Photoluminescence and X-ray studies of thin layers down to single quantum wells

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ABSTRACT

We have demonstrated the capability and limitation of nondestructive photoluminescence and X-ray diffraction tecniques in the characterization of GaAs, AlGaAs, InGaAs matched epitaxial layers grown by molecular beam epitaxy as well as quantum wells grown by metalorganic chemical vapour deposition. The application of the X-ray diffraction and the photoluminescence methods to the same objects made it possible to control Al content in the $Al_xGa_{1-x}As$ layers in the range of x ($0 \le x \le 1$) and to solve some technological questions connected with layers lateral homogeneity.

Keywords: photoluminescence, X-ray diffraction, epilayer, AlGaAs/GaAs quantum well.

1. INTRODUCTION

Development of epitaxial technologies such as molecular beam epitaxy (MBE) or an alternative for this technique metalorganic vapour phase epitaxy (MOVPE) have recently been providing the possibility of producing thin layers with abrupt transition at the interfaces, and precisely controlled composition and doping level. GaAs, InGaAs, AlGaAs epilayers and AlGaAs/GaAs multilayer heterostructures (HS) have recently become a subject of study. Nondestructive characterization methods of heterojunction abruptness and its quality are essential. One of such methods is the X-ray diffraction technique. Photoluminescence is a powerful nondestructive technique for characterization of semiconductor materials and quantum wells (QW) too.

In this paper we present some of our recent experiments to demonstrate the complex application of low-temperature photoluminescence method (PL) and X-ray technique for determination of composition, structural and electronic properties of heteroepitaxial systems with ultra-thin layers down to single quantum wells.

2. EXPERIMENTAL METHODS

We have investigated lattice-matched HS AlGaAs/GaAs ($0 \le x \le 0.8$) and InGaAs/GaAs ($0 \le x \le 0.01$). Such structures have been grown by MBE technique[1]. Single and multiple quantum wells with well width ranging from 2 to 10 nm were grown by MOVPE technique. Quantum well samples were grown on semi-insulating, (SI), undoped (100) GaAs substrates. The typical structure included 1 μm GaAs buffer layer, and 0.5 μm $Al_{0.6}Ga_{0.4}As$, upon which were the QW's and barriers (inset to Fig.3).

The 514.5 and 488.0 nm lines of Ar-ion laser (LG-503) were used as the excitation source and the incident laser power density was between 5 and 50 W/cm^2. The 632.8 nm line of He-Ne laser with incident laser power density < 10 W/cm^2 was used too. Our detection system was based on the spectral comlex KSVU-23 with grating spectrometer MDR-23 controlled by computer. Luminescence was detected by a photomultiplier in the current-flow regime. Photoluminescence measurements were conducted at 300, 77 and 4.2 K. The structural quality of the epitaxial films was monitored by X-ray reflection topography (CuKα radiation in the assymmetric configuration, 531 reflection) and by X-ray diffractometry using a two-crystal spectrometer and CuKα radiation in the (n,-m) geometry. These two methods have allowed to determine not only the film quality (thickness, lattice parameter, microbending, etc.) but also the film composition.

3. EXPERIMENTAL RESULTS

It is known that the composition of ultra-thin layers can be determined with the help of destructive methods such as Auger spectroscopy, mass spectrometry, neutron activative analysis, etc. We have measured composition and it's lateral homogeneity in thin ($d \leq 1$ μm) matched epilayers InGaAs/GaAs ($0 \leq x \leq 0.01$) and AlGaAs/GaAs ($0 < x < 1$) by the combination of nondestructive methods: X-ray diffraction and PL techniques in the following way.

The same experimental procedure was used at 4.2, 77 or 300 K to measure the AlGaAs or InGaAs epilayers energy gap from the known position of PL peak ($h\nu_{max}$). The PL peak is not precisely related with the energy gap[2] ($E_g = h\nu_{max} + \Delta$). We supposed that it is to be the linear low for the energy gap (E_g variation versus Al or In concentration $\Delta E_g = E_g(x) - E_g(0) = K(x)$). The x value was deduced from direct methods (electron probe determination, mass spectrometry, etc.) for the layers with the same film thicknesses and on the same area of samples. This x value was used further for K determination. Unfortunately in this approach we didn't take into account the internal strain and it's relaxation by misfit dislocations[3]. Simultaneously we determined x from X-ray diffraction measurements of lattice parameter a. Then we have used X-ray and PL data to determine the average value of K. We demonstrated our approach for optical and X-ray composition determination for samples which are listed in Table 1. The lattice mismach f between a film with the bulk lattice constant a_1 and a substrate a_2 is defined as $f=\Delta a/a_2=(a_1-a_2)/a_2$.

Table I. Initial parameters of MBE epitaxial $In_xGa_{1-x}As$ films.

No.	Thickness, μm	p,cm^{-3}	$\Delta a/a_2$	Indium content x,mol.%
1388	1.4	8×10^{16}	9.9×10^{-4}	0.2
1389	1.1	8×10^{16}	1.27×10^{-3}	0.4
1390	1.45	$6\text{-}8\times 10^{16}$	1.48×10^{-3}	0.42
1391	1.3	$5\text{-}8\times 10^{16}$	1.85×10^{-3}	0.9

Fig.1 shows the X-ray diffraction diagrams (the rocking curves) given by the samples in the neighbourhood of the 004 peak (a) and typical PL spectrum (b) for the sample No.1389 from Table I. The film composition x was directly derived from the angular separation of the 004 peak given by the GaAs substrate and $In_xGa_{1-x}As$ layer in a double crystal X-ray diffraction experiment. The angular separation has been experimentally shown by using X-ray diffraction, thickness measurements by interferometry on MII-4 or profilometry on M252(A1) (the error in the thickness measurements ~1 μm is less than 0.006 μm) and electron probe microanalysis EPM (detection sensitivity about 100 ppm for all elements down to Be with Z=4) to be linearly related to x by

$\Delta\Theta=(8950\pm1)x$ for InGaAs epilayers and $\Delta\Theta=(180\pm1)x$ for AlGaAs epilayers. (1)

The value of x, derived from Fig.1,a, is 0.04 i.e. $In_{0.04}Ga_{0.96}As/GaAs$. The dimensions of the beam impact were about 0.1×0.5 mm^2 and the x value could be mapped and measured at the same point of the sample surface, which was examined in PL experiment. The typical PL spectrum (Fig.1,b) reveals two main peaks which correspond to band-to-band and band-to-acceptor transitions. The PL experimental conditions have been described earlier[1]. The In concentration could be measured by detecting the bandgap shift in PL spectra. When the In concentration (i.e. atomic fraction x) increases the band gap energy (i.e. E_g) decreases. The energy shift ΔE_g in the band-to-band or band-to-

acceptor lines has been measured at 77 K using PL spectra. We determined the reduction in bandgap from that GaAs epilayers. We found that ΔE_g [eV]=E_g (GaAs)-E_g (InGaAs)=K_2 x, where ΔE_g is the bandgap shift in meV and x is the molar fraction of InAs. The In content of samples was determined by electron probe analysis too. And then these samples were used to establish the PL calibration relating the bandgap at 77 K to In content. Our study was specially aimed on establishing the accurate calibration at small In fraction (x<0.01). The value of K_2 has been corrected with the X-ray measurements data and, in our case, K_2 =1.5. The In concentration listed in Table I was $0.002 \leq x \leq 0.09$.

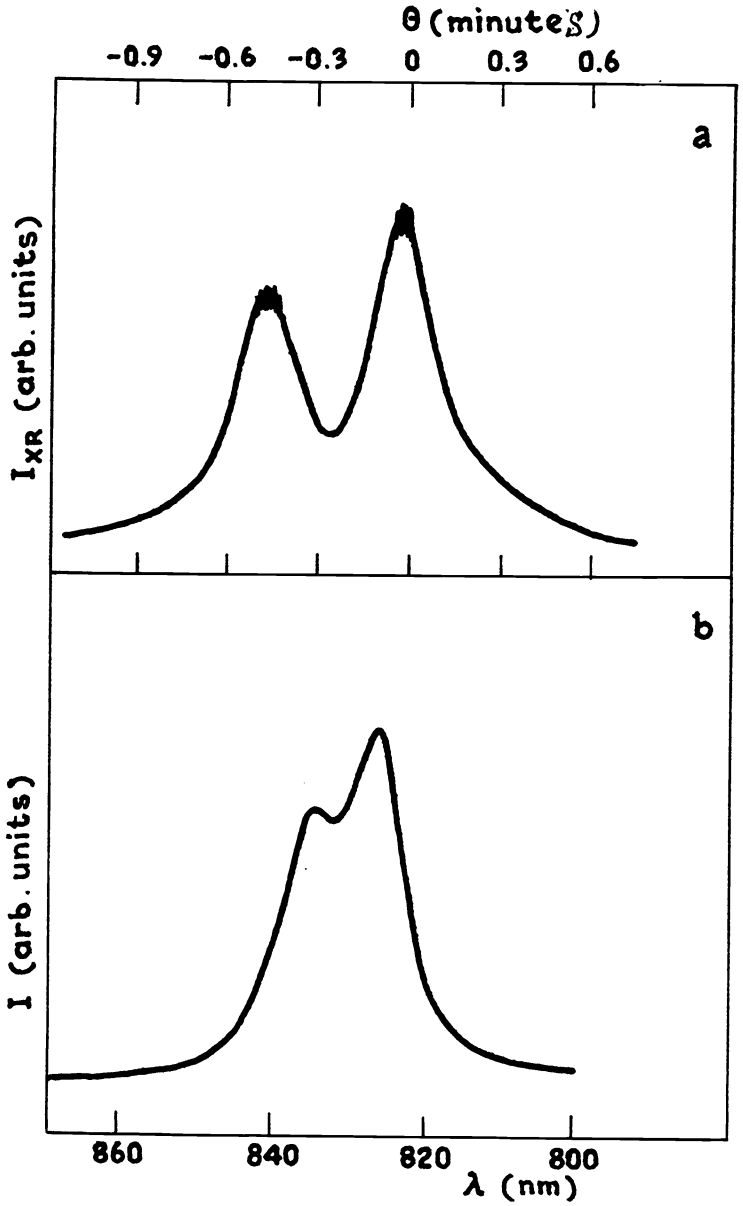

Fig.1. Double-crystal X-ray diffraction pattern in the neigbourhood of the 004 peak (a) and photoluminescence spectra for InGaAs epilayers on GaAs at 4.2 K. (b).

In addition the lateral local fluctuations of x at micro-scale were measured using the PL mapping technique including PL peak derivation[4] with following method:

(i) PL spectrum was recorded with large laser spot (micro-scale) (Fig.2,a).

(ii) The derivative of the PL spectrum was calculated after digital filtering (Fig.2,b).

(iii) Then we were able to record PL intensity profiles at wavelengths p^+ (positive side of derivative) and p^- with high spectral and spatial resolution. Furthermore, a correlation was observed between local PL efficiency of In concentration and point defects - dislocations revealed by photoetching and X-ray topography.

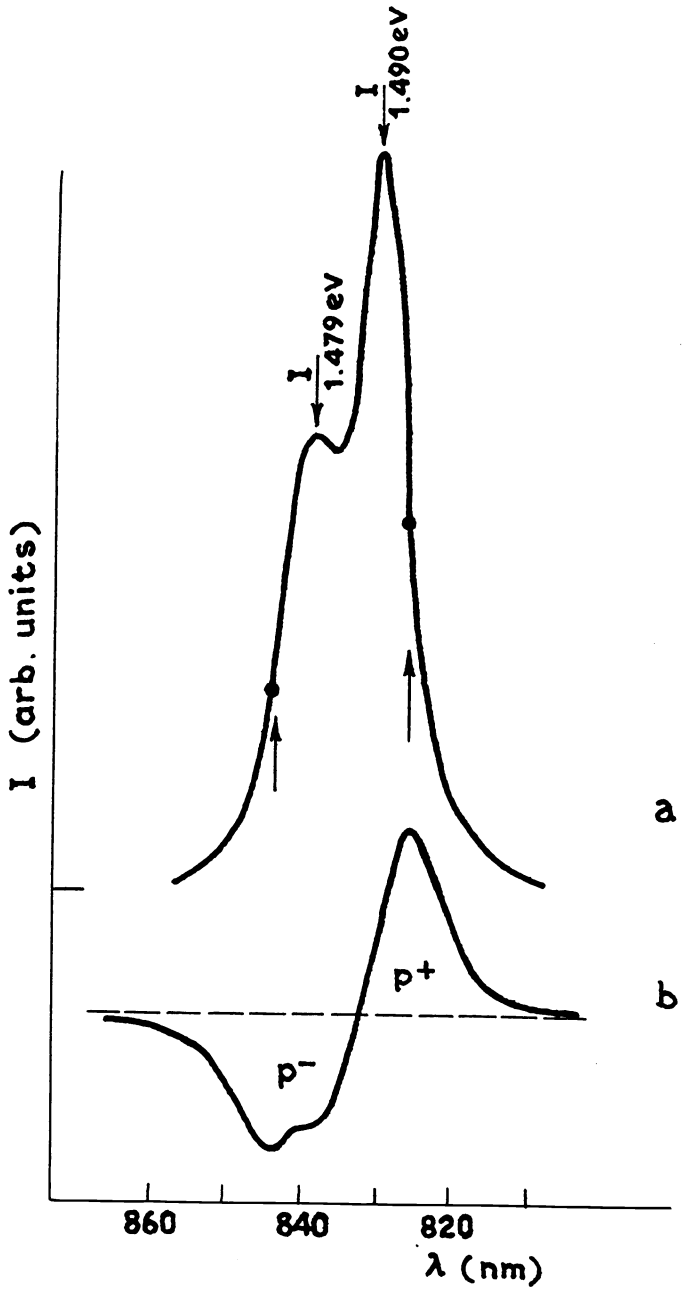

Fig.2. PL spectrum of InGaAs at T=77 K excited at λ=488 nm (a) and it's derivative (b).

4. PHOTOLUMINESCENCE CHARACTERISTICS OF AlGaAs-GaAs SINGLE QUANTUM WELLS

Photoluminescence (PL) is the most popular analitical method for studying the undoped high-quality AlGaAs quantum wells because it is nondestructive. PL provides a quick and relatively simple method of studying the excitonic transitions between two-dimensional electrons and holes.

Fig. 3 shows PL spectra obtained from MOVPE grown structure with three GaAs quantum wells (QW) measured at 4.2, 77 and room temperatures. Quantum wells consist of GaAs layers, 2-10 nm thick, separated by the barriers 100-nm thick $Al_{0.6}Ga_{0.4}As$ layers as shown in the inset to Fig 3. The QWs were grown in the order of thickness decrease from substrate towards the surface for the reason that luminescence from wider wells (lower energy) could not be absorbed by thinner wells above. Signal of the GaAs near-band-edge transition is ~30% of that

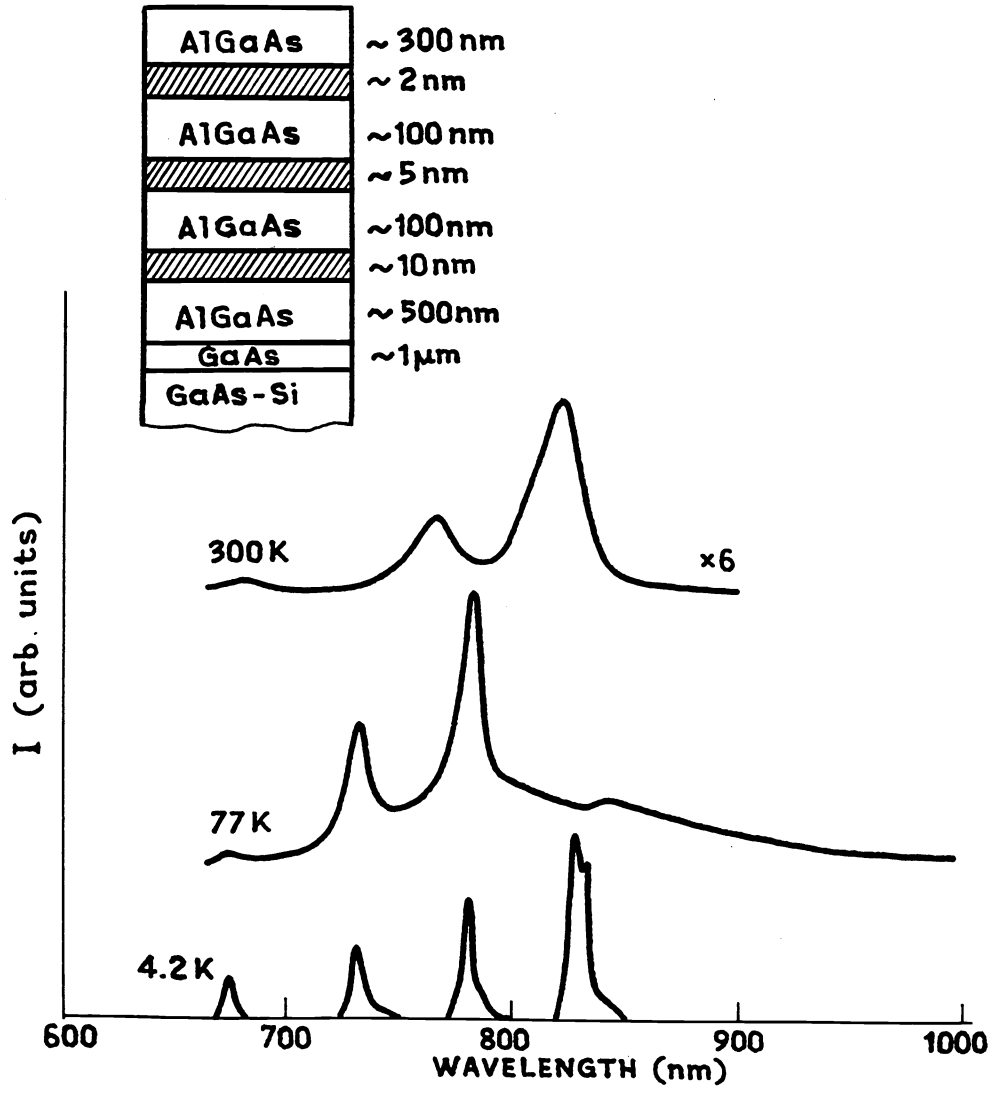

Fig.3 Photoluminescence spectra for GaAs/$Al_{0.6}Ga_{0.4}As$ quantum wells with different well widths L_z=2.5, 4.2, 8.0 nm measured at 4.2, 77 and 300 K. The sample was excited by the He-Ne laser (632.8 nm, incident power density=5 W/cm^2).Basic structure used in this study is shown in the inset.

from wells because GaAs buffer was the deepest lying epilayer. QW photoluminescence signal decreased with well size decreasing, so the well become less efficient at collecting carriers from the barriers. $Al_{0.6}Ga_{0.4}As$ signal with an indirect bandgap was not observed. The main peak of each spectrum is due to an n=1 electron-to-heavy-hole transition as shown in the inset to Fig.4. The well widths L_z were calculated from measured peak energies which corresponds to each well widths. This calculation was necessary in our study under the certain set of x (from x=0 to x=0.6) and temperatures (4.2, 77, 300 K) of measurements. Fig.4 shows the values of the well widths of the quantum wells obtained from the equation for perfect rectangular quantum wells[5]:

$$Tan^2\sqrt{\frac{m_w E L_z^2}{2h^2}} - \frac{m_b(V-E)}{m_w E} = 0, \qquad (2)$$

where V =barrier height; L_Z =well width; m_b (m_w)=barrier (well) mass of the particle; E = eigenvalue in the 1D finite square potential well.

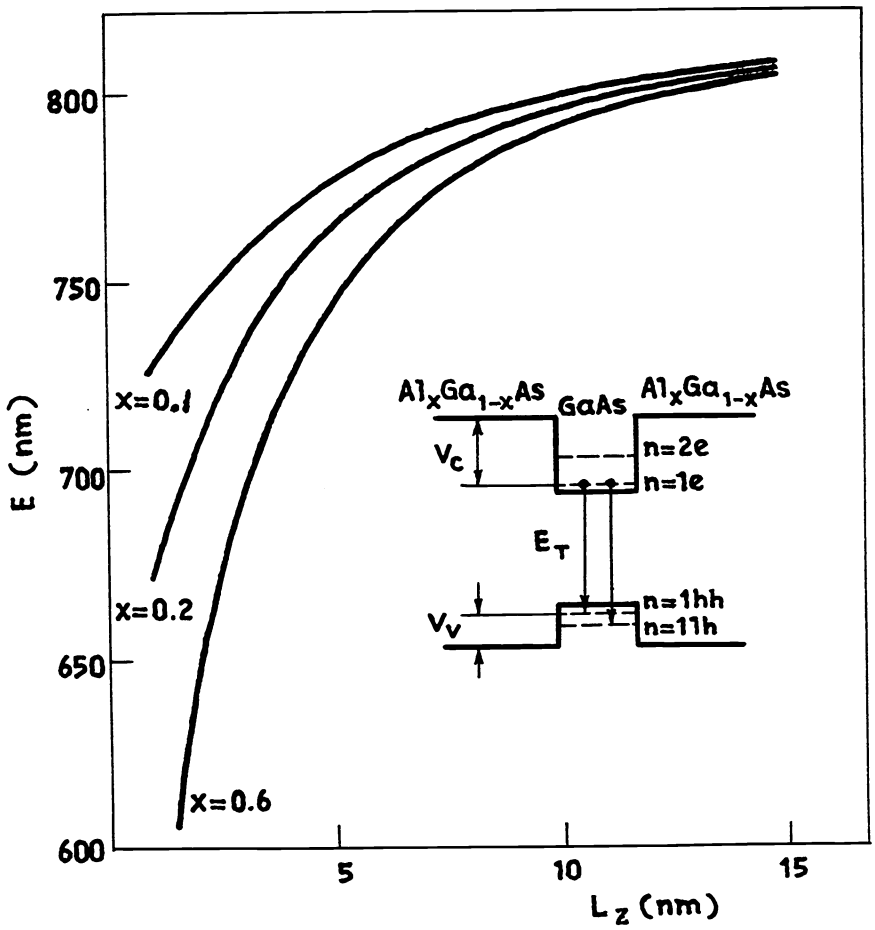

Fig.4 Calculated relationship between emission wave length λ and GaAs well width L_z correspondingly to the n=1 electron to heavy hole transition for a rectangular quantum well with $Al_xGa_{1-x}As$ barriers as shown in the inset. The sample temperature was 4.2 K.

The band parameters used in the calculation are listed in the Table II and Table III. The energy dependent effective mass for an electron was taken into account in the calculation.

Table II. Various band parameters pertaining to the solution of equation (2) of this work.

$V_c = 0.85(E_g(Al_xGa_{1-x}As) - E_g(GaAs))$ $\quad V_v = 0.15(E_g(Al_xGa_{1-x}As) - E_g(GaAs))$
$E_g = \begin{cases} 1.5190 + 1.247x & x \le 0.45 \\ 1.5190 + 1.247x + 1.147(x - 0.45) & x > 0.45 \end{cases}$
$E_g(T) = 1.519 - 5.405 \times 10^{-4} T^2/(204+T)$

Table III. The effective masses m of carriers in the conduction and heavy-hole bands used in this work.

	GaAs	$Al_xGa_{1-x}As$
m^*_{hh}	$0.48m_0$	$(0.48+0.31x)m_0$
m^*_e	$(0.0665+0.0436E+0.236E^2 - 0.147E^3)m_0$	$(0.0665+0.083x)m_0$
E in eV $\quad m_0 = 9.11 \times 10^{-28}$ g		

The well widths were determined by the calculated emission energy of the n=1, e-hh transition (Fig.4) and were 2.5, 4.2 and 8.0 nm for the main peaks positions 1.8397, 1.6972 and 1.5905 eV for 4.2 K, respectively. A comparison between L_z measured by PL and based on the predetermined growth rate showed that agreement between them is not excellent. The difference between two values appears to have a dependence on well width. The maximum observed deviation was in wider wells with L_z >7 nm. The actual thickness L_z of QWs can be determined from cross-section TEM and can be used for PL calibration. This demonstrates the limitation of PL technique for a quick non-destructive indication of a QW thickness.

It is known that sharpness of a PL peak is a good indication of the quantum well quality (heterojunction abruptness and well width fluctuation). The full-widths-at-half-maximum (FWHM) of the main peaks are 24, 31, 26 meV at 77 K and 14,20,7 meV at 4.2 K for the wells with L_z 2.5, 4.2, 7.7 nm respectively. The calculated values assume that one monolayer fluctuation and thermal (Boltzmann) distributions of the carriers are comparable with our experimental results only in narrower wells. But our experimental data in wells with L_z =4.2 and 8.0 nm indicate that well width fluctuation is larger than one monolayer.

Combined application of X-ray diffraction and PL methods to the same samples made it possible to determine the composition, quality and structural properties of HS with thin layers down to single quantum wells.

5. ACKNOWLEDGEMENTS

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6. REFERENCES

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