Influence of strain on the excitonic bandgap of AlN epitaxial layers grown on Si and sapphire substrates

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Motivation

- ✓ Studies of the near band-edge optical properties of AlN layers have been restricted due to technical difficulties involved with optical measurements in the deep UV range.
- ✓ Synchrotron radiation gives a great possibility to obtain unique experimental data shedding new light on the important properties of the compound that is intensively explored recently.

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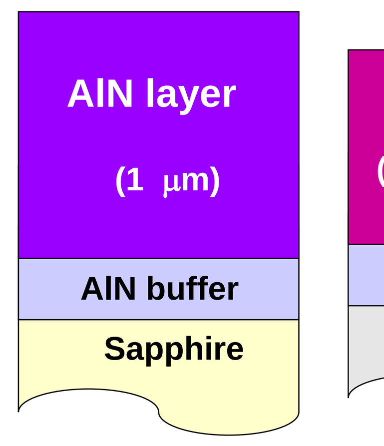
Aim of the work

- ✓ The study of defect-related photoluminescence (PL) and photoluminescence excitation spectra (PLE) of the series of AlN layers with different dislocation densities, grown on sapphire or silicon substrates.
- ✓ Our goal:
  - to to determine experimentally an impact of the substrate type and dislocation densities on the AlN bandgap and its emission properties,
  - to compare experimental results with theoretical analysis using DFT calculations.

Samples



Grown by PA MBE:

1) on c-plane sapphire
2) on silicon substrate

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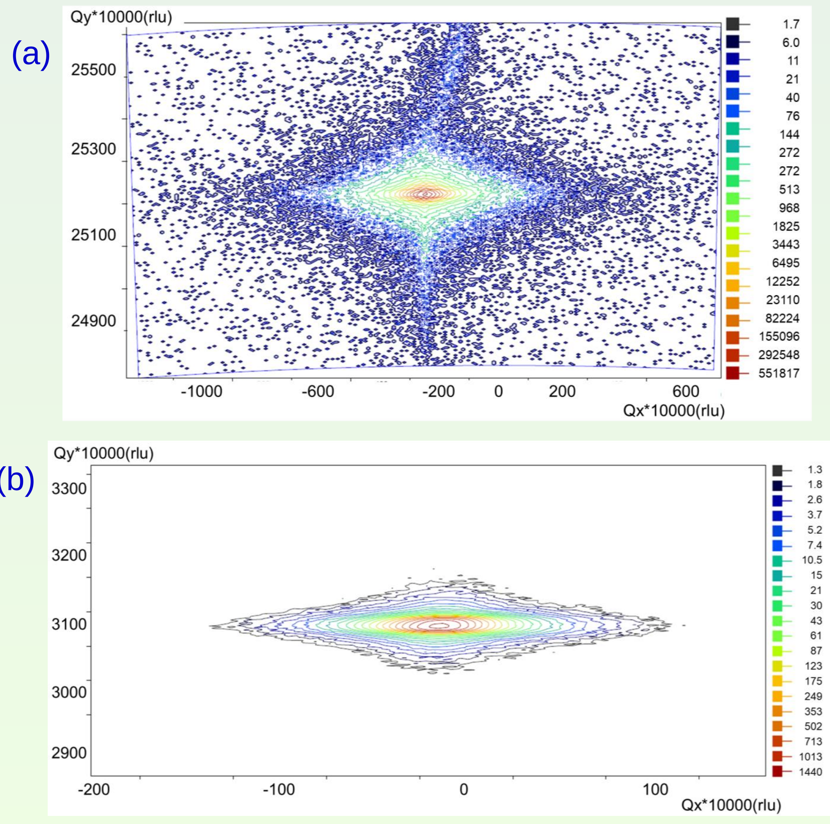
Structural characterisation: High-Resolution X-Ray Diffraction (HRXRD)



Exemplary reciprocal space maps of symmetrical 0002 AlN reflection for (a) AlN/sapphire and (b) AlN/silicon

Type of the substrate, sample names, dislocation densities, lattice parameters, in-plane strains (ε_{xx}) and out-of-plane strains (ε_{zz}) of all samples

Substrate	Sample name	Screw dislocations density (10^9 cm^{-2})	Edge dislocations density (10^9 cm^{-2})	a (Å)	c (Å)	ε_{zz} (10^{-4})	ε_{xx} (10^{-4})
Sapphire	Sa-1	0.22	0.938	3.1033	4.9872	16.3	-27.9
Sapphire	Sa-2	0.48	3.3	3.1034	4.9867	16.1	-27.3
Sapphire	Sa-3	0.82	3.2	3.1108	4.9795	1.8	-3.5
Sapphire	Sa-4	1.0	5.5	3.1080	4.9851	8.8	-15.1
Silicon	Si-1	7.8	28	3.1186	4.9739	-11.6	20.2
Silicon	Si-2	20	60	3.1206	4.9735	-14.6	25.4

Ab initio calculations

- *Ab initio* calculations were performed for the Sa-2 and Si-1 samples using the VASP package.
- The **a** lattice parameter was set equal to that determined by HRXRD measurements; it was fixed, and the structure was allowed to relax freely along the **c** lattice parameter to minimize the elastic energy.
- The calculated values of the bandgap energies are equal to **6.33 eV**, and **6.16 eV** for the AlN/sapphire (Sa-2) and AlN/silicon (Si-1), respectively.
- Additionally, energy gaps for layers on different substrates were calculated according to "model-solid theory": it was assumed that wurtzite AlN thin films grown along the **c**-direction on sapphire or silicon substrates are subjected to biaxial stress induced by the substrate.
- Under such stress, the wurtzite system exhibits biaxial strain in the **c** plane accompanied by the out-of-plane strain along the **c** axis:

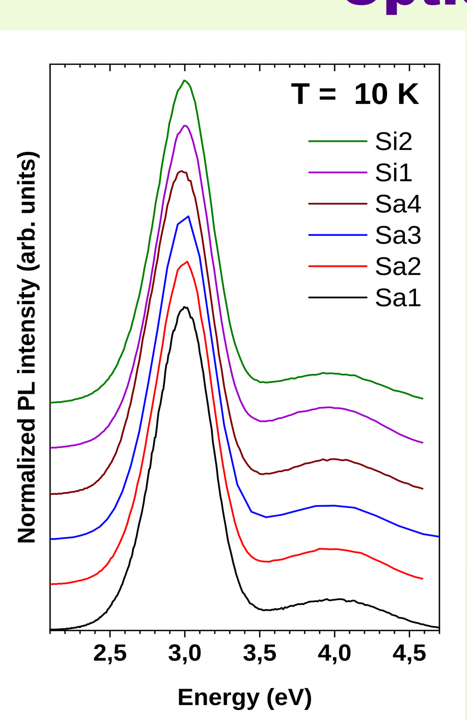
$$\varepsilon_{zz} = -2\frac{C_{13}}{C_{33}}\varepsilon_{xx}$$

- Using the elastic constants C_{13} and C_{33} obtained by DFT calculations performed by I. Vurgaftman et al.[1] bandgap energies were evaluated according to the model presented by Q. Yan et al.[2]: for AlN/sapphire, it was **6.28 eV**, whilst for AlN/silicon, it was **6.13 eV.**

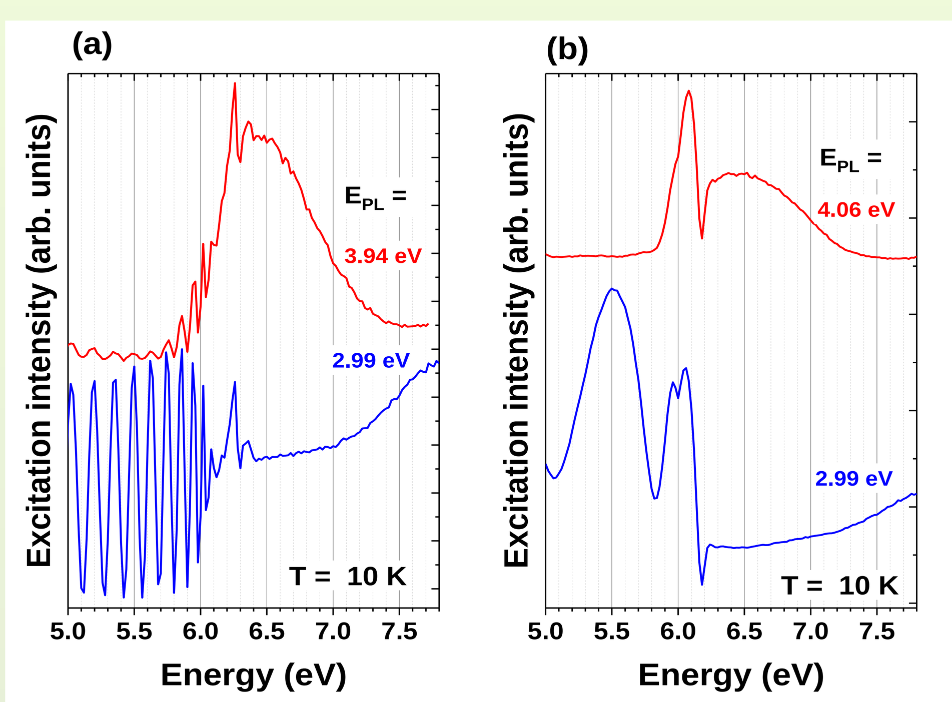
[1] I. Vurgaftman and J. R. Meyer, J. Appl. Phys. **94**, 3675 (2003).

[2] Q. Yan, P. Rinke, A. Janotti, M. Scheffler, and C. G. Van de Walle, Phys. Rev. B **90**, 125118 (2014).

Optical properties of AlN layers @ 10 K



Deep defect-related low-temperature PL spectra of AlN/Al_2O_3 and AlN/Si samples, excited by the synchrotron radiation with the energy of around 6.35 eV or 6.10 eV, respectively.



PLE spectra of the AlN layers (a) Sa-2 and (b) Si-1, monitored at defect-related emission energy of around 4 eV (upper red line) and 3 eV (lower blue line). The Fabry-Pérot oscillations are associated with interference effects.

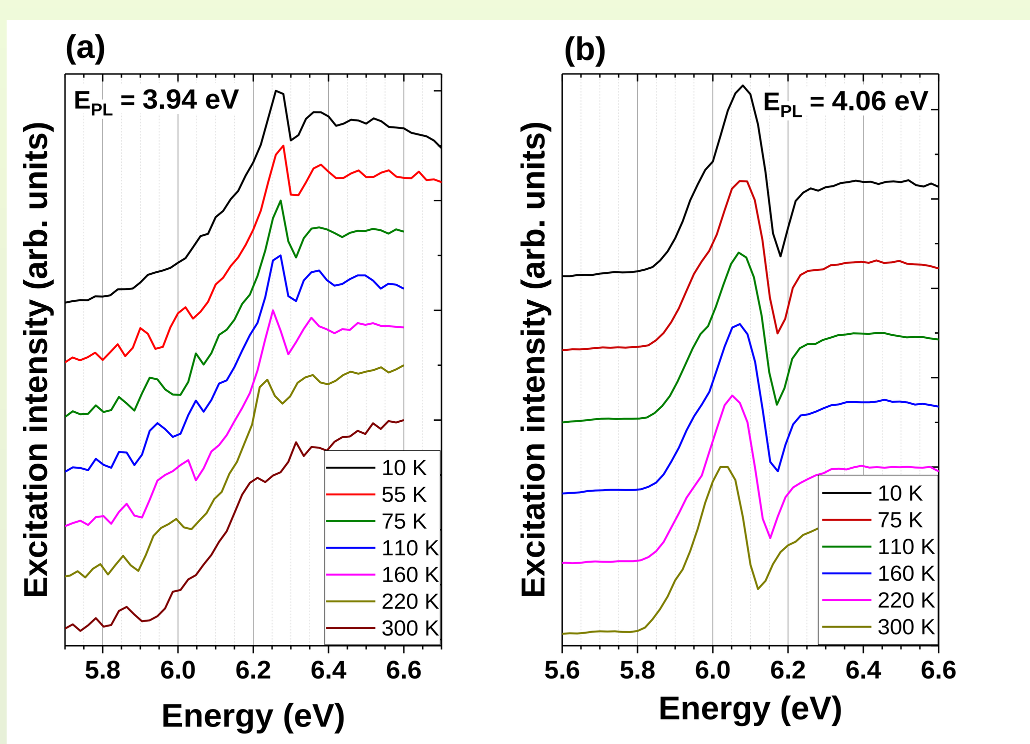
The two broad PL bands of deep defect-related transitions in the AlN are connected the most probably with the presence of Al vacancies (V_{Al}):

- **the band around 3 eV was ascribed by Harris et al.[3] to $(V_{Al}\text{-}O_N)^{2-}$ or $(V_{Al}\text{-}Si_{Al})^{1-}$ complex through DFT calculations,**
- **the band around 4 eV was identified by Sedhain et al.[4] as a donor-acceptor-pair type transition involving a shallow donor and $(V_{Al}\text{-}O_N)^{2-/1-}$ or $(V_{Al}\text{-}Si_{Al})^{2-/1-}$ complex.**

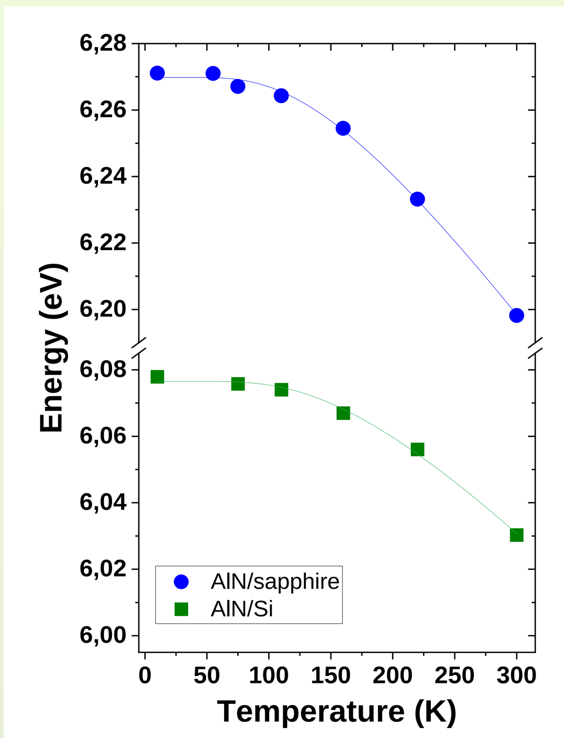
[1]J. S. Harris et al., Appl. Phys. Lett. **112**, 152101 (2018).

[2] A. Sedhain, J. Y. Lin, and H. X. Jiang, Appl. Phys. Lett. **100**, 221107 (2012).

Temperature dependence of optical properties of AlN layers



Temperature dependence of the PLE spectra of the AlN layers (a) Sa-2 and (b) Si-1, monitored at defect-related emission energy of around 4 eV.



Temperature dependence of A excitonic peak energies in AlN/sapphire (Sa-2) and AlN/silicon (Si-1,). Solid lines are the fits using the expression given by Viña et al.[5].

Fitting parameters of the Bose-Einstein expression for the variation of the excitonic bandgap with temperature

Sample	E_0 (eV)	α (meV)	Θ (K)
Sa-2	6.270±0.001	123±21	448±39
Si-1	6.077±0.001	112±30	532±66

$$E(T) = E_0 - \frac{2\alpha}{\exp(\Theta/T) - 1}$$

[5] L. Viña, S. Logothetidis, and M. Cardona, Phys. Rev. B **30**, 1979 (1984).

Considering the AlN exciton binding energy of 58 meV ⇒ the determined bandgap energies of the AlN samples are equal to 6.328 eV for AlN/sapphire (Sa-2), and 6.135 eV for AlN/silicon (Si-1).

Summary

- **We demonstrated the possibility of using synchrotron radiation and defect-related PL and PLE measurements to study the excitonic bandgap of AlN epitaxial layers ⇒ these measurements allow determination of the bandgaps of the investigated AlN samples and their temperature dependencies, and to compare them with the results obtained so far by other techniques.**
- **The structural analysis revealed significant dependence of the dislocation densities and strain directions in the AlN layer on the substrate used for the growth.**
- **The optical results revealed that AlN bandgap energies are dependent on the substrate type, and independent of the dislocation density.**
- **The bandgap energies obtained by *ab initio* calculations are in very good agreement with experimental data.**
- **The obtained results indicate that the dependence of bandgap energy of AlN layers on a substrate is induced by the tetragonal strain related to the lattice mismatch between the substrate and the AlN layer[6]. This effect has a strong influence on the spectral positions of the intrinsic excitons, and consequently on the bandgap of AlN layers.**

[6] A. Kaminska, K. Koronski, P. Strak, A. Wierzbicka, M. Sobanska, K. Klosek, D. V. Nechaev, V. Pankratov, K. Chernenko, S. Krukowski, Z. R. Zytkiewicz, Appl. Phys. Lett. **117**, 232101-1-6 (2020).

Acknowledgements

The authors acknowledge the financial support from the Polish National Science Center under grant 2021/43/D/ST7/01936 and the MAX IV Laboratory for time on Beamline FinEstBeAMS under Proposal 20180493. Research conducted at MAX IV, a Swedish national user facility, is supported by the Swedish Research Council under contract 2018-07152, the Swedish Governmental Agency for Innovation Systems under contract 2018-04969, and Formas under contract 2019-02496. We would like to thank the FinEstBeAMS beamline staff for expert assistance during the experiments.

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