Photoluminescence surface mapping as a probe for interface disorder in quantum well structures

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Abstract. We show here that a simple photoluminescence surface mapping across the sample indicates whether the nano-quantum well layer is interface disordered or alloy-disordered. Two sets of CdSe-ZnSe nano-quantum well structures, one grown by metalorganic vapour phase epitaxy and atomic layer epitaxy are analysed both structurally and optically. The former structures are found to be predominantly interface disordered whereas the latter samples are found to be alloy-disordered. Photoluminiscence mapping across the sample surface reveals that there is a strong correlation between the spatial variations in the exciton band peak energy position and the linewidth (~ 60 – 100 meV and 40 – 70 meV, respectively) in the well-width disordered systems compared to the alloy-disordered or quantum dot systems. A contour plot of the photoluminiscence shows for the rough structures orderly patterned contour lines for both the peak energy and linewidth. On the contrary, contour plots for the quantum dot samples show uncorrelated maps with the linewidths scattered in relation to those of the peak energy. We show that even without structural characterisation, one can tell the quality of the sample under study (whether rough or quantum dot) by carrying out a surface mapping of the emission and studying the contour maps and scatter plots of peak energies and their corresponding linewidths. These results can assist in shedding more light on similarly lattice-mismatched material combinations in the III-Vs and III-Nitrides nanostructures. This knowledge is crucial for understanding not only of what is behind the optical properties, but also of the growth dynamics of these systems and can be utilised in calibrations of the growth process to optimise sample quality.

1. Introduction

Problems of structural disorder associated with growth in ultra-thin structures include interface roughness, interfacial alloy formation, and well width fluctuations. Nanostructure heterointerfaces between quantum wells and barriers are quite rough within certain lateral regions that change in size at small terraces or islands with heights generally being in monolayers so that excitonic photoluminescence (PL) linewidths are closely related to the heavy-hole excitons bound in the potential well/barrier caused by the fluctuations formed by submonolayer deposition [1-13].

Here, we draw attention to photon emission characteristics of CdSe/ZnSe ultra-thin nanostructures surveyed on single quantum well structures using surface mapping measurements of the PL spectra. While a great deal has already been learned about the luminescence from these II-VI heterostructures, insufficient attention has been given of the correlation between the luminescence and the structural details of the CdSe insertion layer responsible for the optical effects reported. This knowledge is

crucial for understanding not only of what is behind the optical properties, but also of the growth dynamics of this lattice-mismatched system. This presentation tries to demonstrate that before any further investigation, surface mapping of the sample is important in determining the overall behaviour of the sample in terms of uniformity and nano-quantum well width fluctuations. In some areas where the structural variations are large over a sample, PL mapping can yield information which would otherwise require several samples to be grown.

2. Samples

The structures studied in this work have been grown by two methods: atomic layer epitaxy (ALE) for samples KC1 and KC2 and metal-organic vapour phase epitaxy (MOVPE) for samples KC7 and KC9. All QWs in this case are ZnSe/CdSe/ZnSe systems. All the samples are single QWs (SQWs) consisting of the GaAs substrate, followed by a ZnSe buffer layer, the CdSe QW, and finally a ZnSe capping layer. Details of the sample structures are given in Table 1 below.

Table 1: Summary of the sample structure

Sample Name	ZnSe buffer thickness (nm)	CdSe QW thickness (ML*)	ZnSe cladding thickness (nm)
KC1	150	0.5	50
KC2	150	1.0	50
KC3	150	1.8	50
KC4	150	1.0	50

*ML = monolayer

3. PL measurements and surface mapping

The emission was excited using a 357 nm argon-ion continuous-wave (cw) laser. The laser beam first passed through a mechanical chopper, which modulates the beam at 330Hz beam then through a 395 nm interference filter to remove the laser plasma lines, after which it was focused with a converging lens onto the sample held at a fixed temperature in a continuous flow cryostat. The sample was placed at an angle such that the direct reflection of the laser is not collected by the collimating lens between the sample and the spectrometer. The luminescence from the sample was then focused with the help of two lenses onto the slits of a grating spectrometer, of which the slitwidth could be varied (giving an appropriate spectral resolution) depending on the intensity of the luminescence from the sample in the wavelength range of interest. Generally the slit width was between 200 – 500 μm giving resolutions of ~1 meV. The output from the spectrometer was detected using a photomultiplier tube which was linked via an amplifier to the photon counter also fed with the 330 Hz reference signal. The data was then collected by a computer which, in addition to data acquisition, also regulated the temperature controller and drove the spectrometer motor.

The sample was divided up into a grid of equal small point areas. Then the PL spectra of each of the point areas across the entire surface were measured. From these spectra, the peak photon energy and full-wave half-maximum (FWHM) of the exciton band for each point was extracted. An analysis of the results yielded a relation between the peak energy and FWHM. Also 2-D contour maps of these parameters across the *xy*-plane of the surface were plotted. PL peak intensity mapping was not considered because of fluctuations in the intensity of the measuring incident laser beam.

4. PL Results

Figure 1 shows the PL spectra of all the samples studied in this work. All spectra are obtained at ~ 10K and the emissions are quite bright compared with standard ZnSe. Figure 1A shows spectra for KC7 and KC9 samples grown by MOVPE and Figure 1B shows that for ALE-grown KC1 and KC2. The exciton bands of KC7 and KC9 with nominal well-widths 1.8 and 1.0 ML sit on peak energies ~ 2.60 and 2.75 eV, respectively, with linewidths of ~ 150 and 60 meV, respectively. The emission bands in the ALE-grown single QWs KC1 and KC2 with nominal well-widths of 1.0 ML and ½ ML, sit on ~ 2.72 and 2.76 eV, respectively. Unlike the MOVPE-grown samples, these have much narrower PL lines of linewidths ~ 6 and 10 meV, respectively. A comparison of all the samples reveals that in the ALE systems, the PL lines feature relatively weaker exponential tails on the low energy side extending between ~ 20-50 meV, compared to the much stronger tails in the MOVPE samples.

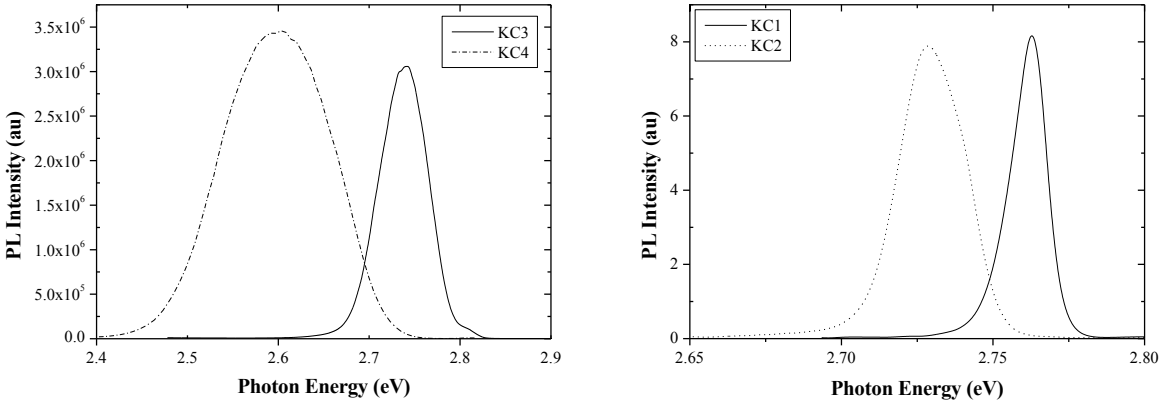

Figure 1. PL spectra of the studied samples taken at temperature of 12 K.

5. PL surface maps

In this part, spatial variations in the exciton band PL spectra were measured by surface mapping of the emission by scanning the sample laterally in the plane of the QW, i.e., across the sample area. The luminescence intensity was then mapped as a function of the emission peak energy and emission linewidth and the spatial coordinates *x* and *y*, resulting in contour plots shown in Figure 2. Even at the scale of our plots, it can be seen that there is some good correlation in the contour patterns of the peak energy and linewidth in the MOVPE structures with large variations in these values across the sample compared to the ALE structures with little variatians. In the former samples, the variations in peak energy and linewidth across the sample area depict well matched patterns showing a strong spatial correlation between the variations in the peak shift and linewidth. These large variations of the order of magnitude ~50 meV and ~30 meV for peak energy and linewidth, respectively, are associated with structures that predominantly exhibit large well width fluctuations across the plane of the well region, in good agreement with the structural results previously reported [14]. On the other hand the latter structures exhibit peak energy and linewidth variations across the sample of the order of ~1.5 meV, respectively, a far smaller change compared to that in the former. Emission in the ALE samples is expected predominantly from self-assembled quantum dots (QDs). The rather narrow Pl lines and very weak fluctuations in the peak energy and linewidth with position suggests two possibilities: (i) either there are too few of these QDs for their size-effects to be noticed or/and (ii) the dots possess size homogeinity. From structural results, we suggest that an interplay between low dot densities and less spread in size of the QD recombination centres influence the quality of the luminescence.

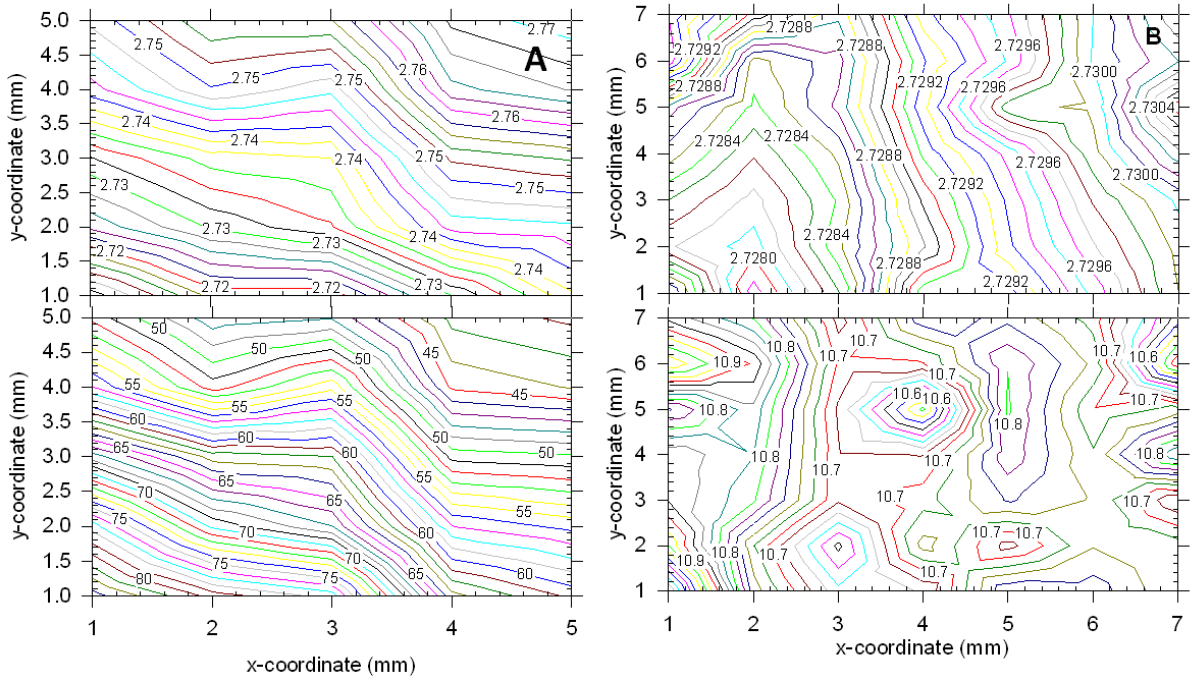

Figure 2. Contour maps showing the variation of the exciton peak energy and linewidth across the (A) MOVPE and (B) ALE grown samples.

Figure 3 gives a plot of linewidth versus peak energy. This shows more vividly the difference in quality between the MOVPE and ALE grown structures with the former systems possessing large PL linewidths than the latter. The strong correlation between the well thickness and linewidth in the former case shows a general decrease in linewidth with increasing exciton band peak energy in these systems. This means that regions with smaller well-widths have narrower PL bands, and this pattern is generally reflected in the overall behaviour of both the MOVPE and ALE systems. This is contrary to the III-V and other II-VI, systems reported in which the opposite is observed to be the case [15-20]. Furthermore, within the range of well-width sizes studied here, ½ - 3.5 ML, there appears to be a linear relationship between the well-width size and its exciton band linewidth. From the mapping, we reveal a well-defined linear variation of well-width and PL linewidth across the samples, with linear fits, $\Gamma(E) \sim \beta_0 + \beta_1 E$ (where the β's coefficients or empirical parameters are related to the sample quality) to the data points yielding individual slope values (giving a measure of the roughness) of -0.64, and –0.73, respectively. As already observed earlier, the data points for the more homogeneous ALE series are clouded over the same point, emphasising the uniformity of the active layer. For clarity, we apply the curve fits shown. A linear relationship would entail the line width continuously increases with peak energy. On the other hand, a more detailed analysis reveals, $\Gamma(E) \sim \beta_0 + \beta_1 E - \beta_2 E^2$, i.e. a quadratic dependence offers a more befitting model considering that for bulk materials, the PL linewidth is generally less than for QW structures. If we were to extrapolate the $\Gamma(E)$ curve down towards the band-edge energy 1.84 eV for bulk strained CdSe, an ideal peak energy dependence of linewidth should approach bulk values so that a quadratic function, with accurately defined β–coefficients, provides a more sensible approximation to the overall behaviour. A precise account of the β-coefficients would require a more detailed and rigorous study which is not within the scope of this work. Nonetheless, a closer analysis of this quadratic form leads us to conclude the parameters are strong functions of the growth procedure and depend a great deal on the interface and alloy disorder in the systems.

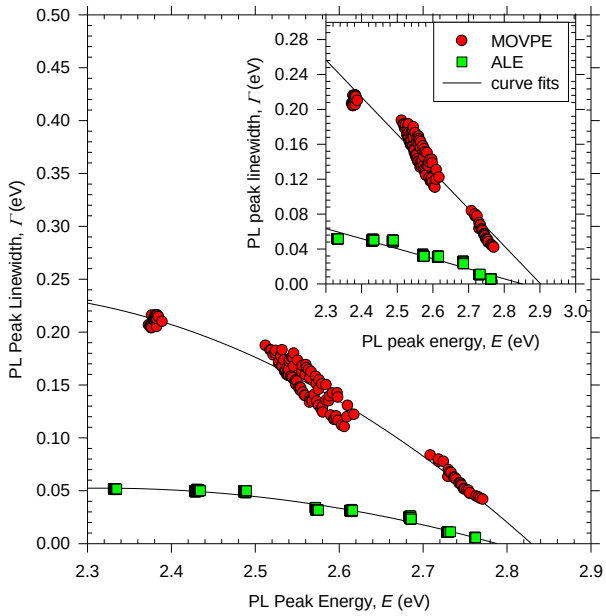

Figure 3. Overall plot of all the mapping data points comparing the MOVPE and ALE grown systems. The insert shows the linear fits to the points.

6. Conclusion

The luminescence of the QWs, the alloy-disordered and rough structures, has also revealed some new information that further distinguishes these two categories. Our PL characterisation shows that knowledge about whether the structures are alloy-disordered or rough can be derived by a simple PL surface mapping across the sample area. In rough structures, there is strong correlation between variations in peak energy position and the linewidth of the main exciton band. There is a generally a linear decrease in linewidth with increasing peak energy across the sample, displaying the typical behaviour of the CdSe-ZnSe QW systems, that thinner well widths give narrower PL lines than thicker ones. On the other hand, in alloy-disordered structures, there is virtually no correlation between the peak energy and linewidth. The variations in peak energy and linewidth across the sample area is weak and very random. In fact this is because they do not show any significant spatial fluctuations either in peak energies or linewidths across the sample, at least relative to the rough structures. A contour plot of the luminescence will show, for the rough structures, orderly patterned contour lines for both peak energy and linewidth, with their overall contour maps well correlated. On the contrary, contour plots for the QD samples show uncorrelated maps, with the linewidth contour lines rather very randomly distributed in relation to those of the peak energy. So for the first time we have shown that, even without structural characterisation, one can tell the quality of the sample (whether alloy-disordered or rough) by carrying out a prior surface mapping of the emission and producing a contour map of peak energies and linewidths. Surface mapping has also revealed variations in peak energy across the rough samples do not only correspond to monolayer fluctuations in well-width, but fluctuations in steps of submonolayers are very common. This explains why thinner QWs give narrower lines compared to thicker one in these systems. Further, this mapping analysis has shown alloy-disordered structures give good quality structures in terms of optical properties in comparison to the rough ones. We see more intense and less broadened exciton emission bands in the former relative to the latter.

Clearly the PL peak energy is a strong function of the growth sequence so that it is still impractical to accurately tie calculated and measured QW exciton-band peak energies. In other words, a ‘universal’ PL peak energy for QW insertions that can be precisely tied with a particular well-width

does not exist for these II-VI structures, unlike the well established III-V systems where it may be possible because of the high quality of the produced structures.

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