Rev. B 50, 8930 (1994).
- ²¹ P. S. Kop'ev, B. Y. Meltser, I. N. Uraltsev, Al. L. Efros, and D. R. Yakovlev, Pis'ma Zh. Eksp. Teor. Fiz. **42**, 327 (1985) [JETP Lett. **42**, 402 (1985)].
- ²² L. Goldstein, Y. Horikoshi, S. Tarucha, and H. Okamoto, Jpn. J. Appl. Phys. 22, 1489 (1983).
- ²³ B. Deveaud, J. Y. Emery, A. Chomette, B. Lambert, and M. Baudet, Appl. Phys. Lett. 45, 1078 (1984).
- ²⁴ P. S. Kop'ev, I. N. Ural'tsev, Al. L. Efros, D. R. Yakovlev, and A. V. Vinokurova, Fiz. Tekh. Poluprovodn. **22**, 424 (1988) [Sov. Phys. Semicond. **22**, 259 (1988)]; M. Baudet, Appl. Phys. Lett. **45**, 1078 (1984).
- ²⁵ L. D. Landau and E. M. Lifshitz, *Relativistic Quantum Theory*, 2nd ed. (Pergamon Press, Oxford, 1965).
- ²⁶ We note here that half of the heavy-hole excitons are in a "triplet" state and not radiative. These are sometimes called dark excitons.
- ²⁷ R. C. Miller, D. A. Kleinman, W. T. Tsang, and A. C. Gossard, Phys. Rev. B 24, 1134 (1981).

- ²⁸ G. Bastard, Wave Mechanics Applied to Semiconductor Heterostructures (Les Editions de Physique, Les Ulis, France, 1988).
- ²⁹ N. N. Ablyazov, M. E. Raikh, and A. L. Efros, Fiz. Tverd. Tela (Leningrad) **25**, 353 (1983) [Sov. Phys. Solid State **25**, 199 (1983)].
- ³⁰ M. E. Raikh and Al. L. Efros, Fiz. Tverd. Tela (Leningrad) 26, 106 (1984) [Sov. Phys. Solid State 26, 61 (1984)].
- ³¹ D. J. Thouless and M. E. Elzain, J. Phys. C **11**, 3425 (1978).
- ³² T. Takagahara, Phys. Rev. B **31**, 6552 (1985).
- ³³ A. G. Abdukadirov, S. D. Baranovskii, S. U. Verbin, E. L. Ivchenko, A. U. Naumov, and A. N. Resnizkii, Zh. Eksp. Teor. Fiz. **98**, 2056 (1990) [Sov. Phys. JETP **71**, 1155 (1990)]; A. G. Abdukadirov, S. D. Baranovskii, and E. L. Ivchenko, Fiz. Tverd. Tela (Leningrad) **24**, 136 (1990) [Sov. Phys. Solid State **24**, 82 (1990)].
- ³⁴ H. F. Hess, E. Betzig, T. D. Harris, L. N. Pfeiffer, and K. W. West, Science **264**, 1740 (1994).
- ³⁵ T. Y. Wang and G. B. Stringfellow, J. Appl. Phys. 67, 344 (1990).
- ³⁶ B. K. Meyer, M. Drechsler, C. Wetzel, V. Härle, F. Scholz, H. Linke, P. Omling, and P. Sobkowicz, Appl. Phys. Lett. **63**, 657 (1993).
- ³⁷ Physics of Group IV Elements and III-V Compounds, edited by O. Madelung, Landolt-Börnstein, New Series, Group III, Vol. 17, Pt. a (Springer-Verlag, Berlin, 1982); Intrinsic Properties of Group IV Elements and III-V, II-VI and I-VII Compounds, edited by O. Madelung, Landolt-Börnstein, New Series, Group III, Vol. 22, Pt. a (Springer-Verlag, Berlin, 1987).
- ³⁸ H. Brugger, H. Müssig, C. Wölk, K. Kern, and D. Heitmann, Appl. Phys. Lett. **59**, 2739 (1991).
- ³⁹ B. Kuhn-Heinrich, W. Ossau, A. Waag, and G. Landwehr, J. Appl. Phys. **75**, 8046 (1994).

^{(1986).}