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Linköping University Post Print

N.B.: When citing this work, cite the original article.

Original Publication:

Ivan Shtepliuk, Volodymyr Khranovskyy and Rositsa Yakimova, Effect of Zn-Cd interdiffusion on the band structure and spontaneous emission of ZnO/Zn_{1-x}Cd_xO/ZnO quantum wells, 2015, Superlattices and Microstructures, (85), 438-444.

<http://dx.doi.org/10.1016/j.spmi.2015.06.013>

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Postprint available at: Linköping University Electronic Press

<http://urn.kb.se/resolve?urn=urn:nbn:se:liu:diva-121305>

Effect of Zn-Cd interdiffusion on the band structure and spontaneous emission of ZnO/Zn_{1-x}Cd_xO/ZnO quantum wells

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Abstract

Needs in more-efficient visible light sources based on quantum wells (QWs) requires the diversification of traditional optoelectronics' materials as well as development of the cost-effective approaches for reliable quantum confinement engineering. Interdiffusion approach has a great potential to become a simple method for controlling the optical properties of QWs and diminishing the quantum confined Stark effect (QCSE). In this work we theoretically study the effect of Zn-Cd interdiffusion in ZnCdO/ZnO QWs on their band structure, optical matrix elements and spontaneous emission properties. The QW intermixing leads to improving both the transverse electric (TE) and transverse magnetic (TM) optical matrix elements due to enhancement of the overlap integral between electron and hole wave functions and modification of the confinement potential from triangle-shaped to parabolic-like. The optimized diffusion length 4 Å provided by the annealing at 700 K during 60 s was determined for 2 nm-thick Zn_{0.85}Cd_{0.15}O QW, which offers higher spontaneous emission rate in comparison to conventional QW. The reasonable interpretation of the interdiffusion effect on the optical properties of QWs is proposed in terms of low diffusion length and high diffusion length regimes. Thus, suitable combination of annealing duration and annealing temperature with the geometrical/compositional parameters of QWs can be the efficient way for improving the optical performance of ZnO-based QWs.

Key words: interdiffusion, quantum well, ZnCdO alloy, spontaneous emission, matrix elements

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1. Introduction

The conventional type-I $\text{Zn}_{1-x}\text{Cd}_x\text{O}$ quantum well (QW) with ZnO barriers have gained considerable attention due to prospective applications in optoelectronics [1-10]. One of the major challenges that prevent high performance of $\text{ZnO}/\text{Zn}_{1-x}\text{Cd}_x\text{O}/\text{ZnO}$ QW is the large spontaneous and piezoelectric polarization in the QW, which induces the low electron and hole wave functions overlap, especially for QW with high cadmium content and thick QW active layer as well as the redshift in transition energy (well-known QCSE) [2, 7]. Therefore, the main efforts of investigations should be focused on a searching the ways of the effective suppression of the carrier separation effect. To our best knowledge, there are several approaches, which were proposed to suppress the polarization induced effects in type-I QWs, such as (i) layer growth on non-polar and semi-polar substrates [11-13], (ii) formation of the staggered QW [14], (iii) using the embedded delta-doped layer [15], formation of interdiffused QW [16]. In contrast to other approaches, the interdiffusion technology attracts a lot of attention due to the fact that it can provide controllable/reliable tailoring the confinement profile by means of a simple post-growth treatment of the as-grown conventional triangle-shaped QW with high build-in internal electric field [17]. As far as we know there are several ways in which intermixing could be achieved, such as impurity-induced disordering [18], ion implantation intermixing [19], impurity free disordering [20] and laser annealing [21]. These features suggest that the interdiffused quantum well structure can be used to produce polarization insensitive light-emission diodes.

Despite of the unquestionable availability of the intermixing approach for improving the properties of $\text{ZnO}/\text{Zn}_{1-x}\text{Cd}_x\text{O}/\text{ZnO}$ QWs there is a lack of literature data devoted to this effect [9, 22]. Taking into account a necessity to understand the correlation between interdiffusion and QW's properties, we performed the

theoretical analysis of intermixed $\text{ZnO}/\text{Zn}_{1-x}\text{Cd}_x\text{O}/\text{ZnO}$ QWs using the approximation of a double error function potential and self-consistent 1D Schrödinger-Poisson solver. The band structure and optical properties of the interdiffused QWs with different interdiffusion lengths were compared to that of the conventional $\text{ZnO}/\text{Zn}_{1-x}\text{Cd}_x\text{O}/\text{ZnO}$ QWs.

2. Theoretical approach

The calculation of the band structure and optical properties of the $\text{ZnO}/\text{Zn}_{1-x}\text{Cd}_x\text{O}/\text{ZnO}$ QWs is based on a self-consistent 1D Schrödinger-Poisson solver using $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian for wurtzite QWs [7, 23-25]. The model takes into account the intermixing effect, strain effect, spontaneous and piezoelectric polarizations [23].

For the interdiffused $\text{ZnO}/\text{Zn}_{1-x}\text{Cd}_x\text{O}/\text{ZnO}$ QWs, the extent of the Zn-Cd interdiffusion process is generally characterized by a diffusion length L_D , defined here as $L_D = \sqrt{Dt}$ [26], where D and t are the Zn-Cd interdiffusion coefficient and annealing time, respectively. In the model presented here, we have assumed an isotropic interdiffusion of Zn and Cd with the diffusion coefficients being independent of their respective concentrations, which were also assumed to be equal and constant, i.e., we assumed Fick's second law of diffusion applies [27]. The diffused Cd composition profile, $x(z)$, across the QW structure is given by [27]

$$x(z) = \frac{1}{2}x_0 \left[\text{erf}\left(\frac{0.5L_w - z}{L_D}\right) + \text{erf}\left(\frac{0.5L_w + z}{L_D}\right) \right] \quad (1)$$

where x_0 is the as-grown Cd mole fraction in the well, L_w is the as-grown width of the QW, z is both the quantization and the growth axis (QW centered at $z=0$), and erf denotes the error function [28]. Zn-Cd interdiffusion coefficient D can be expressed for $\text{Zn}_{1-x}\text{Cd}_x\text{O}$ alloy system as follows:

$$D = D_{\text{Cd}}(1-x) + D_{\text{Zn}}x \quad (2)$$

where D_{Cd} and D_{Zn} are diffusion coefficients of cadmium and zinc, correspondingly, and they can be described by an Arrhenius equation as follows [29]:

$$D = D_0 \exp\left(-\frac{\Delta E_a}{kT}\right) \quad (3)$$

where ΔE_a is an activation energy, D_0 is a diffusivity. For the calculation the following parameters were used: $\Delta E_a=2.26$ eV and $D_0=0.6$ cm²/s (for Cd diffusion in ZnO [30]); $\Delta E_a=2.66$ eV and $D_0=1.57 \cdot 10^{-3}$ cm²/s (for Zn diffusion in ZnO [31]). The parameters involved in the calculation of optical properties for ZnO/Zn_{1-x}Cd_xO/ZnO QWs were obtained from Refs. [7] and [10].

At the final stage of our theoretical analysis we calculated the total spontaneous emission spectra of the interdiffused QWs by means of the following equation [32]:

$$r^{spon}(E = \hbar\omega) = \frac{n_e^2 \omega^2}{\pi^2 \hbar c^2} \frac{2(2g_{sp}^{TE} + g_{sp}^{TM})}{3} \quad (4)$$

where ω is the frequency, $\hbar$ is the Planck constant, c is the light speed, g_{sp}^{TE} and g_{sp}^{TM} are spontaneous emission rates for TE and TM polarization modes, which includes all types of interband optical transitions between the conduction subbands (C1, C2) and valence subbands (HH1, LH1, HH2, LH2):

$$g_{sp}^{TE, TM} = \frac{2q^2 \pi}{n_r c \varepsilon_0 m_0^2 \omega L} \sum_{\sigma=U, L} \sum_{n, m} \int \frac{k_t dk_t}{2\pi} \left| (M_{TE, TM})_{nm}^{\sigma}(k_t) \right|^2 \frac{f_n^c(k_t)(1 - f_{cm}^v(k_t))(\Gamma/\pi)}{(E_{\sigma, nm}^{cv}(k_t) - \hbar\omega)^2 + \Gamma^2} \quad (5)$$

where q is the elementary charge, k_t is in-plane wave-vector, n_r is the effective refraction index, L is the well thickness, m_0 is free-electron mass, ε_0 is the vacuum permittivity, $\hbar\Gamma$ is the half-linewidth of the Lorentzian function, (for our calculation $\Gamma=(0.1 \text{ ps})^{-1}$ was accepted), $\left| (M_{TE, TM})_{nm}^{\sigma}(k_t) \right|^2$ is the momentum matrix elements and $f_n^c(k_t)$ and $f_{cm}^v(k_t)$ are Fermi–Dirac distribution functions for the electrons in conduction band and valence band. Matrix elements $\left| (M_{TE, TM})_{nm}^{\sigma}(k_t) \right|^2$ for all types of optical transitions for TE and TM modes can be determined as follows [32]:

$$\left\{ \begin{aligned} \left| (M_{TE})_{nm}^{\sigma=U} (k_t) \right|^2 &= \frac{|\langle S|p_x|X \rangle|^2}{4} \left\{ \langle \psi_n | g_m^{(1)} \rangle^2 + \langle \psi_n | g_m^{(2)} \rangle^2 \right\} \\ \left| (M_{TE})_{nm}^{\sigma=L} (k_t) \right|^2 &= \frac{|\langle S|p_x|X \rangle|^2}{4} \left\{ \langle \psi_n | g_m^{(4)} \rangle^2 + \langle \psi_n | g_m^{(5)} \rangle^2 \right\} \\ \left| (M_{TM})_{nm}^{\sigma=U} (k_t) \right|^2 &= \frac{|\langle S|p_z|Z \rangle|^2}{4} \langle \psi_n | g_m^{(3)} \rangle^2 \\ \left| (M_{TM})_{nm}^{\sigma=L} (k_t) \right|^2 &= \frac{|\langle S|p_z|Z \rangle|^2}{4} \langle \psi_n | g_m^{(6)} \rangle^2 \end{aligned} \right. \quad (6)$$

where $\langle \psi_n | g_m^{(v)} \rangle$ is overlap integral between components of the valence-band and conduction band wave functions. The terms $|\langle S|p_x|X \rangle|$ and $\langle S|p_z|Z \rangle$ are related with Kane's parameters via following relationships [33]:

$$\left\{ \begin{aligned} |\langle S|p_x|X \rangle| &= \frac{m_0}{\hbar} P_2 \\ |\langle S|p_z|Z \rangle| &= \frac{m_0}{\hbar} P_1 \\ P_1^2 &= \frac{\hbar^2}{2m_0} \left(\frac{m_0}{m_e^z} - 1 \right) \frac{(E_g + \Delta_1 + \Delta_2)(E_g + 2\Delta_2) - 2\Delta_3^2}{E_g + 2\Delta_2} \\ P_2^2 &= \frac{\hbar^2}{2m_0} \left(\frac{m_0}{m_e^t} - 1 \right) \frac{E_g [(E_g + \Delta_1 + \Delta_2)(E_g + 2\Delta_2) - 2\Delta_3^2]}{(E_g + 2\Delta_2)(E_g + \Delta_1 + \Delta_2) - \Delta_3^2} \end{aligned} \right. \quad (7)$$

where E_g is the band gap energy of active layer, $\Delta_1 = \Delta_{cr}$ and $\Delta_2 = \Delta_3 = \Delta_{so}/3$ are splitting energies, m_e^t and m_e^z electron effective masses in the transverse and longitudinal direction.

Integrating the spontaneous emission spectra (which were defined by Eq. 4) within the whole energy range gives a possibility to obtain the values of spontaneous emission rate per unit volume [32]:

$$R_{SP} = \int_0^\infty r^{spon}(\hbar\omega) d(\hbar\omega) \quad (8)$$

3. Results and discussion

In general, the quantum well intermixing is based on a fact that a QW is an intrinsically metastable system due to the large concentration gradient of atomic

species across the interface between the QW layer and the barrier. At high temperatures, diffusion of atom species from one lattice to another makes the QW smeared with adjacent barrier materials (Fig. 1). This intermixing process can be significantly enhanced by the presence of impurities or point defects, in the vicinity of the interfaces of the QW. This mechanism promotes the smoothing the initially abrupt cadmium content profile to approximately a double error function as illustrated in Fig. 2. As interdiffusion proceeds, the Cd content in active layer decreases and the QW depth/width is changed (Figure 2). Immediately after, the change in compositional profile will lead to modification of band structure and optical properties of QWs. Using six-by-six **k.p** approach and finite difference scheme we computed the subband bound states in QW in order to understand the role of interdiffusion.

Fig.1. Schematic view of crystal structure of the barrier and well layers for (a) conventional and (b) interdiffused ZnO/Zn_{1-x}Cd_xO/ZnO QWs.

Fig. 2. Cadmium profile for interdiffused ZnO/Zn_{1-x}Cd_xO/ZnO QWs at the different values of the interdiffusion length L_d (the case of $L_d=0$ corresponds to the conventional QWs with abrupt interface).

Fig. 3 Band structure of the conventional (a) and interdiffused (b) 7nm-ZnO/2nm-Zn_{1-x}Cd_xO/7 nm-ZnO QWs

Figure 3 shows calculated valence band structures for the valence subbands for a conventional and interdiffused 2 nm-Zn_{0.85}Cd_{0.15}O quantum wells. In the case of conventional QW the self-consistent energy profile of quantum well is triangle-shaped. The huge build-in electric field exists will push the valence subbands upwards, as shown on Figure 3a. It was revealed that the dispersion of HH1 subband was quite parabolic in shape. It was also found that there is a strong coupling of all heavy-hole (HH1, HH2) and light-hole (LH1, LH2) valence

subbands at region k_t smaller than 0.3 nm^{-1} and coupling of HH2 and LH1 within wavenumber range between $0.6\text{-}0.8 \text{ nm}^{-1}$. It is interesting to note that increase in diffusion length significantly modifies the valence band structure of $\text{ZnO}/\text{Zn}_{1-x}\text{Cd}_x\text{O}/\text{ZnO}$ quantum well. First of all, the interdiffusion leads to change of the confinement profile from triangle-shaped to parabolic-like. In this regard, all valence subbands shift downwards and energetic distance between these subbands is reduced (Fig. 3b). Nevertheless, the HH1 subband remains parabolic shape. Another observable feature is the strong mixing between the HH2 and LH2 valence subbands at region k_t greater than 1 nm^{-1} .

Based on the analysis of Bloch wave functions for carriers and overlap integrals, the matrix elements for main interband transitions for TE- and TM-polarization modes were calculated. It is appropriate to note that in the case of TM-polarization, electric field vector is directed along the c axis ($E \parallel c$), whereas in the case of TE-polarization electric field vector is perpendicular to the c axis ($E \perp c$). In many cases TM-polarization mode is undesirable because the radiation output (with such polarization) extracted from the wide-gap semiconductors in the direction perpendicular to the (0001) plane is virtually impossible. Fig. 4 shows the typical dependence of the matrix elements on the wave vector for conventional and interdiffused QWs. The optical matrix elements of interdiffused QW structures [see Fig 4c and 4d] are shown to be larger than that of a conventional QW structure [see Fig4a and 4b]. This is caused by the reduction of the internal electric field due interdiffusion process. The main contribution to the spontaneous emission rate was from the C2-HH1 and C2-LH1 transition matrix elements. We also observed that the transition matrix element at the zone center between the conduction band and heavy-hole subband is similar to that between the conduction band and light-hole subband. It is interesting to note that the transition matrix elements for TE mode are much greater than in the case of TM-polarization for both QW structures. It can be explained by the fact that TM matrix elements are mainly determined by the g_3 and g_6 components of the carrier wavefunction, having low amplitude for heavy hole and light hole subbands.

Fig. 4 Transition matrix elements for the conventional (a, b) and interdiffused (c, d) 2 nm- $Zn_{0.85}Cd_{0.15}O$ quantum wells

Notably, the knowledge of the matrix elements for all types of direct interband transitions gives a possibility to calculate spontaneous emission spectra with consideration of TE and TM polarization modes. The spontaneous emission spectra for both conventional and interdiffused 2 nm- $Zn_{0.85}Cd_{0.15}O$ quantum wells were calculated from low sheet-carrier density $1 \cdot 10^{12} cm^{-2}$ up to high low sheet-carrier density $9 \cdot 10^{12} cm^{-2}$ (in steps of $1 \cdot 10^{12} cm^{-2}$). The resulting spectra are demonstrated in Fig. 5. As a consequence of the reduced QCSE, the emission intensity increases and shifts to blue with increasing the diffusion length. The reduction in the QCSE in combination with the changes in the matrix elements of interband transitions improves the gain when going from the $L_d = 0$ to $L_d = 0.37$ nm. It is due to the fact that the electron confinement in the parabolic-shaped well is larger than that for the conventional QW structure. Figure 5 also shows the emission spectra as function of

the sheet carrier density for both QW structures. We can see that the emission spectra increase with increasing carrier density because of the screening effect of the internal field.

Fig. 5 Calculated spontaneous emission spectra of conventional and interdiffused 7 nm-ZnO/2 nm-Zn_{0.85}Cd_{0.15}O/7 nm-ZnO QWs at different values of the sheet-carrier density.

In order to understand the behavior of the spontaneous emission rate governed by interdiffusion process, we calculated the enhancement ratio of the conventional and interdiffused QW at the different values of the diffusion lengths. As interdiffusion proceeds, this ratio rises initially to a maximum and then decrease monotonically as QW intermixing becomes stronger [See Fig. 6]. It should be mentioned that the R_{SPint}/R_{SPconv} ratio attains its maximum at approximately 4 Å. According to equation $L_D = \sqrt{Dt}$ such a value of optimized diffusion length can be reached after rapid thermal annealing ($\tau=60$ s) of QW structure at the annealing temperature 700 K. The non-trivial behavior of the enhancement ratio can be explained in terms of an existence of two different regimes of the interdiffusion process. At the initial stage of intermixing (the case of the low values of the diffusion length $L_d < 4$ Å), only the cadmium and zinc species locating near to the boundary between well and barrier layers can diffuse through heterointerface, thereby changing the

confinement profile. While the atoms that are locating far from the heterointerface don't have sufficient speed to reach the interface. In fact, such a small atomic interdiffusion leads to not only the forming of parabolic-shaped profile, but also to decrease of the quantum well thickness in the limited region of the quantum well structure (closed to heterointerface). In other words, confinement profile remains unchanged in the center of ZnCdO quantum well and the large part of ZnO barriers. Therefore, the value of the well depth is virtually unaltered at the low diffusion lengths. Under such conditions, the overlap integral between electron and hole wave functions is enhanced (due to lower well width and improved confinement profile), whereas the optical transition energy C1-HH1 is blue-shifted. In fact, this phenomenon compensates the negative role of the quantum confined Stark effect. That's why the intensity of the spontaneous emission spectrum of the interdiffused QWs with short diffusion length is larger than that of the conventional QW, as was demonstrated on Fig 5. Further thermal-induced increase in diffusion length helps both Zn atoms to reach the center of QW and the Cd ones to move deeply into barriers. It gives rise to decrease in the well depth (which is defined by the modified value of the band-gap offset) and gradual increase in the well width. This causes the red-shift in the transition energy and decrease in the wavefunction overlapping. Thus, the large values of diffusion length are associated with the lower enhancement ratios, as shown in Fig. 6. Furthermore, according to the cutoff rule (that defines an upper limit for the quantum well thickness, above which the quantum confinement effect is considered to be inessential [34]), the complete QW intermixing leads to reduction of the two-dimensional nature of the confined states and their transformation into three dimensional (3D) unbound bulk-like continuum states. Obviously that such vanishing of the quantum well confinement will play a negative role in forming high-efficiency radiative recombination channels in ZnO-based QW structures.

Fig. 6 Dependence of the ratio of spontaneous emission rate of the interdiffused to similar one for conventional 7 nm-ZnO/2 nm-Zn_{0.85}Cd_{0.15}O/7 nm-ZnO QWs QW on the diffusion length

Conclusions

We have presented a comparative study of the band structure, matrix elements and spontaneous emission spectra of the conventional and interdiffused ZnO/ZnCdO/ZnO QWs. Our calculation is based on the valence band Hamiltonian derived using the k.p method and solving the self-consistent Schrödinger-Poisson equations. We showed that an increase in the diffusion length enhances the TE and TM matrix elements for main optical transitions. We also demonstrated that intermixed QW offers pronounced higher spontaneous emission rate reaching the maximum at the diffusion length of 4 Å due to the improved quantum confinement characteristics. The obtained results suggest that the time- and temperature-regulated Zn-Cd interdiffusion can be cost-effective tool for selective enhancement of the visible light emission from ZnO-based QWs.

Acknowledgments

This publication is part of Dr. I. Shtepliuk's research work at Linköping University, thanks to a Swedish Institute scholarship.

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