Interface disorder and inhomogeneous broadening of quantum well excitons: Do narrow lines always imply high-quality interfaces?

I. V. Ponomarev,[a] L. I. Deych, and A. A. Lisyansky
Department of Physics, Queens College of the City University of New York, Flushing, New York 11367

(Received 5 April 2004; accepted 27 July 2004)

It is a commonly assumed that narrow lines in absorption or luminescence of quantum well excitons at low temperatures indicates high quality of quantum well interfaces. We show, that at least for narrow quantum wells, this is not always the case. Correlations between morphological fluctuations of two interfaces confining a quantum well, which were neglected in previous studies of exciton line shape, strongly suppress an inhomogeneous broadening due to interface disorder. *© 2004 American Institute of Physics*. [DOI: 10.1063/1.1793341]

Absorption and luminescence exciton spectroscopies are among the most informative tools for studying quantum wells (QW) as well as other semiconductor heterostructures. Therefore, a great deal of effort has been devoted to establishing the connection between spectral line shapes and the microscopic properties of QW structures. In particular, it has been established that low-temperature exciton linewidths in absorption and photoluminescence in QWs is predominantly inhomogeneous.[1] The shape of the spectra is determined by various types of disorder, and it is currently generally accepted that the spectral width in QW is directly related to the quality of interfaces, so that the luminescence spectra provide a quick and simple quality assurance tool for QW growth.[2] We show in this letter, however, that interwall correlations could significantly alter the quantitative relation between the exciton line width and quality of the interfaces.

It is usually assumed that fluctuations of QW interfaces are not correlated, so that their contribution to the total exciton line width is additive. At the same time there exist direct and convincing evidences that the morphological inhomogeneities of these interfaces are cross correlated. These correlations were seen by cross-sectional scanning tunneling microscopy,[3] scattering ellipsometry,[4] and x-ray reflection measurements.[5,6] Theoretically the importance of such correlations for physics of low-dimensional structures was first emphasized in Refs. 7–9, where conductivity of ultrathin metallic films was considered. Surprisingly, all existing theories of the inhomogeneous exciton linewidth unjustifiably neglect these correlations, although, as we show here, they significantly affect inhomogeneous broadening of exciton spectra. One of the important practical conclusions of this work is that, contrary to popular belief, narrow spectral lines do not always imply good quality interfaces but can be the result of a line narrowing effect of the interwall correlations.

QWs are generally heterostructures formed by a binary semiconductor (AB) and a ternary disordered alloy ($AB_{1-x}C_x$). There are two major disorder mechanisms responsible for the inhomogeneous broadening. One is compositional disorder caused by concentration fluctuations in a ternary component of the QW.[10,11] The other, which is the main object of our study, is the roughness of the QW walls caused by the formation of monolayer islands at the interfaces.[2,12–15] The main goal of this letter is to calculate the r.m.s fluctuation, W, of a random potential acting on QW excitons in the presence of interwall correlation. In order to establish the connection between our results and experimental observations, we will use the results of Refs. 1 and 16, where relations between W and the exciton linewidth, Δ, were extensively studied.

The r.m.s value of the random potential is defined as $W=\sqrt{\langle U_{eff}(\mathbf{R})^2\rangle}$, were U_{eff} is the effective potential acting on the center-of-mass (COM) exciton wave function due to interface disorder. An expression for this potential for a simplified model of a symmetric QW (both conduction and valence bands are nondegenerate and have an isotropic parabolic dispersion characterized by masses m_e and m_h, respectively) can be presented in the form $U_{eff}(\mathbf{R})=U_e(\mathbf{R})+U_h(\mathbf{R})$, where $U_{e,h}(\mathbf{R})$ is

$$U_{e,h}(\mathbf{R}) = V_{e,h}\chi^2_{e,h}(L/2)\int[\eta_1(\mathbf{R}\pm\beta_{h,e}\boldsymbol{\rho}) - \eta_2(\mathbf{R}\pm\beta_{h,e}\boldsymbol{\rho})]\phi^2(\boldsymbol{\rho})d^2\rho. \quad (1)$$

Here $V_{e,h}$ are the heights of the electron (hole) confining potentials, $\chi_{e,h}(z)$ are single electron (hole) wave functions, describing the quantized motion in the growth (z) direction, L is the well thickness, $\boldsymbol{\rho}=\boldsymbol{\rho}_e-\boldsymbol{\rho}_h$ and $\mathbf{R}=(m_e\boldsymbol{\rho}_e+m_h\boldsymbol{\rho}_h)/(m_e+m_h)$ are the relative and the COM coordinates of an electron-hole pair in the plane of the QW, $\phi(\boldsymbol{\rho})$ is a ground-state exciton wave function of in-plane relative motion, and $\beta_{h,e}=m_{h,e}/(m_e+m_h)$. Zero-mean random functions $\eta_{1,2}(x,y)$ characterize deviations of the ith interface from its average position. The presence of two functions, $\eta_{1,2}(x,y)$, distinguishes Eq. (1) from the corresponding equations of Ref. 12, where the roughness of a single interface was taken into account. The statistical properties of the interfacial roughness in multilayered systems can be characterized by the height-height correlation functions

$$\langle\eta_i((\boldsymbol{\rho}_1)\eta_j((\boldsymbol{\rho}_2)\rangle = h^2 f_{ij}\zeta(|\boldsymbol{\rho}_1-\boldsymbol{\rho}_2|), \quad (2)$$

where h is the average height of the interface inhomogeneity, and $\langle\ldots\rangle$ denotes the ensemble average. We assume that the dependence of both diagonal and nondiagonal correlations on the lateral coordinates $\boldsymbol{\rho}$ is described by the same function $\zeta(\boldsymbol{\rho})$. For the diagonal elements, $f_{ii}\equiv 1$, and the respective functions describe lateral correlation properties of a given interface (*self-correlation functions*). Nondiagonal elements

[a]Author to whom correspondence should be addressed; electronic mail: ilya@physics.qc.edu

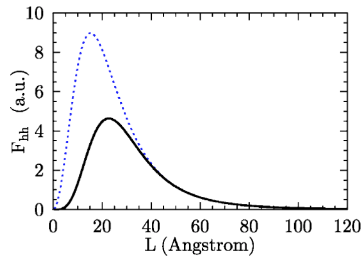

FIG. 1. (Color online) The dotted line shows $\chi_h^4(L/2)$ and the solid line presents function $F_{hh}(L)$. All calculations were made with parameters typical for $In_{0.12}Ga_{0.88}As/GaAs$ QW.

with $i \neq j$ introduce correlation between different interfaces; the respective quantity $f_{12}(L/\xi)$, which can be called a *cross*-or *vertical-correlation function*, is a function of the average width of the well and is characterized by the vertical correlation length ξ. In the limit $L/\xi \ll 1$ it is reasonable to assume that the interwall correlation function f_{12} tends to unity, which means that for a very small separation between the interfaces one random surface follows the contour of the other. In the opposite limit, $L/\xi \gg 1$, of a large well thicknesses f_{12} should vanish. The value of the vertical correlation length ξ depends on the growth process.

The r.m.s value of the random potential, W, is a sum of three terms $W \equiv T_{hh} + 2T_{eh} + T_{ee}$, where functions $T_{\alpha\beta}$ (indexes α and β take values e or h) can be presented as $T_{\alpha\beta}(L) = 2h^2 V_\alpha V_\beta F_{\alpha\beta} G_{\alpha\beta}$. Functions $F_{\alpha\beta}$, defined as,

$$F_{\alpha\beta}(L) = \chi_\alpha^2 \chi_\beta^2 [1 - f_{12}(L)], \tag{3}$$

determine the dependence of W on the thickness of the QW and are the main object of our attention.

$$G_{\alpha\beta} = \int d^2\rho d^2\rho' \phi^2(\rho)\phi^2(\rho')\zeta(|\beta_\beta \boldsymbol{\rho}' - \beta_\alpha \boldsymbol{\rho}|) \tag{4}$$

describe the effect of partial averaging of microscopic potentials by the lateral motion of the exciton. For QWs with a heavy hole and a light electron ($m_h \gg m_e$), the main contribution to the variance comes from the hole-hole correlator T_{hh}, since a rather massive hole averages only a small volume around the COM of the exciton, whereas a light electron is spread out over a much greater area of the order of the square of Bohr's radius, a_B^2. Let us consider the function F_{hh} in detail. The dependence of F_{hh} on L comes from two factors. The first is the fourth power of the electron QW wave function, χ_h^4, calculated at the interfaces, $z = \pm L/2$. This dependence for $In_{0.12}Ga_{0.88}As/GaAs$ QW is shown in Fig. 1 by the dotted line. In the limit of wide wells, when $L \gg 1/\kappa_{0h} = 1/\sqrt{2m_h V_h}$, $\chi_h(L)$ decreases with L according to $\chi_h^2 \sim 1/L^3$. In the opposite case of narrow QWs, $L \ll 1/\kappa_{0h}$, $\chi(L)$ behaves as $\chi_h^2 \approx \kappa_{0h}^2 L/2$. If one neglects vertical correlations, χ^4 determines[14] $W(L)$ with asymptotes given by these two equations and a maximum at $L \sim \kappa_{0h}$.

Interwall correlation, however, significantly modifies this behavior. First, it introduces a length scale, the vertical correlation length ξ, which is independent of κ_{0h}. For lengths smaller than ξ, the interwall correlation function behaves as, $1 - f_{12} \sim (L/\xi)^\gamma$, where γ is determined by the form of f_{12}. For example, if $f_{12}(L)$ is a Gaussian or Lorentzian, the parameter $\gamma = 2$, while for an exponential function f_{12}, $\gamma = 1$. For narrow QWs, for which L is smaller than both ξ and κ_{0h}, the r.m.s of the random potential W becomes

$$W \sim L^{2+\gamma}, \quad L < \xi, 1/\kappa_0, \tag{5}$$

and is strongly suppressed compared to the case in which vertical correlations are absent. Since κ_0 and ξ are independent parameters, the transition between the two asymptotic behaviors of χ and f_{12} also occurs independently of one another. Experimental data suggest that there are situations in which $\xi \gg \kappa_0$. In this case, interwall correlation affects not only the $L \to 0$ asymptotic of W, but also its behavior for $L \gg \kappa_0$. Instead of a $1/L^3$ behavior one would have a much slower decrease of W with L: $W \propto L^{\gamma-3}$. This result is obtained under the assumption of positive interwall correlations, $f_{12} > 0$. Theoretically, a negative sign of f_{12} is also possible.[7,9] In this case, instead of the narrowing of exciton spectra, the intercorrelations would result in extra broadening. It appears, however, on the basis of structural studies,[3–6] that in known systems the correlations are positive.

The presented analysis demonstrates, that the vertical correlation function f_{12} can significantly shift the position of the maximum of $W(L)$ as well as change its height and shape. Experimentally observed dependences $W(L)$ are characterized by a great variety of positions, widths, and heights of the maxima,[17,18] which cannot be explained by existing theories even when compositional disorder is taken into account. Since the vertical correlation length is strongly dependent on the growth conditions of the QW, interwell correlations can naturally explain the full range of experimental results. The graph of the function F_{hh} with interwall correlations taken into account is shown in Fig. 1. One can see that these correlations indeed significantly affect the shape of this function. While we only discussed properties of the hole-hole correlator, it is clear that the behaviors of the electron-electron and hole-electron terms are similar.

In order to compare calculations of W with experimental absorption spectra, the dynamics of excitons in a random potential should be evaluated for various correlation properties. Exciton absorption in the situations under consideration can be reliably described[16] by the semiclassical theory, in which the exciton inhomogeneous line shape is given by a Gaussian with width $\Delta = 2\sqrt{2\ln 2}(W_{comp}^2 + W_{int}^2)^{1/2}$, where W_{comp} takes into account alloy disorder. Using the results of Refs. 1, 11, 16, and 19 in conjunction with our analysis of $W(L)$, we can find the interface contribution to Δ. However, in order to fit this function to experimental data we also need to take compositional disorder into consideration. R.m.s of the disorder potential due to compositional disorder was calculated in Ref. 12, while theories of exciton broadening in this case were given in Refs. 11 and 12. Unfortunately, all existing theories are in a strong quantitative disagreement with experimental results. It is not the goal of this letter to uncover the causes of this discrepancy. However, common wisdom tells us that in the limit of very wide QW only the alloy disorder contribution should survive. The simplest way to adjust the theories is to scale alloy disorder contribution to the linewidth, Δ_{comp}, to the value that would coincide with experimental results in the limit of large L. The results of the best fit performed in this way are shown in Fig. 2. We were able to achieve a similarly good fit to the experimental data obtained for other material systems[17] as well. This fit would not be possible at all without incorporating interwall correlation even if alloy disorder scaling is used as an additional fitting parameter. We conclude, therefore, that interwall cor-

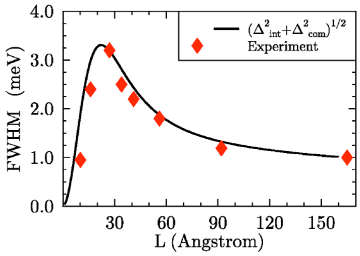

FIG. 2. (Color online) The thick solid line is the best fit for the total linewidth displayed together with experimental results (diamonds) from Ref. 18.

relation must be taken into account to properly interpret experimental results.

In conclusion, we have shown that the presence of positive interwall correlation strongly suppresses the interface disorder contribution to inhomogeneous broadening. The main consequence of this finding is that narrow exciton lines do not necessarily imply that the grown interfaces are of very good quality. We also found that the differences in interwall correlation lengths can account for the variety of positions, strengths, and sharpness of the full width at half maximum dependence on the well width for experimental data obtained by different research groups.

The work is supported by AFOSR Grant No F49620-02-1-0305.

[1] A. L. Efros and M. E. Raikh, *Optical Properties of Mixed Crystals* (North-Holland, Amsterdam, 1988).
[2] C. Weisbuch, R. Dingle, A. Gossard, and W. Wiegmann, Solid State Commun. **38**, 709 (1981).
[3] Y. Yayon, A. Esser, M. Rappaport, V. Umansky, H. Shtrikman, and I. Bar-Joseph, Phys. Rev. Lett. **89**, 157402 (2002).
[4] T. A. Germer, Phys. Rev. Lett. **85**, 349 (2000).
[5] V. Holý and T. Baumbach, Phys. Rev. B **49**, 10668 (1994).
[6] E. A. Kondrashkina, S. A. Stepanov, R. Opitz, M. Schmidbauer, R. Kohler, R. Hey, M. Wassermeier, and D. V. Novikov, Phys. Rev. B **56**, 10469 (1997).
[7] A. E. Meyerovich and A. Stepaniants, Phys. Rev. B **58**, 12242 (1998); *ibid.* **60**, 9129 (1999).
[8] A. E. Meyerovich and A. Stepaniants, J. Phys.: Condens. Matter **12**, 5575 (2000).
[9] A. E. Meyerovich and I. V. Ponomarev, Phys. Rev. B **65**, 155413 (2002).
[10] S. Baranovskii and A. Efros, Sov. Phys. Semicond. **12**, 1328 (1978).
[11] A. Efros, C. Wetzel, and J. Worlock, Phys. Rev. B **52**, 8384 (1995).
[12] R. Zimmermann, F. Grosse, and E. Runge, Pure Appl. Chem. **69**, 1179 (1997).
[13] R. Zimmermann, Phys. Status Solidi B **173**, 129 (1992).
[14] J. Singh and K. K. Bajaj, J. Appl. Phys. **57**, 5433 (1985).
[15] H. Castella and J. W. Wilkins, Phys. Rev. B **58**, 16186 (1998).
[16] E. Runge, in *Solid State Physics*, edited by. H. Ehrenreich and F. Spaepen (Academic, San Diego, 2002).
[17] A. Patanè, A. Polimeni, M. Capizzi, and F. Martelli, Phys. Rev. B **52**, 2784 (1995); P. B. Kirby, J. A. Constable, and R. S. Smith, *ibid.* **40**, 3013 (1989); J.-P. Reithmaier, R. Höger, and H. Riechert, *ibid.* **43**, 4933 (1991).
[18] D. Bertolet, J.-K. Hsu, K. M. Lau, E. Koteles, and D. Owens, J. Appl. Phys. **64**, 6562 (1988).
[19] M. E. Raikh, N. N. Ablyazov, and A. L. Efros, Sov. Phys. Solid State **25**, 199 (1983).