Exciton line broadening by compositional disorder in alloy quantum wells

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Spectral lines of photoluminescence in $ZnSe_xTe_{1-x}$/ZnTe single quantum wells (QW)'s are studied experimentally and theoretically. Special attention is given to the linewidth as a measure of the QW quality. The analysis of band offsets shows that the structures under study are type-II QW's, where electron levels are quantized inside a QW, whereas holes are situated in the barriers. The spectral linewidth depends rather slightly on the thickness of QW's, ruling out fluctuations of QW thickness as the dominant broadening mechanism. On the other hand, the theory of line broadening by compositional disorder fits the experimental data well without any adjustable parameters, suggesting that it is the compositional disorder that dominates the line broadening in these systems.

I. INTRODUCTION

One of the most important problems connected with the growth of heterostructures and, in particular, quantum wells (QW's), is the structural disorder present in such systems. The disorder determines to a large extent the properties of devices based on heterostructures such as lasers, high-speed transistors, photodetectors, etc.,[1] and the study of the disorder is necessary for improving the quality of heterostructures. One of the most powerful tools for studying the structural disorder in QW's is photoluminescence (PL). The quantity of primary interest is the inhomogeneous width of PL spectral lines, because it is this quantity that is determined by the structural disorder. As has been shown by many investigations,[1,2] fluctuations of the QW thickness (so-called interface roughness) play a very important role in QW's based on III-V semiconductors, in particular in the case when the QW is built on the basis of a binary compound. This kind of disorder could be, at least theoretically, eliminated by improving the preparation procedure.

An additional mechanism of disorder leading to PL line broadening arises in QW's based on alloys. Fluctuations of the alloy composition in space cause potential fluctuations, which influence the behavior of photoexcited carriers. These fluctuations can localize not only electrons or holes, but also excitons as a whole.[3] It is known that the compositional fluctuations have a strong influence on the exciton luminescence in bulk systems and, in particular, in II-VI compounds.[3–5] The role of this kind of disorder in QW's is therefore of great interest. In order to investigate the influence of compositional disorder on the PL properties in alloy QW's we carried out experimental and theoretical studies of PL in single QW's, based on the alloy material $ZnSe_{0.3}Te_{0.7}$ with barriers of ZnTe. Comparison of our experimental data with theoretical results shows that PL spectral lines are broadened by compositional disorder and not by the interface roughness. The theory of line broadening by compositional disorder yields a favorable fit for the experimental data without any adjustable parameters.

In Sec. II, the preparation procedure of the samples is described and experimental data for the PL measurements are summarized. In Sec. III the theoretical description of PL lines inhomogeneously broadened by various kinds of disorder in QW's is presented. The comparison of the theoretical results with the experimental data shows that the alloy disorder is quite probably responsible for the line broadening while the mechanism of interface roughness fails to account for the experimental data. Concluding remarks are given in Sec. IV.

II. EXPERIMENTAL RESULTS

The studied structures of QW's formed by thin $ZnSe_xTe_{1-x}$ alloys and thick ZnTe barriers (Fig. 1) were grown by atmospheric pressure metalorganic-vapor-phase epitaxy (MOVPE) on (100)-GaAs substrates. We have used di-isopropyltelluride (DiPTe) and diallyselenide (DASe) as metalorganic precursors in combina-

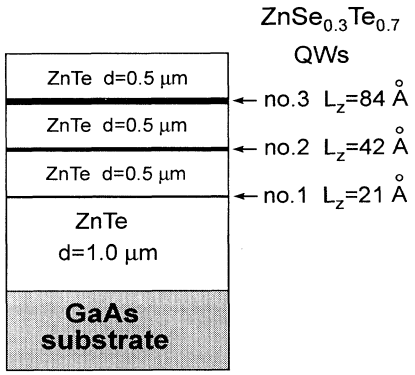

FIG. 1. Schematic picture of the sample structure with three $ZnSe_{0.3}Te_{0.7}$ QW's.

tion with dimethylzinc-triethylamine (DMZn:TEN) diluted in H_2 gas. The growth was performed at $T=340\,^{\circ}C$ with 10-min interruptions at the interfaces. The composition of the mixed crystal was derived from the lattice constant of the $Zn_xSe_{1-x}Te$ epilayer (with the thickness of about 1.5 μm) using Vegard's law. The quantum-well thicknesses were estimated from the growth rate and were determined from high-resolution electron-microscope (HRTEM) analysis. Detailed growth conditions and characterization of $ZnSe_xTe_{1-x}$ mixed crystals and QW's will be published elsewhere.[6] After the optimization of the growth conditions the best structural and optical quality was achieved in the sample containing three $ZnSe_{0.3}Te_{0.7}$ QW's with thicknesses $L_z=21$, 42, and 84 Å separated by 0.5-μm-thick ZnTe barriers. The optical spectra were measured in a backscattering geometry when the samples were mounted in a helium bath cryostat at $T=2$ K. The photoluminescence spectra were obtained under the Ar-ion laser lines (457.9 and 514.5 nm) excitation (above ZnTe barriers) or using a continuously pumped dye laser operated with Rhodamin 110 in the spectral region of 530–600 nm for the excitation below the energy gap of ZnTe. The PL spectra were recorded with a Jarrel-Ash 1-m double spectrometer and a thermoelectrically cooled GaAs photomultiplier.

The studied structures with $ZnSe_{0.3}Te_{0.7}$ show a bright yellow-red photoluminescence at low temperatures. The barriers of ZnTe are sufficiently thick (0.5 μm) and we assume that there is no interaction between different quantum wells. The emission spectra of the sample under study with the excitation above and below the gap of the ZnTe barrier are shown in Fig. 2. Three main peaks are attributed to the recombination of carriers in three QW's with different thicknesses. Two lines at 1.972 and 2.025 eV dominate the PL spectrum under the excitation above

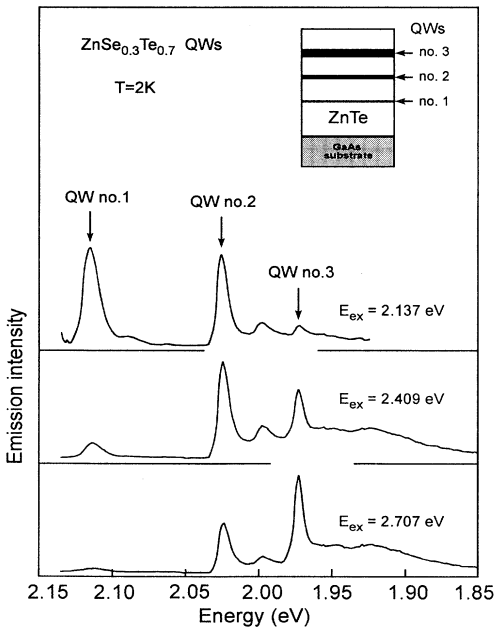

FIG. 2. Emission spectra of the studied structure with three QW's at different excitation energies.

the band gap of the ZnTe barrier. These lines correspond to the radiative recombination in the QW's Nos. 2 and 3 (Fig. 2) with thicknesses $L_z=84$ and 42 Å. Intensity of the PL from the QW No. 1 with $L_z=21$ Å under this excitation is much smaller because of a strong absorption of the exciting light in the ZnTe barrier. We have obtained an increase of the relative intensity of the PL peak at 2.115 eV from this QW using the dye-laser excitation below the energy gap of ZnTe. Under these conditions the excitation is almost homogeneous through the sample thickness and the relative intensities of PL lines reflect the probabilities of recombination in the QW's with different thicknesses.

The spectral position of the recombination lines in QW's shifts to higher energies with the decrease of the QW thickness due to the confinement effect in the $ZnSe_xTe_{1-x}$ quantum well. At the same time the half-width of the emission line monotonically increases from 7 to 12 meV. The half-width of the main peak in our sample is much smaller than that reported for $ZnSe/ZnSe_xTe_{1-x}$ strained-layer superlattices[7] and indicates a high quality of the grown $ZnSe_{0.3}Te_{0.7}$ QW structures.

In order to identify the type of transitions in these structures, let us consider the band alignment in the $ZnSe_{0.3}Te_{0.7}$ system. The position of the valence-band edge changes almost linearly[8] with composition x and the valence-band offset between ZnSe and ZnTe is about 0.9 eV, the position of the valence-band maximum of ZnSe being below that of ZnTe.

In order to find the offset for the conduction band in our sample, we need the compositional dependence of the band gap in addition to that of the valence-band maximum. The variation of the band gap with composition for molecular-beam epitaxy grown $ZnSe_xTe_{1-x}$ epilayers was studied in Ref. 9, where it was found that

$$E_g(x)=E_g(1)x+E_g(0)(1-x)-bx(1-x) \tag{1}$$

with the bowing parameter $b=1.504$ eV and with the band gaps of binary compounds $E_g(0)=2.40$ eV and $E_g(1)=2.82$ eV.

Concentration dependence for the conduction-band minimum can be written as

$$E_c(x)=E_v(x)+E_g(x)\ , \tag{2a}$$

where

$$E_v(x)=\text{const}+\Delta E_v(1-x)\ . \tag{2b}$$

The compositional variation of both the band edges is presented in Fig. 3. The variation of the band gap $E_g(x)$ of the $ZnSe_xTe_{1-x}$ mixed crystals is also shown in this picture. On the basis of these concentration dependences we deduce a band alignment in the structure containing $ZnSe_{0.3}Te_{0.7}$ as a quantum-well material. Obtained band alignment in $ZnSe_{0.3}Te_{0.7}/ZnTe$ structures (Fig. 4) corresponds to the type-II recombination between electrons on quantized levels in a QW and holes near the valence-band edge of the ZnTe barrier. We believe that the obtained band alignment is qualitatively correct and we have the type-II recombination indeed. It is difficult, however, to

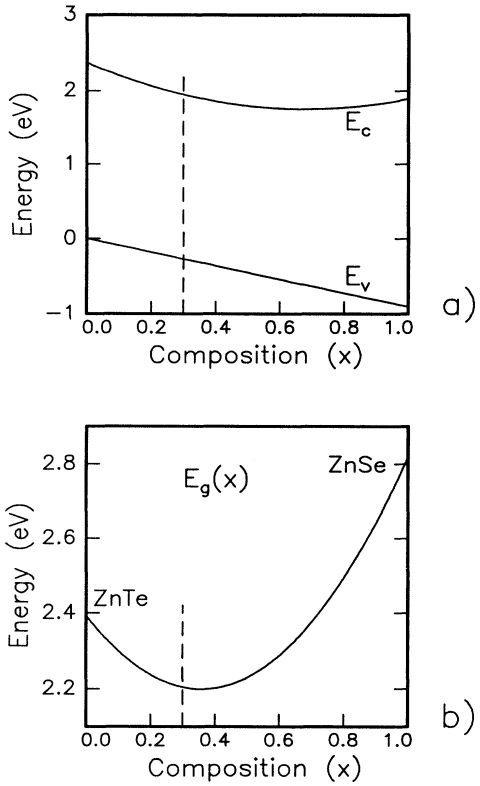

FIG. 3. Concentration dependence of the band edges (a) (Ref. 8) and the band gap (b) (Ref. 9) in $ZnSe_xTe_{1-x}$ mixed crystals.

find the exact values of all energetic alignments in our system. The reason is the unknown values of the deformation coefficients for each band and the unknown magnitude of the strain effect. Indeed, the large lattice mismatch between the $ZnSe_{0.3}Te_{0.7}$ layer and the ZnTe barrier (about 2%) makes strain effects play an important role in determining the band-edge positions. The energy shifts of band-edge positions due to the lattice mismatch can be of the order of 100–150 meV. This, however, does not change the type of alignment in this system. For accurate description of the band structure of strained-layer QW's it is necessary to know the degree of strain relaxation in each QW layer and the exact value of the valence-band offset for the ZnSe/ZnTe system, which is still under discussion.[10]

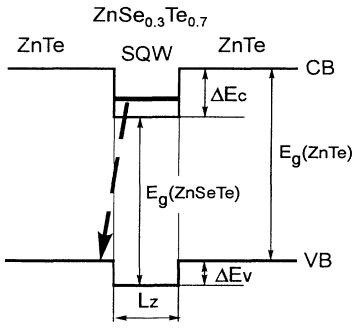

FIG. 4. Band alignment in $ZnSe_{0.3}Te_{0.7}$/ZnTe structure.

Only the depth V of the QW in the conduction band will be important for our further considerations. We calculate this depth to be $V \approx 0.45$ eV by fitting the positions of the PL peaks using the routine procedure for calculating quantized energies in QW's with finite barriers. This calculation is valid if the QW layers are below the critical thickness for strain relaxation, which we assume to be the case. This assumption is supported by HRTEM measurements, where no misfit dislocations were observed at the interface between the $ZnSe_{0.3}Te_{0.7}$ QW and the ZnTe barrier.

The aim of this paper is to study the influence of a disorder on the PL spectral lines. This influence is mostly pronounced on the width of spectral lines, therefore in the rest of the paper we will discuss the linewidth and its dependence on the QW thickness and pay less attention to the position of spectral lines. In Fig. 5 the PL linewidths δ^{exp} for three QW's are shown as a function of the QW thickness L_z. Under the linewidth we mean the full width at half maximum of the photoluminescence spectral lines. This quantity is the subject of the following theoretical consideration.

III. THEORETICAL CALCULATION OF THE LINEWIDTH

A theory of the exciton PL in disordered bulk systems has been developed in Ref. 3, where the localization of excitons by potential fluctuations has been suggested and a quantitative calculation of the PL linewidth has been performed. The theory can be applied to the exciton photoluminescence in heterostructures and QW's as well.

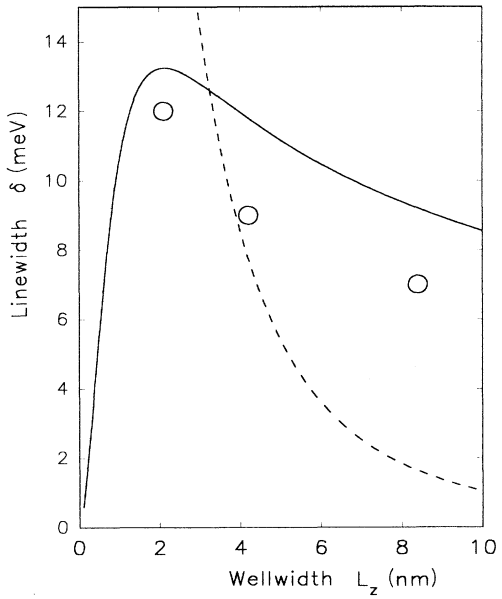

FIG. 5. Experimental data (open circles) and theoretical curves for the full width at half maximum of the main line in $ZnSe_{0.3}Te_{0.7}$ quantum wells as a function of the well thickness L_z. Dashed line: Dependence $V_e(L_z)$ for interface roughness. Solid line: Exciton line broadening by compositional disorder inside the quantum well.

We describe this theory for bulk material and proceed by modifying it for QW's.

Let $m_e, \mathbf{r}_e$ and $m_h, \mathbf{r}_h$ be the electron and hole masses and coordinates, and $V_e(\mathbf{r}_e)$ and $V_h(\mathbf{r}_h)$ the fluctuating potentials acting on the electron and the hole, respectively. The Schrödinger equation for the exciton wave function has the form

$$\left[-\frac{\hbar^2}{2M}\nabla_R^2-\frac{\hbar^2}{2\mu}\nabla_r^2-\frac{e^2}{\kappa r}+V_e\left[\mathbf{R}+\frac{m_h}{M}\mathbf{r}\right]-V_h\left[\mathbf{R}-\frac{m_e}{M}\mathbf{r}\right]\right]\psi(\mathbf{R},\mathbf{r})=E\psi(\mathbf{R},\mathbf{r}) , \tag{3}$$

where $\mathbf{R}=(m_e\mathbf{r}_e+m_h\mathbf{r}_h)/M$ and $\mathbf{r}=\mathbf{r}_e-\mathbf{r}_h$ are the exciton center-of-mass coordinate and the coordinate of the relative motion, respectively, $M=m_e+m_h$, $\mu=m_e m_h/(m_e+m_h)$. We suppose that the linewidth we are interested in is small compared to the exciton binding energy E_{exc}. We can then factorize the wave function as $\psi=\varphi(\mathbf{r})\chi(\mathbf{R})$, where φ satisfies the equation

$$\left[-\frac{\hbar^2}{2\mu}\nabla_r^2-\frac{e^2}{\kappa r}\right]\varphi=-E_{\text{exc}}\varphi , \tag{4}$$

which describes the relative motion of the electron and the hole. Substituting this wave function into Eq. (3), multiplying by $\varphi^*(\mathbf{r})$, and integrating over $\mathbf{r}$, using Eq. (4), we find the equation for $\chi(\mathbf{R})$,

$$\left[-\frac{\hbar^2}{2M}\nabla_R^2+V_e'(\mathbf{R})-V_h'(\mathbf{R})\right]\chi(\mathbf{R})=(E-E_{\text{exc}})\chi(\mathbf{R}) , \tag{5}$$

where

$$V_e'(\mathbf{R})=\int V_e\left[\mathbf{R}+\frac{m_h}{M}\mathbf{r}\right]|\varphi(\mathbf{r})|^2 d^3r , \tag{6}$$

$$V_h'(\mathbf{R})=\int V_h\left[\mathbf{R}-\frac{m_e}{M}\mathbf{r}\right]|\varphi(\mathbf{r})|^2 d^3r . \tag{7}$$

This yields the equation for a particle of mass M in a fluctuating potential. We suppose that the kinetic energy of the exciton is so small that the exciton is localized by a potential fluctuation. The broadening of the exciton line occurs because the exciton localization energy is different at different points in the sample.

Before discussing the disorder in QW's it is worth remarking about treatments of compositional disorder in bulk systems. It has been shown in Ref. 3 that for compositional disorder the spatial size of the fluctuations that influence the movement of electrons and holes is of the order of the de Broglie wavelength $\lambda=\hbar/(mE_0)^{1/2}$, where E_0 is the energy scale of the band-edge smearing due to the disorder. For holes with $m_h \gg m_e$ the size of the fluctuations is much less than that for electrons and hence the random potential of the compositional disorder influences the movement of holes much stronger than the movement of electrons.[3] From Eq. (7) it is seen that for most systems with $m_e \ll M$ the potential $V_h'(\mathbf{R})$ acting on the exciton is equal to the potential $V_h(\mathbf{R})$ acting on the hole, i.e., the potential fluctuations of the valence-band edge influence the exciton movement without any averaging. Hence it is the de Broglie wavelength of holes which determines the spatial size of fluctuations affecting the excitons most effectively[3] and not the exciton volume determined by its Bohr radius as was claimed in many publications.[11]

Now let us discuss a possible mechanism responsible for the potential fluctuations in the QW's under study. As long as we consider the disorder inside a quantum well, we shall concentrate our discussion on the term $V_e(\mathbf{r}_e)$, because only electrons are situated inside the well of our type-II QW structures. The holes inside the barriers are influenced by the quantum-well disorder to a much smaller extent and we shall ignore the term $V_h(\mathbf{r}_h)$ in the following consideration.

Disorder in QW's is mostly discussed in terms of fluctuations of the QW thickness.[1,2,12] For a QW with infinitely high barriers, V_e has the form

$$V_e=\frac{\hbar^2}{m_e}\frac{\pi^2}{L_z^2}\frac{\delta L_z}{L_z} , \tag{8}$$

where δL_z is the change of the QW thickness in the fluctuation. In order to treat a QW with finite barriers, one has to solve the corresponding quantum-mechanical problem.[1] The parameters of the problem are the effective mass of carriers m_e, the thickness L_z, the fluctuation of the thickness δL_z, and the energetic depth V of a QW. In Fig. 5 we show the results of such a calculation for V_e when the potential is caused by a single-monolayer fluctuation $\delta L_z=3$ Å as a function of L_z for the QW of $ZnSe_{0.3}Te_{0.7}$. In this calculation we used the value $m_e=0.132m_0$ for the electron effective mass obtained by a linear extrapolation between the values of m_e for ZnSe and ZnTe. The depth V of the QW's was assumed to be 0.45 eV (see above). It is seen that whereas V_e is much bigger than the observed linewidth δ^{exp} for the narrowest QW with $L_z=21$ Å, it is much smaller than the linewidth δ^{exp} for the widest QW with $L_z=84$ Å. From Eqs. (5) and (6) it is clear that the potential energy which influences an exciton is not V_e itself, but must be obtained from V_e by the corresponding averaging with the wave function of the relative motion of the electron. The interface roughness does not lead to disorder in the growth direction, hence the averaging is important in the lateral directions only. If the lateral size d of the thickness fluctuation is smaller than the exciton Bohr radius a_B, this averaging yields a value of V_e' much smaller than V_e. The typical value of the potential energy in Eq. (5) will then be of the order of

$$V_e' \simeq V_e(d/a_B)^2 . \tag{9}$$

Thus by an appropriate choice of $d<a_B$ one can fit the δ^{exp} value for the well with $L_z=21$ Å, where δ^{exp} is much smaller than V_e. If only such fluctuations were present in the system we would obtain δ^{theor} always smaller than V_e as described above. However, the value of δ^{exp} for the

widest QW with $L_z = 84$ Å is obviously larger than the corresponding value of V_e (see Fig. 5), so this mechanism cannot account for the observed results at all. In this discussion we compare the linewidth δ^{exp} with the typical value of the potential energy V_e'. In fact, δ^{exp} is to be compared with the spread of energies obtained by solving a Schrödinger equation for excitons in the potential fluctuations[3] V_e'. But if the typical value of V_e' is much smaller than δ^{exp}, the spread of energies obtained by solving the Schrödinger equation is much smaller than δ^{exp} as well. That is obviously the case for a QW with $L_z = 84$ Å. Roughly speaking, the L_z dependence of δ^{theor} in the case of these QW thickness fluctuations is too strong to account for the experimental data, and we may claim that the interface roughness does not play a dominant role in the observed line broadening. Analogous results have been recently obtained[13] for QW's $ZnSe/ZnS_xSe_{1-x}$ and $Zn_xCd_{1-x}Se/ZnS_xSe_{1-x}$, so it seems to be a general property of QW's on the basis of II-VI semiconductors.

Therefore we now turn to a different kind of disorder in alloy QW's, namely compositional disorder. In the case of compositional disorder the random potential $V(\mathbf{r})$ can be represented by a white-noise potential with the correlation function[14]

$$\langle V(\mathbf{r})V(\mathbf{r}')\rangle = \gamma\delta(\mathbf{r}-\mathbf{r}') , \tag{10}$$

where $\langle\ \rangle$ denotes averaging over different configurations of atoms, and

$$\gamma = \alpha^2 x(1-x)/N , \tag{11}$$

where x is the composition of the alloy, N is the concentration of lattice sites occupied by the alloy atoms, and $\alpha = dE_c/dx$. The parameter α characterizes the shift of the bottom of the conduction band with composition. In Ref. 3 localization of excitons by this kind of disorder has been studied in detail. There are two regimes for the spatial scale R_l of potential fluctuations localizing excitons. If $R_l \gg a_B$ the averaging in (6) is not important and $V_e'(\mathbf{R}) \approx V_e(\mathbf{R})$. However, usually[14] R_l is less than α_B and in order to obtain the potential $V_e'(\mathbf{R})$ acting on the exciton, one has to use the averaging procedure (6). In this case the smoothing of small-scale fluctuations becomes important, and the exciton moves in the potential whose minimum spatial length scale is of the order of a_B. In order to perform this averaging, one has to know the wave function $\varphi(\mathbf{r})$ for the relative motion of the electron and the hole. We approximate this function by[15]

$$\varphi(r) = C\tilde{\chi}(z)\exp(-\rho/a_B) , \tag{12}$$

where ρ is the lateral distance between the electron and the hole and z is the electron coordinate perpendicular to the interface. The function $\tilde{\chi}(z)$ is the single-particle ground-state wave function in a QW,

$$\begin{aligned} &\tilde{\chi}(z) = A\exp(-k_b|z|) \quad \text{for } |z| > L_z/2 , \\ &\tilde{\chi}(z) = B\cos(k_w z) \quad \text{for } |z| < L_z/2 , \end{aligned} \tag{13}$$

where A, B, C are the normalization coefficients and

$$k_w = (2m_e E_e)^{1/2}/\hbar, \quad k_b = [2m_e(V-E_e)]^{1/2}/\hbar ,$$

V being the depth of the well for the conduction band (0.45 eV) and E_e is the position of the first quantum level with respect to the bottom of the well. Using the continuity conditions for the wave function and its derivative at the interfaces and the normalization of the wave function (12), we obtain for the mean-square-root fluctuation W of the potential

$$W^2 = \langle (V_e'(R))^2\rangle = \frac{\gamma}{8\pi}\left[\frac{M}{m_h a_B}\right]^2 k_w \times \frac{6x + 8\sin(x) + \sin(2x)}{[x + \sin(x) + 2(k_w/k_b)\cos^2(x/2)]^2} \quad \text{with } x \equiv L_z k_w . \tag{14}$$

Since the potential $V_e'(R)$ is proportional to the Gaussian fluctuation of the composition, the potential distribution function must also be a Gaussian,

$$F(V) = \frac{1}{\sqrt{2\pi}W}\exp(-V^2/2W^2) . \tag{15}$$

Assuming that the fluctuations are classical, i.e., a potential well contains many levels, we come to the conclusion that the distribution function of exciton energies due to localization by compositional fluctuations is a Gaussian as well with the parameter W calculated above. The important parameters to determine the value of W are α, N, x, and a_B, which appear in Eq. (14). The values of parameters N and a_B in $ZnSe_{0.3}Te_{0.7}$ are obtained from linear extrapolations between the corresponding values for ZnSe and ZnTe. Using the values $\alpha = 1$ eV, $x = 0.3$, $N = 1.88 \times 10^{22}$ cm^{-3}, and $a_B = 52$ Å, we obtain from Eq. (14) the full width at half maximum δ^{theor} of the distribution (15). The dependence of δ^{theor} on L_z is shown in Fig. 5. It appears to be rather weak in good agreement with experimental data. At small values of L_z the wave function (12) more strongly penetrates into the binary barriers, where there is no compositional disorder, and the value of W decreases. This effect has been recently observed.[13] It has not been taken into account in our calculation that the Bohr radius decreases with the decrease of L_z because this dependence in type-II QW's is not known well enough.[16] At this level of accuracy the main conclusion from this theoretical consideration is that the theoretical values δ^{theor} obtained in consideration of exciton localization by compositional disorder without any adjustable parameters are close to the experimentally observed values of the linewidths δ^{exp}.

IV. CONCLUSIONS

High-quality quantum-well structures formed by thin $ZnSe_xTe_{1-x}$ alloys and thick ZnTe barriers have been grown by metalorganic-vapor-phase epitaxy. The analysis of band offsets shows that these structures are type-II quantum wells. The photoluminescence is due to radiative recombination of excitons, formed by holes in the barriers close to the interfaces and quantized electrons inside the quantum wells. The inhomogeneous width of spectral lines characterizes the structural disorder present in our samples. Two mechanisms of the line

broadening by structural disorder are analyzed theoretically. The interface roughness gives a much more pronounced dependence of the linewidth on the well thickness than that observed experimentally. We conclude that the roughness is not responsible for the line broadening in our samples. On the other hand, our theoretical investigation of the line broadening due to compositional disorder inside the quantum well fits the observed linewidths sufficiently well without any adjustable parameters, manifesting that this mechanism dominates the line broadening. Note, however, that this result does not rule out the presence of interface roughness in our systems at all. Rather, it suggests that excitons recombine from regions of quantum wells having a particular thickness. These regions are probably those with the lowest dimensional quantization levels. Excitons can relax into these regions during their lifetime, which is sufficiently large in the type-II quantum wells. The problem of exciton migration in the type-II quantum wells needs further investigation.

ACKNOWLEDGMENTS

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