Optical properties of GaN/AlN multi-quantum wells under high hydrostatic pressure: experimental investigations and *ab initio* study

A. Kaminska [1,2], P. Strak [3], J. Borysiuk [1], J. Z. Domagala [1], E. Grzanka [3], M. Beeler [4], K. Sakowski [3], E. Monroy [4], S. Krukowski [3,5]

[1] *Institute of Physics, Polish Academy of Sciences, Al. Lotników 32/46, 02-668 Warsaw, Poland*

[2] *Cardinal Stefan Wyszynski University, College of Science, Department of Mathematics and Natural Sciences, Dewajtis 5, 01-815 Warsaw, Poland*

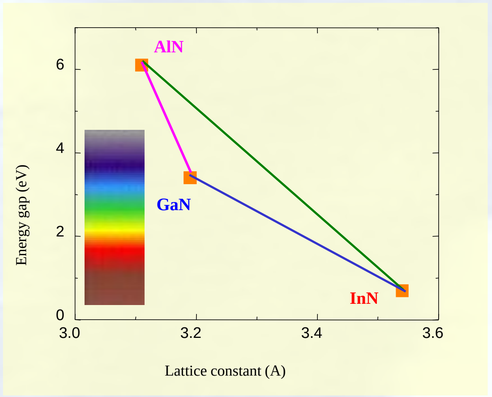
[3] *Institute of High Pressure Physics 'Unipress', Polish Academy of Sciences, Sokołowska 29/37, 01-142 Warsaw, Poland*

[4] *CEA Grenoble-INAC/SP2M/NPSC, 17 rue des Martyrs, 38054 Grenoble, France*

[5] *Interdisciplinary Centre for Mathematical and Computational Modelling, University of Warsaw, Pawinskiego 5a, 02-106 Warsaw, Poland*

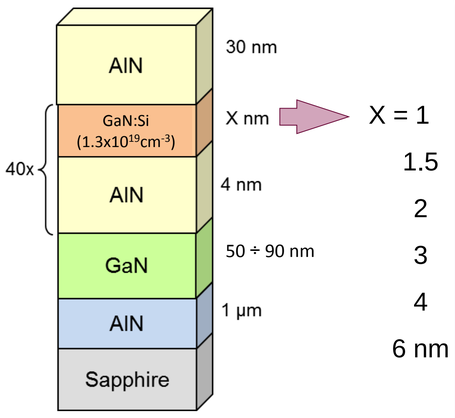
Motivation

- ✓ **Nitrides crystalize in wurtzite structure ⇒ the performance of nitride devices is hampered by the presence of strong spontaneous and piezoelectric polarization, and resulting built-in electric fields along the typical [0001] growth direction;**
  - **a built-in electric field reduces an overlap of electron-hole wavefunctions which causes lower optical output efficiencies of both LEDs and LDs.**
- ✓ **The problem of polarization is generally not well solved, and the proposed theoretical approaches provide results which do not have experimental verification;**
  - **the segregation problems in ternary InGaN and AlGaN alloys may lead to drastic differences in the optical properties of such layers creating the MQWs structures; segregation/clustering of In in AlInN and InGaN seems to play play a crucial role in the band structure and luminescence properties of these alloys**
- ✓ **High pressure PL measurements offer a possibility of investigations of the nature of the quantum states**



Our idea: to synthesize binary nitride MQWs systems which could be simulated theoretically and investigated experimentally.

Samples



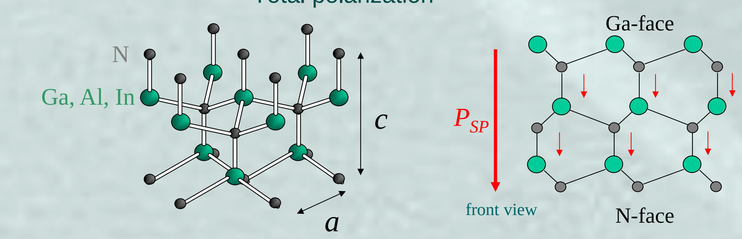
Grown by PA MBE on c-plane sapphire,
Mark Beeler, Eva Monroy, CEA Grenoble

Electric field effects in wurtzite quantum well heterostructures

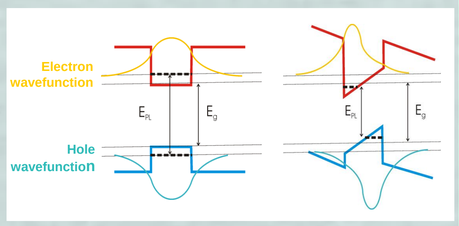
Piezoelectric polarization = = (strain) x (piezoelectric tensor)

Spontaneous polarization (in absence of strain)

Total polarization



- Spontaneous and piezoelectric polarizations in wurtzite heterostructures lead to interface charge and therefore induce built-in electric fields fields in quantum wells and heterostructures
- Quantum Confined Stark Effect:
  - ✓ spatial separation of the electron and hole wave functions in the wells
  - ✓ red-shift of the PL energy



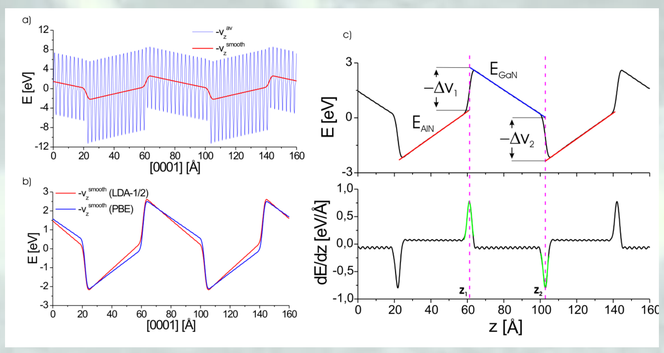
QW transition energy — electric field — exciton binding energy

$$E_{PL}=E_g+E_e+E_h-qLF-E_x$$

band gap — confinement energy — QW width

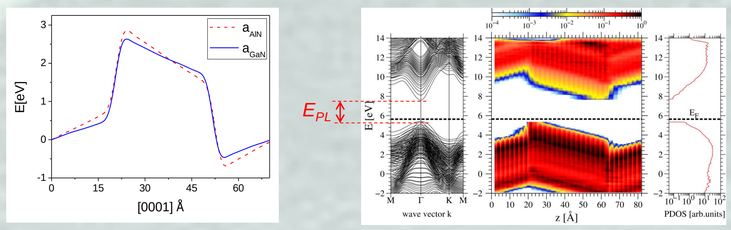
Ab initio calculations

- *Ab initio* calculations were performed for number of AlN/GaN MQWs using the Siesta software.
- The **a** lattice parameter was set equal to either bulk AlN or GaN in the separate calculations; it was fixed, and the structure was allowed to relax freely along **c** lattice parameter to minimize the elastic energy.
- It has been assumed that the Fermi level was controlled by the dominant silicon donor in the system, and the electric potential drop across the whole structure was close to zero.



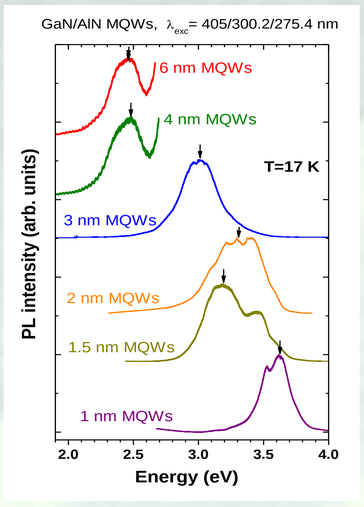
DFT results of 16 GaN/16 AlN layers (4 nm/4 nm) periodic QW system:
a) total electric potential (i.e. electron energy);
b) averaged smoothed potential, obtained from LDA-1/2 and PBE formulations;
c) the electric field determination in well/barrier interior;
d) derivation of a dipole layer localization at the heterointerface.

- In order to determine the effect of the built-in electric field, the potential profile was obtained by averaging in the plane perpendicular to c-axis: the potential slope changes as a function of the strain state, proving that piezoelectric effects related to the substrate-induced strain play an important role in the MQWs.

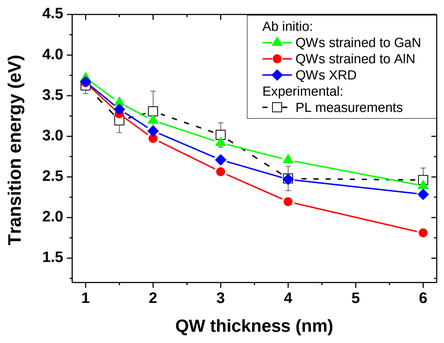


- The emission energy shift, related to change of the energy of quantum states caused by electric field (QCSE) was obtained directly from *ab initio* calculations

Dependence of PL energy on MQWs width at ambient pressure – experimental vs. theoretical data

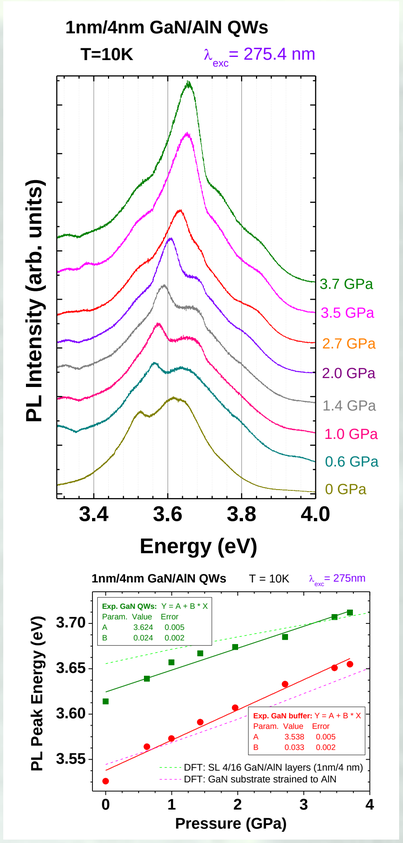


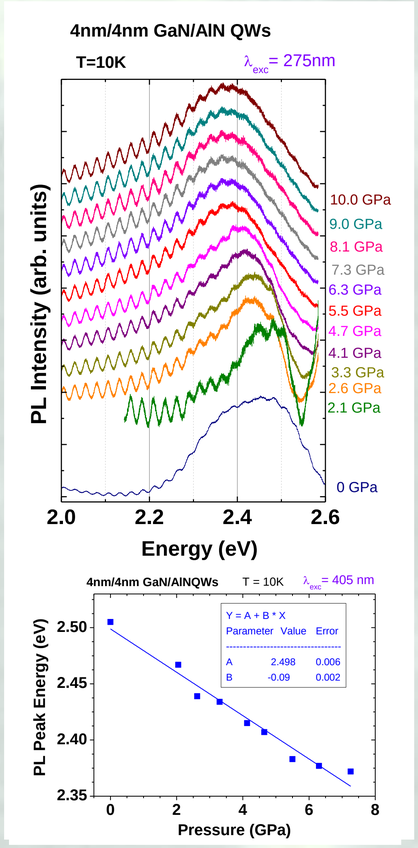
Normalized low-temperature PL spectra of GaN/AlN MQWs of different QW widths

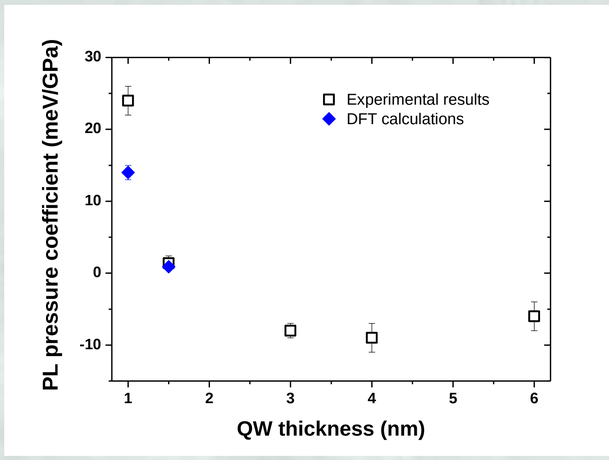


PL peak energies as a function of MQWs widths, determined experimentally (dashed lines and empty symbolds) and obtained using GGA-1/2 calculations (solid lines and full symbols).

High pressure dependence of PL spectra – experimental vs. theoretical data







PL peak pressure coefficient as a function of MQWs widths, determined experimentally (black open squares) and obtained using DFT calculations (blue diamonds)

Summary

- Good quality GaN/AlN relaxed multi-qauntum well structures were obtained by PA MBE growth.
- Due to the Quantum-Confined Stark Effect the ambient pressure PL peak energies of GaN/AlN MQWs decreased by over 1 eV, i.e. below the value of E_G(GaN), for QW widths increasing from 1 nm up to 6 nm (which is the fingerprint of the presence of internal electric field in the structure); this effect was accompanied by the drastic drop of the PL efficiency.
- The optical transition energies in these structures and their pressure coefficients obtained from *ab initio* calculations are in good agreement with experimental measurements.
- High pressure measurements show that pressure coefficients strongly depend on the well width.
- The obtained results indicate that piezoelectric effects constitute the important factors determining energy of transitions in nitride MQWs systems.
- Good agreement of theoretical predictions with experimental data proves correctness of the theoretical model, which can be useful in understanding and controlling the basic properties of nitride heterostructures.

Acknowledgements

The research was partially supported by the Polish National Science Centre by grants:
DEC-2011/03/D/ST3/02071
DEC-2011/03/B/ST5/02698
DEC-2012/05/B/ST3/03113