Electric-field-induced narrowing of exciton linewidth

I. V. Ponomarev,* L. I. Deych, and A. A. Lisypansky

Department of Physics, Queens College of the City University of New York, Flushing, New York 11367, USA

(Received 1 June 2005; published 2 September 2005)

Considering the effects of the electric field on the low temperature absorption line of quantum well excitons, we show that, for moderate strength of the electric field, the main contribution to the field dependence of the linewidth results from the field induced modifications of the inhomogeneous broadening of excitons. We find that the strength of the random potential acting on the quantum well excitons due to alloy disorder and interface roughness can either decrease or increase with the field depending upon the thickness of the well. This means that under certain conditions one can observe counterintuitive narrowing of the exciton spectral lines in an electric field.

DOI: 10.1103/PhysRevB.72.115304

PACS number(s): 73.21.Fg, 78.67.De, 71.35.Cc

In the case of three-dimensional excitons, it is well known that application of an electric field significantly reduces the exciton lifetime because of a finite probability of exciton tunneling through the field distorted potential barrier. An obvious spectroscopic consequence of this effect is the significant broadening of the exciton spectral lines at moderate electric fields. However, excitons confined in a quantum well (QW) are much more robust with respect to the electric field, \( \mathcal{E} \), applied perpendicular to the plane of a QW. As a result, QW excitons demonstrate an appreciable field induced shift of their spectral lines, while the spectral width does not change too much. The physical origin of this quantum confined Stark effect (QCSE) lies in the significantly increased stability of QW excitons compared to the bulk case. Indeed, Stark broadening shows strong exponential dependence on the field

\[
\Gamma_{\text{Stark}} \sim E_0 \exp(-2\hbar^2/3m_e|\mathcal{E}|E^3),
\]

where \( E_0 \) is a typical value of the resonance energy, \( m \) is an effective particle mass, and \( \ell \) is a tunneling length. In QWs, \( \ell \) is determined by the confining potential of the well rather than by the binding energy of excitons. Since the former is one or two orders of magnitude greater than the latter, it is clear that QW excitons in the perpendicular field can withstand much stronger fields than their three-dimensional counterparts. QCSE has received a great deal of attention during the last two decades and was exploited in a number of electro-optic devices. However, the issue of the electric-field-induced changes in the exciton linewidth, and their origin, still remains largely unstudied (see Ref. 2).

In this paper we consider the electric-field-induced modification of the exciton inhomogeneous broadening, which determines the exciton linewidth at low temperatures. We show that modification of the random potential caused by the reconstruction of electron-hole wave functions in the applied electric field results in a power law field dependence of the exciton linewidth, which yields a much stronger change in the linewidth than the exponential dependence caused by the Stark effect, Eq. (1).

We find that for QWs whose thickness, \( L \), is smaller than some critical value, \( L_{cr} \), an electric field actually reduces fluctuations of this potential resulting in a counterintuitive narrowing of the exciton linewidth with the electric field. When \( L > L_{cr} \), the sign of the electric field contribution to the linewidth changes and the exciton lines becomes broader with the field increase. The critical thicknesses, as well as \( L \) and \( \mathcal{E} \) dependencies of the exciton linewidth, are different for compositional disorder and interface roughness mechanisms of the inhomogeneous broadening. Therefore, the experimental observation of field induced changes in the low temperature exciton linewidth can yield a unique method of the characterization of QWs allowing the separation of these two contributions to the exciton’s spectral broadening.

Let us consider a QW formed by a binary semiconductor, \( AB \), as a barrier material, and a ternary disordered alloy, \( AB_{1-x}C_x \), as a well. Throughout the paper we use effective atomic units: the effective Bohr radius for length, \( a_B = \hbar^2/e^2m_e \), the Bohr electron-hole radius, \( r_B = \mu^* \hbar^2/\varepsilon e^2 = 2 \text{ Ry} \) for energy, and reduced electron-hole mass \( \mu^* \) for masses, \( 1/\mu^* = 1/m_e + 1/m_h \). In the isotropic effective mass approximation, the Hamiltonian for the exciton in a QW with a disorder is

\[
H = H_0^{\text{el}}(\mathbf{r}_e) + H_0^{\text{int}}(\mathbf{r}_e) - \mathbf{r}_e - \mathbf{r}_h^{-1} + U^{\text{el}}(\mathbf{r}_e) + U^{\text{int}}(\mathbf{r}_h),
\]

where \( U^{\text{el}}(\mathbf{r}_e)(e^{\text{el}}(\mathbf{r}_e)) \) are disorder induced potentials, and \( H_0^{\text{el}}(\mathbf{r}_e) \) are Hamiltonians for an electron(hole) in a QW

\[
H_0^{\text{el}}(\mathbf{r}) = \frac{p^2}{2m_e} + V_0(2z^2 - L^2/4) + \frac{F(z)z^2}{2m_e},
\]

where \( F(z) = 2m_e|\mathcal{E}|z \) and \( \theta(z) \) is the step function. The inhomogeneous broadening of excitons in such a well is determined by a combination of two types of disorders

\[
U^{\text{int}}(\mathbf{r}_e) = U^{\text{int}}(\mathbf{r}_e) + U^{\text{int}}(\mathbf{r}_e).
\]

Compositional disorder, \( U^{\text{int}}(\mathbf{r}_e) \), arising due to the concentration fluctuations in a ternary component, which produce local band gap fluctuations, and interface roughness, \( U^{\text{int}}(\mathbf{r}_e) \), caused by the formation of monolayer islands on the QW, interfaces that result in local changes in the QW thickness.

Usually, the exciton binding energy in a QW is much larger than disorder-induced local energy fluctuations. Therefore the excitons are expected to move through a QW as a
whole entity. It is formalized in a representation of the total wave function of the electron pair in the form of a product

\[ \Psi(r_x, r_h) = \Phi(R) \psi(\rho) \chi_h(z_h), \]

where

\[ r_x = (p_{xh}; z_x), \quad \rho = p_x - p_h, \quad R = (m_x \rho_x + m_h \rho_h)/M, \]

\[ \Phi(R) \] is a wave function for the center-of-mass motion, \[ \psi(\rho) \] is an exciton relative lateral motion wave function, and \[ \chi_h(z_h) \] are one-dimensional electron and hole QW ground state wave functions. Functions \( \psi \) and \( \chi \) are solutions of the corresponding Schrödinger equation for a perfect QW and the Schrödinger equation for the center-of-mass motion includes effective random potentials, \( V_{\text{eff}}(R) = V_{\text{int}}(R) + V_{\text{alloy}}(R), \) obtained from the averaging of the original random potentials \( U^h(r_x), U^m(r_h) \) over \( \rho \) and \( z \) coordinates. [In these calculations we do not take into account the disorder-induced renormalization of functions \( \psi, \chi, \) and the corresponding energies, \( E_x, E_h, \) which result in effective decreasing of Bohr’s radius \( \lambda. \) This effect does not change, the qualitative conclusions of our work [see Eqs. (18) and (19)].]

Our primary goal is to calculate the variance of the effective random exciton potential defined as

\[ W = \sqrt{\langle V_{\text{eff}}(R)^2 \rangle}. \]

This parameter determines a number of experimentally observable quantities such as the exciton radiative lifetime and the absorption linewidth. \( ^{3-5} \) The radiative lifetime can be extracted from the exciton absorption line shape, \( ^8 \) whose calculation in the dipole approximation is equivalent to the estimation of the optical density function: \( A(\epsilon) = \langle \Theta[\epsilon - \phi(R)] \rangle \delta(\epsilon - \epsilon_0) \rangle. \) The interpolation procedure \( ^{3,8} \) gives an asymmetric line shape towards high frequencies. In many cases, however, for estimation of the linewidth it is reasonable \( ^{4-5} \) to consider that the underlying disorders are described by the Gaussian random processes. Then the shape of the exciton line is also Gaussian, and the corresponding full width at half maximum is given by \( \lambda = \sqrt{8 \ln(2)} W. \)

Since contributions from the alloy and the interface disorders can be considered statistically independent of each other, \( W_{\text{tot}}^2 = W_{\text{alloy}}^2 + W_{\text{int}}^2. \) Estimations show that usually both disorders yield comparable contributions to the total width. This imposes an additional difficulty for experimental identification of the interface quality in QWs from optical spectra, since the absolute values of each contribution are usually unknown.

The effective random potentials acting on the exciton’s center-of-mass, for each type of disorder, is presented as a sum of two terms \( V(R) = V_h(R) + V_e(R), \) representing hole and electron contributions

\[ V_{h,e} = \int U_{h,e}(R + m_x \rho/M; z) \psi^2(\rho) \chi_{h,e}(z) dz^2 d\rho. \]

Correspondingly, each variance will have three terms \( W^2 = \langle V^2 \rangle + 2 V_h V_e + V_e^2. \) For a QW with a heavy hole and light electron \( (m_h \gg m_e) \) as in In\(_x\)Ga\(_{1-x}\)As/GaAs heterostructures, the main contribution stems from the hole-hole part due to the enhancement factor \( (M/m_h) \) which appears after averaging over lateral coordinates \( \rho \) in Eq. (7). Considering only this case, we neglect terms containing a \( v_e \) factor. Then the microscopic potential representing the alloy can be presented as (hereafter the subscript “h” is omitted)

\[ U_{\text{alloy}}(R) = \alpha \xi(R) \delta(L^2/4 - z^2)/N, \]

where \( \delta(z) \) is a step function, \( N \) is the concentration of lattice sites \( (N = 4/a_{lat}^2 \) for zinc blende materials, \( a_{lat} \) is a lattice constant), \( \xi(R) \) is the random fluctuation of the local concentration of atoms in the alloy from the average value \( \langle x \rangle \), and \( \alpha = dE_e/dx \) characterizes the rate of the shift of the valence bands with composition \( x \). The interface roughness potential can be presented in the following form \( ^{12,13} \)

\[ U_{\text{int}}(R) = V_0 \langle \eta_{x}(\rho) \delta(z + L/2) - \eta_{z}^{2}(\rho) \delta(z - L/2) \rangle, \]

where \( \delta(z) \) is a \( \delta \) function, \( V_0 \) is a hole off-set band energy. Random functions \( \eta_{x,z}(\rho) \) with zero mean characterize a deviation of the \( i \)th interface from its average position.

The statistical properties of alloy and interfacial roughness are characterized by the correlators: \( ^{6,12-15} \)

\[ \langle \xi(r_1) \xi(r_2) \rangle = x(1 - x) \delta(r_1 - r_2), \]

\[ \langle \eta_{x}(p_1) \eta_{x}(p_2) \rangle = h^2 f_{x} \xi_{x}(p_1 - p_2), \]

where \( h \) is an average height of interface inhomogeneity, and \( \langle \cdots \rangle \) denotes an ensemble average. For the interface height-height correlator we assume that the dependence of both diagonal and nondiagonal correlations on the lateral coordinates \( \rho \) is described by the same function \( \xi(\rho) \). The diagonal elements \( f_{x} \) are different if two interfaces are grown under different conditions, which happens naturally for GaAs based structures. (Growth of a ternary alloy on GaAs occurs differently from growth of GaAs on the alloy; besides using techniques of growth interruption one can significantly modify the statistical properties of the grown interfaces.) The nondiagonal element \( f_{z} \) introduces correlations between different interfaces. The respective quantity, which can be called the cross-correlation or vertical-correlation function, \( ^{13,16} \) is a function of the average width of the well and is characterized by the vertical correlation length \( \sigma_{z}. \) The presence of these correlations suppresses the interface disorder contribution into inhomogeneous broadening, especially, for narrow QWs. \( ^{13,17} \)

In order to calculate the effective potential, one needs to know the exciton wave functions \( \psi \) and \( \chi \) for an ideal QW in the perpendicular uniform electric field. They can be found with the help of the variational method. It is a well-known fact \( ^{11,18} \) that the lateral relative motion is very weakly affected by the perpendicular electric field. The corresponding trial function can be chosen in a form of the hydrogen 1S-like orbital, \( (2/\pi \lambda^2) \exp(-r/\lambda), \) with the quasitwo-dimensional Bohr’s radius \( \lambda \) as a variational parameter. In principle, the one-dimensional single-particle function \( \chi(z) \) for a hole in a QW, which satisfies the Schrödinger equation

\[ [p^2/2m + V_0(\theta z^2 - L^2/4) + Fz^2/2m] \chi = E \chi, \]

(12)

can be found exactly in terms of the Airy functions. This solution corresponds to a quasistationary hole state, and de-
scribes a possibility for the hole to tunnel out of the well. It diverges at infinity, and is not suitable, therefore, for the calculation of the effective potential, Eq. (7). In this paper, however, we are interested in the range of parameters where the Stark broadening is small. This restricts our consideration to a certain region on the $F-L$ plane (grey shaded area in Fig. 1), where the exponent of Eq. (1) is smaller than unity. It is worth it to note, that these fields can reach very high values since the relevant energy scale is the confining QW potential rather than the Coulomb interaction between the hole and the electron. For example, for In$_{0.15}$Ga$_{0.85}$As/GaAs QWs the maximum in Fig. 1 corresponds to the field $8 \times 10^5$ V/cm.

Previous studies of the QCSE showed$^{19}$ that within this range of parameters, an approximation of $\chi(z)$ by a real function, which can be found with the help of the variational method, gives a very good description of both the energy and the wave function of a hole in the presence of a perpendicular electric field. Reasonable results can be obtained even for the simplest one-parameter variational function of the following form:$^{18}$

$$\chi(z; \beta) = B(\beta, k) \exp(-\beta z) \chi_0(z),$$

where the wave function $\chi_0(z)$, represents, the hole ground state in a QW without the electric field.

$$\chi_0(z) = \begin{cases} \cos(\kappa z), & z \leq |L/2| \\ \exp[-\kappa(|z|-L/2)], & z \geq |L/2| \end{cases},$$

$B$ is the normalization constant,

$$B(\beta, k) = \sqrt{\frac{2\beta(\kappa^2 - \beta^2)(k^2 + \beta^2)}{k^2[2\kappa \beta \cosh(\beta L) + (\kappa^2 + \beta^2) \sinh(\beta L)]}},$$

and we introduced the following notations $k = \sqrt{2mE/u} = \sqrt{2mV_0}$, and $\kappa = \sqrt{u^2 - k^2}$. The wave number $k$ is given by a root of the transcendental equation, $kL/\pi = 1 - (2/\pi) \arcsin(k/u)$. Equation (14) for $\chi_0(z)$ guarantees a continuity of the wave function and its derivative at interfaces $z = \pm L/2$. Parameter $u$ defines a natural length scale for the QW width, $L_0 = \pi/u$. It counts the number of levels in a QW: $[L/L_0] = n+1$.

In general, the solution for the variational parameter $\beta(F, L, u)$ can be found only numerically. One can show, however, that for a moderate field, $\beta$ is proportional to the electric field, $\beta = C(L)F$, where

$$C(L) = \frac{1}{\kappa^2} - \frac{1}{k^2} + \frac{L + L\kappa + L^2\kappa^2/6}{\kappa(2 + L\kappa)}. \quad (16)$$

The constant $C(L)$ introduces the natural scale for electric field units $F_0 = C(L)^{-3/2} = \ell^3$, where $\ell$ defines an average extension of wave function in a QW. Analytical expression for $C(L)$ can be obtained in two important limits: a very wide well, which can be approximated by an effective infinite QW (See Ref. 19) with $k = \pi/L$, $\kappa = u$, and a very narrow shallow QW, which can be described by a model of a $\delta$-functional QW potential with $k = u$, $\kappa = mV_0L$:

$$C_\infty = \frac{L^2}{2} \frac{\pi^2 - 6}{6\pi^2}, C_s = \frac{1}{2(mV_0L)^3}. \quad (17)$$

Using correlators, Eqs. (10) and (11), and the wave functions $\psi(r)$ and $\chi(z; \beta)$ we obtain the following expressions for alloy and interface roughness variances:

$$W_{\text{all}}^a = \frac{a_{\text{int}}^2}{8\pi\kappa^2} \frac{\chi^4}{m^2} \int_{-L/2}^{L/2} \chi^4(z; \beta) dz, \quad (18)$$

$$W_{\text{int}}^a = \frac{V^2_{\text{int}}}{\pi^2} G(y)[f_{11}x_0^4 + f_{22}x_0^4 - 2f_{12}x_0^2], \quad (19)$$

where $\chi_{LR} = \chi(\mp L/2; \beta)$. The function $G(y)$, defined as

$$G(y) = \int d^2\rho d^2\rho' \psi^2(\rho)\psi^2(\rho') \xi(\|\rho' - \rho\| m_2 M), \quad (20)$$

with $y = \sqrt{2}\sigma_{\perp} M/(\kappa m_2)$, depends on the lateral correlations of interface roughnesses, characterized by the in-plane correlation radius $\sigma_{\perp}$.

Since, for small $F$, parameter $\beta$ is proportional to the field, both Eqs. (18) and (19) can be expanded in terms of $F$:

$$W_{\text{all}}(F) = \Omega_{\text{all}}[\gamma^a_0 + \gamma^a_2 F^2], \quad (21)$$

$$W_{\text{int}}(F) = \Omega_{\text{int}}[\gamma^a_0 + \gamma^a_2 F + \gamma^a_2 F^2 F^2], \quad (22)$$

where

$$\Omega_{\text{all}} = \left[ k^2 a_{\text{all}}^2 x(1-x) \right] (\kappa^2 + L^2)^2 \kappa^2 m_2^2, \quad (23)$$

$$\Omega_{\text{int}} = \frac{4\kappa^2 k^4 V^2_{\text{int}}}{\pi^2} G(y)[(\kappa L + 2)^2 m_2^2 m_2^2], \quad (24)$$

and all other $\gamma_i$ are also monotonic functions of the QW width. Equations (21) and (22), which present the main results of the paper, show that in the range of parameters where the Stark width is exponentially small, there exists a strong power law field dependence of inhomogeneous exciton broadening caused by the field induced changes in the variance of the effective exciton potential.

FIG. 1. (Color online) Phase diagram $F-L$ of dominant contributions to the exciton linewidth: the gray shaded region—disorder induced mechanisms, the outside region—Stark broadening. Dashed lines are the shallow well and the infinite well approximations. The QW width is measured in units $L_0 = \pi/u$, which determines the number of levels in the QW, and $F_0$ defines the natural scale for electric field (see text).
The first remarkable feature of Eqs. (21) and (22) that we would like to point out is the presence of the linear-in-field term (see Fig. 2) in the interface roughness

\[ \gamma_1^{\text{lin}}(L) = 2LC(L)(f_{11} - f_{22}), \]

which results from the asymmetry between the two interfaces of the well, which manifest itself through different roughnesses, \( f_{11} \neq f_{22} \). In GaAs-based heterostructures, this asymmetry appears naturally because of the polar nature of GaAs, but it can also be engineered by preparing different interfaces under different growth conditions. The presence of the linear term gives rise to an interesting effect: one can switch between field-induced narrowing or broadening of the exciton line by simply changing the polarity of the applied field. This effect has a simple physical interpretation: if QW holes are pushed by the field toward a less disordered interface, the exciton line narrows, but it broadens in the opposite situation.

The quadratic in the field terms in Eqs. (21) and (22) also possess nontrivial properties (see Fig. 3). In the limit of shallow \( \delta \)-functional QWs, factors \( \gamma_2(L) \) can be presented as

\[ \gamma_2^{\text{aff}} = -2C_\delta^2(mV_0L)^2 - L^2/3, \]

\[ \gamma_2^{\text{int}} = -2C_\delta^2 \left( \frac{\gamma_0^{\text{int}}}{(mV_0L)^2} - 2L^2(\gamma_0^{\text{int}} + 4f_{12}) \right). \]

At small widths, \( L < L_{cr} \), the first term in square brackets in both equations dominates making respective contributions to the linewidth negative. In the opposite limit of an effective infinite wall, these factors are positive

\[ \gamma_2^{\text{aff}} = C_\delta^2 L^2 [1/3 - 3/\pi^2], \]

\[ \gamma_2^{\text{int}} = C_\delta^2 [\gamma_0^{\text{int}}(5/3 + 2/\pi^2) + 4f_{12}(1 + 2/\pi^2)]. \]

Different signs corresponding to the opposite limits mean that at some particular QW widths, factors \( \gamma_2(L_{cr}) \) vanish. Numerical analysis shows (see Fig. 3) that these “critical” widths are different for the alloy disorder and the interface roughness contributions. For parameters used in constructing Fig. 1, \( L_{cr} \) for the alloy disorder corresponds to \( L/L_0 \approx 1.5 \), and for the interface disorder to \( L/L_0 = 0.55 \). This means that we can effectively turn off the quadratic contribution from one of two sources of the inhomogeneous broadening by growing a QW with a width close to the respective critical value, \( L_{cr} \). In this case, all the field induced changes in the exciton broadening will be caused mostly by the other broadening mechanism. In principle, this can allow for an unambiguous discrimination between the alloy and the interface disorder contributions to the exciton linewidth. For example, growing a QW with a size \( L/L_0 \approx 1.5 \) and measuring field-induced changes in the exciton linewidth, we can guarantee that these changes originate from the interface roughness mechanism only, since the contribution from the alloy disorder mechanism vanishes up to the third order in field terms (see the dotted-dashed line in Fig. 4).

Different signs of \( \gamma_2 \) for shallow and deep QWs can be explained by a competition of two processes. On the one hand, the electric field pushes part of the wave function outside of the well and away from the influences of the disorders, promoting a narrowing of the exciton line. On the other hand, the field changes the shape of the wave function, pushing it towards an interface and slightly squeezing. The latter results in a greater localization of the wave function and, hence, broadens the exciton line. It is clear that the first process dominates for shallow QWs, while the second one prevails for QWs with larger widths.

To estimate quantitatively the critical QW widths we chose the example of In_{0.18}Ga_{0.82}As/GaAs. For this QW the material parameters are: \( m_r = 0.052m_0, m_x = 0.31m_0, \mu_r = 0.045m_0, U_0 = 79 \text{ meV}, E_B = 6.7 \text{ meV}, a_B = 16 \text{ nm} \). We obtained \( L_{cr}^{\text{all}} = 1.5 \text{ nm} \) and \( L_{cr}^{\text{int}} = 4.2 \text{ nm} \) for the critical interface and the alloy disorder lengths, respectively. Quantitative values for QWs made from different materials can be readily recalculated using the dimensionless values of the field and length in Figs. 1–4 and the following formulas for the electric field and QW lengths in standard units:

\[ L_{cr}^{\text{all}} = 0.3 \text{ nm}, \theta = 79 \text{ meV}, \theta_B = 6.7 \text{ meV}, a_B = 16 \text{ nm}. \]
In conclusion, we considered the effects of an electric field on the inhomogeneous linewidth of QW excitons. It is shown that the interface roughness can result in linear with respect to the field contribution to the exciton linewidth. This gives rise to the effect of switching between the narrowing and broadening of the exciton line by changing the field polarity. Quadratic contributions to the field dependence of the linewidth from both the alloy and the interface disorders are negative for shallow QWs, but change signs with the increase of the QW depth. These effects reveal a rich physics of inhomogeneously broadened excitons in an electric field, and can be used in the applications for narrowing exciton spectral lines and the experimental study of the roles of different mechanisms of the inhomogeneous broadening of excitons. We would like to stress that even though we considered a simplified model of a QW, the incorporation of such effects as valence-band mixing, nonparabolicity of the conduction band, dielectric constant, and effective mass mismatches would not change the qualitative results of the paper, which are related to the presence of two competing mechanisms affecting the exciton effective potential rather than to any particular model of the electron (hole) band structure or the confinement potential.

We are grateful to S. Schwarz for reading and commenting on the manuscript. We thank A. Efros for helpful discussions. The work is supported by AFOSR Grant No.F49620-02-1-0305 and PSC-CUNY grants.

FIG. 4. (Color online) The electric field induced change of the exciton variance $\Delta W^2_{\text{d} \text{a} \text{l} \text{o} \text{y}}$ for the alloy disorder contribution for three different QW widths: $L/L_0=3$ (solid line), $L/L_0=1.5$ (dotted-dashed line) and $L/L_0=0.3$ (dashed line). Curves are drawn only for the gray shaded area in Fig. 1. The contribution is negative for small thicknesses and positive at larger thicknesses. The critical value of the QW width is determined as a moment when the contribution changes its sign: $L_{\text{cr}}^{\text{alloy}} \approx 1.5L_0$.

$$L(\text{nm}) = a_B(\text{nm}) \frac{L}{L_0} \sqrt{\frac{\pi^2 \mu^* E_B}{2m_h V_0^2}},$$

$$F(\text{V/cm}) = 10^6 F_0 \frac{E_B(\text{meV})}{a_B(\text{nm})} \frac{\mu^* F}{2m_h V_0^2},$$

(31)

where $F_0$ can be extracted from Eq. (16).