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Predictions for Yellow Luminescence in Cubic Gallium Nitride Under Pressure

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Outline

- Motivation
  - Background on Yellow Luminescence
  - Prior calculated results
- Method for calculation
  - LMCC in one minute
  - Defect levels and vertical transitions
- LMCC Results for Cubic GaN
  - Defect Band structure
  - Frontier states under pressure: Predicted band gap
- Pressure Results for selected defects
  - V_{Ga}
  - C_N
- Conclusion

Motivation

Yellow Luminescence in w-GaN

- Observed in OMCVD GaN films
- Scales with carbon content
- Numerous theoretical and experimental model systems
  - V_{Ga}
  - V_{Ga}:H_3
  - V_{Ga}-O_N:H_2
  - C_N
  - C_N-O_N

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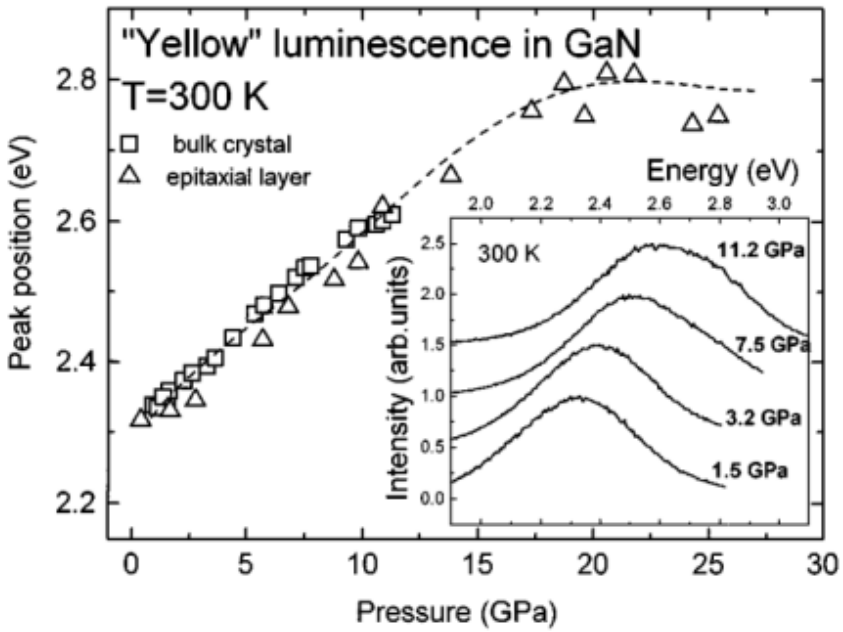
Blue indicates the defect is considered in this work

Motivation

Yellow Luminescence in w-GaN

Suski *et al.* (Appl. Phys. Lett. **67** 2188 (1995))
Pressure dependent PL in w-GaN

- dE_g/dP = 40meV/GPa
- dE_{PL}/dP = 30 meV/GPa
- Significant broadening increase with pressure
  => multiple defects?



Clear need for pressure dependent theory

Method of Calculation

Local Moment Counter Charge (LMCC) DFT

- Current practice:Hybrid functionals including exact exchange
  - Fit Kohn-Sham gap to experiment

- Underlying argument:
  Only the correct K-S band gap can lead to accurate defect levels.

- LMCC argument:
  - *The K-S gap is mostly irrelevant (qualitative test of localization)*
  - *Defect levels can be accurate using only total energies*
  - *Calculate defect levels from ionization potentials referenced to a common level.*

A supercell theory of defect energies

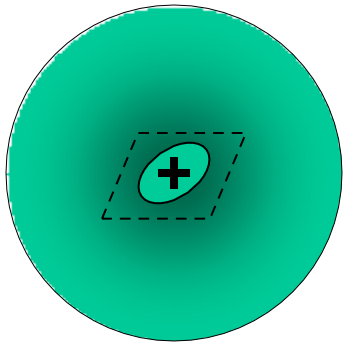
Peter A. Schultz, Phys. Rev. Lett. **96**, 246401 (2006).

Target system: isolated defect

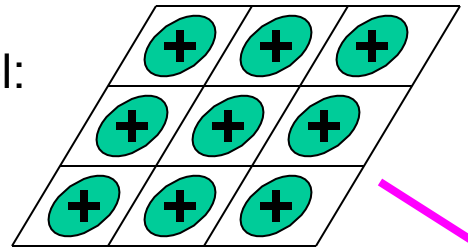
=

Computational model for isolated defect

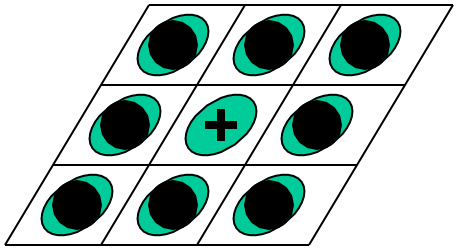
(+ DDO for defect banding)



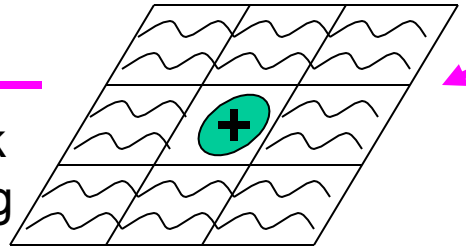
Standard DFT model: Supercell



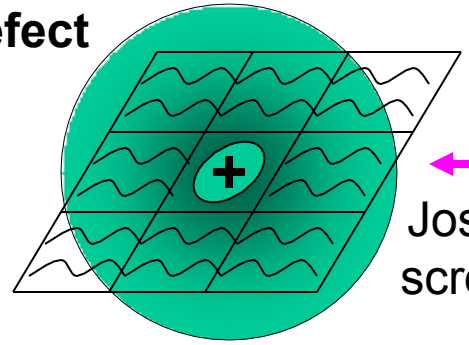
LMCC to fix boundary conditions



Crystal embedding to fix μ_e



Jost Bulk screening



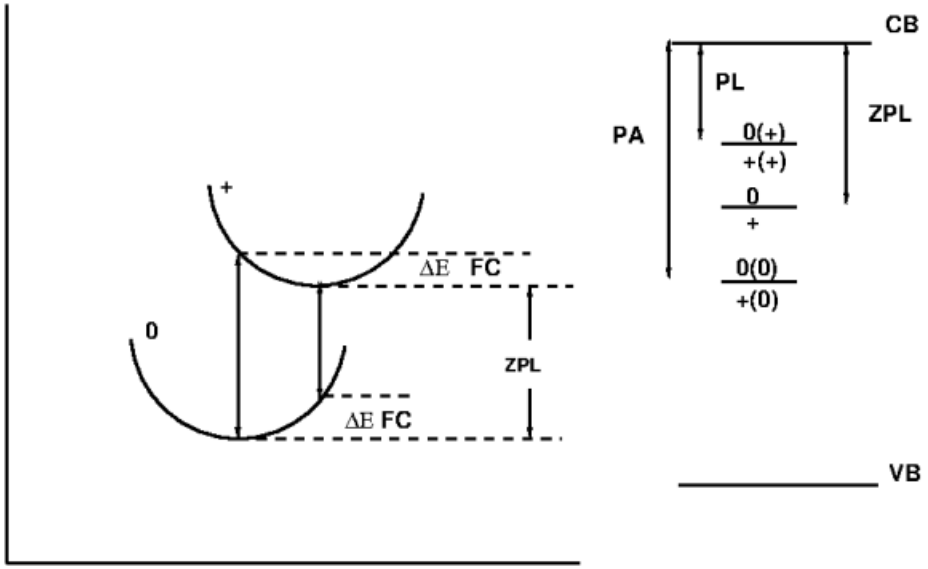
Finite Defect Supercell Model

"*Ab initio*" *computational model* – connect model to physics
Calculations with rigorous control of charge boundary conditions

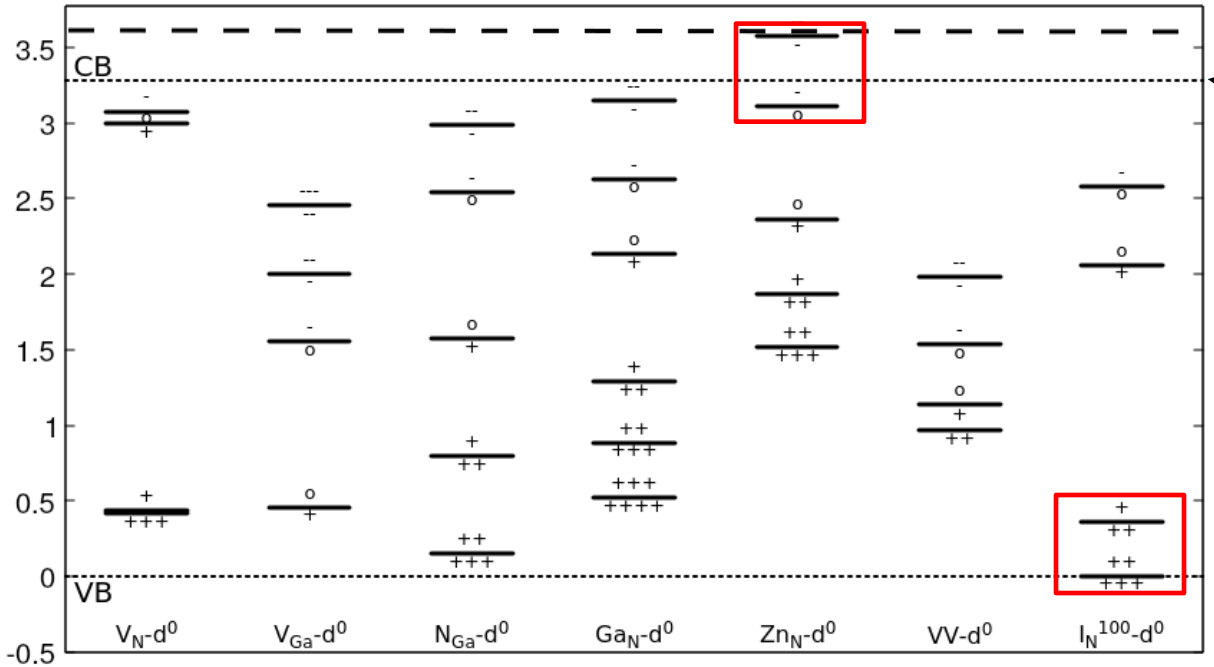
(*i.e.*,not jellium-based)

Method Summary

- Calculate defect level spectra for intrinsic and extrinsic defects as function of supercell size (64, 216, 512, and 1000 atom for c-GaN)
  - E_g from total breadth from all defects
- For chosen defects, use standard CC diagram method to calculate:
  - Zero phonon line (ZPL) from thermodynamic levels
  - Frank-Condon shifts
- Perform same calculation at 12 Gpa (0.98 scale of lattice parammeters)



c-GaN Defect Band Gap

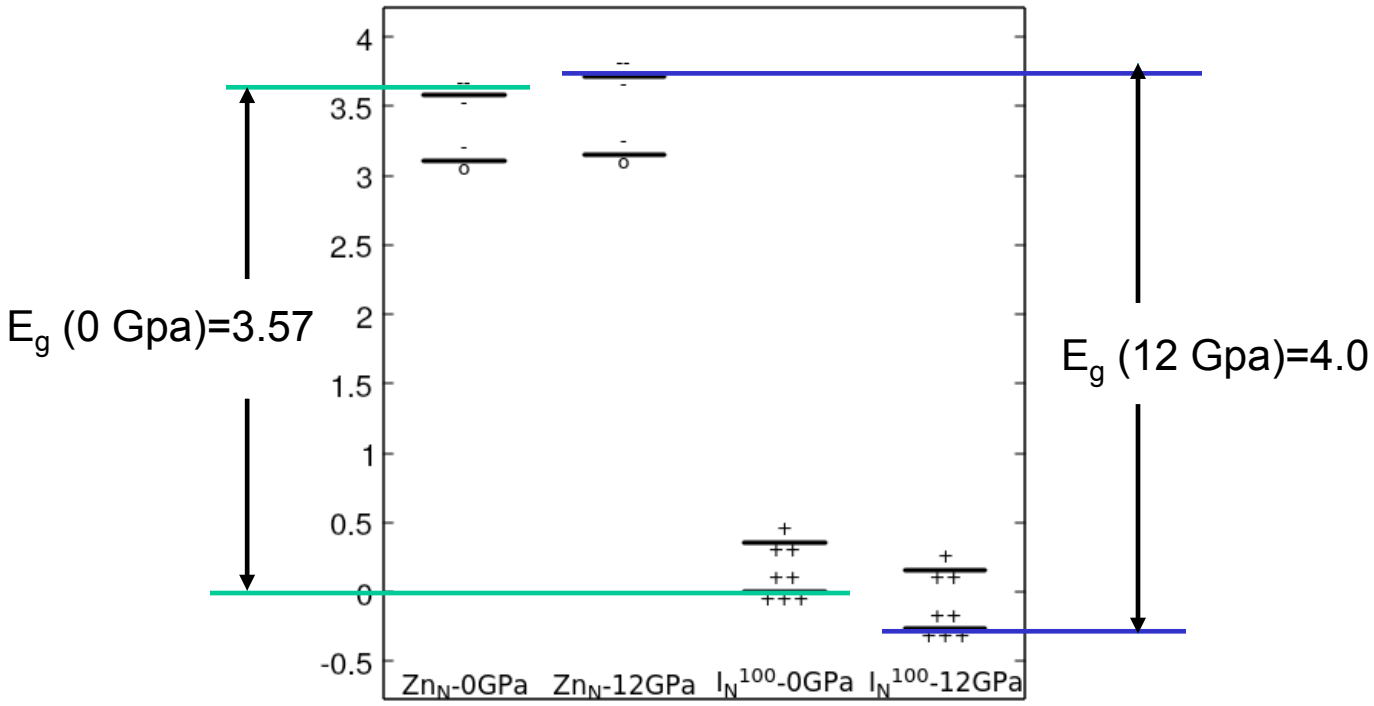


Exp. CB (c-GaN)

theoretical band edges

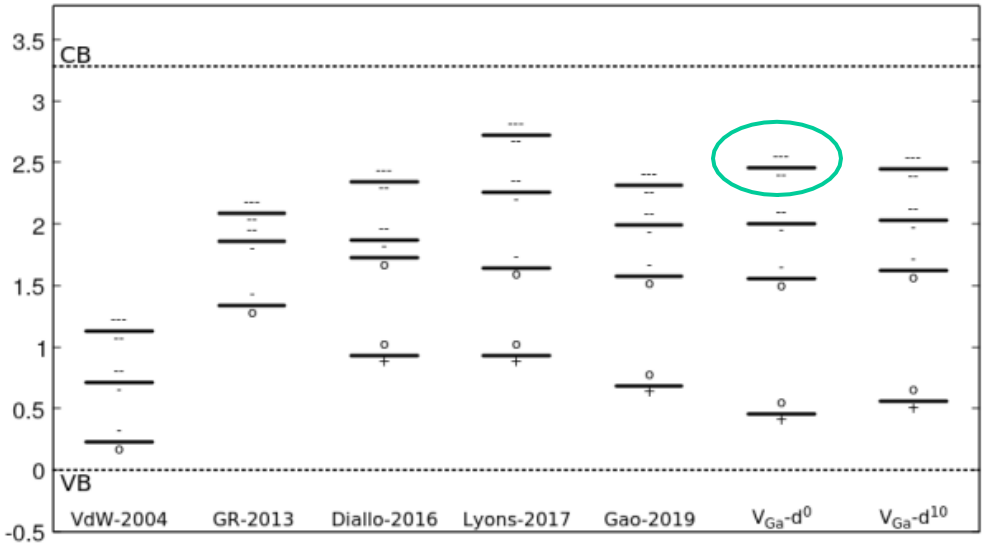
- LMCC band gap ~10% too large (no fitting)
- Kohn-Sham band gap = 2.4 eV
- Frontier levels are lower and upper bounds for band edges

c-GaN Defect Band Gap 12 GPa



- Levels calculated at 0 Gpa and 12 GPa
- dEg/dP (LMCC)=36 meV/GPa
- dEg/dP (KS)=41
- PL peak measured relative to either extremal level

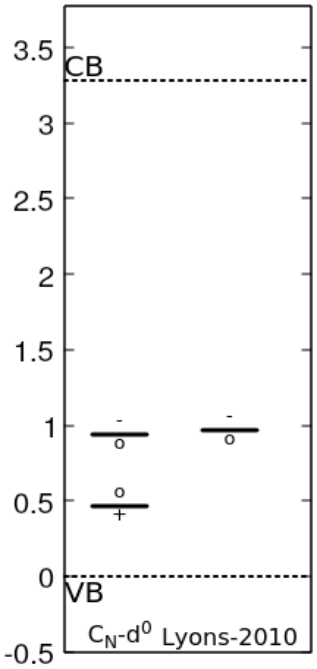
V_{Ga}



- YL from hole absorption/emission from 3-/2- defect level
- ZPL position varies across theory by (0.4 eV)

	0 GPa	12 GPa	Δ	dE/dP (meV/Ga)
ZPL (eV)	2.39	2.71	.323	
PL (eV)	2.19	2.51	.319	26.5
FCS (eV)	0.199	0.204	.005	

C_N

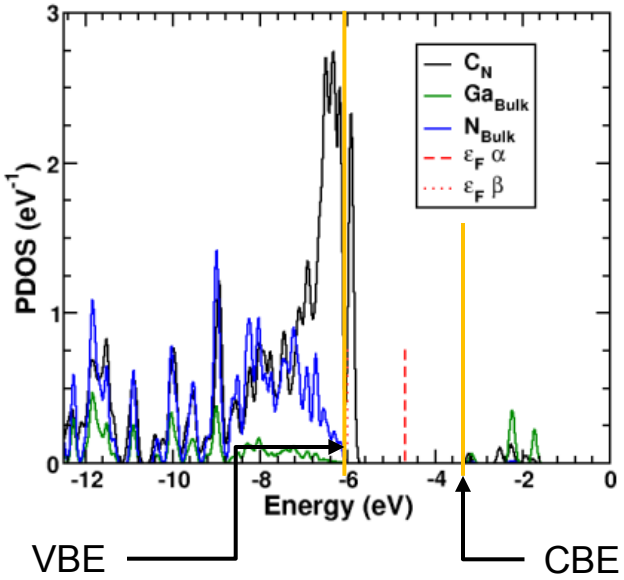


- YL from hole absorption/emission from -1/0 defect level
- wrt VBE, -1/0 levels within 28 meV
- ZPL's differ because of difference in CBE

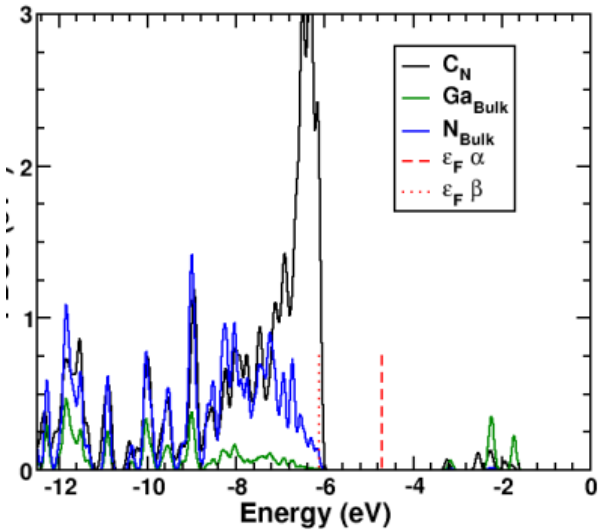
	0 GPa	12 GPa	Δ	dE/dP (meV/Ga)
ZPL (eV)	2.62	3.06	.44	
PL (eV)	2.247	2.70	.45	45.8
FCS (eV)	0.374	0.355	.020	

C_N-O_N

+1 at +1 eq. geometry

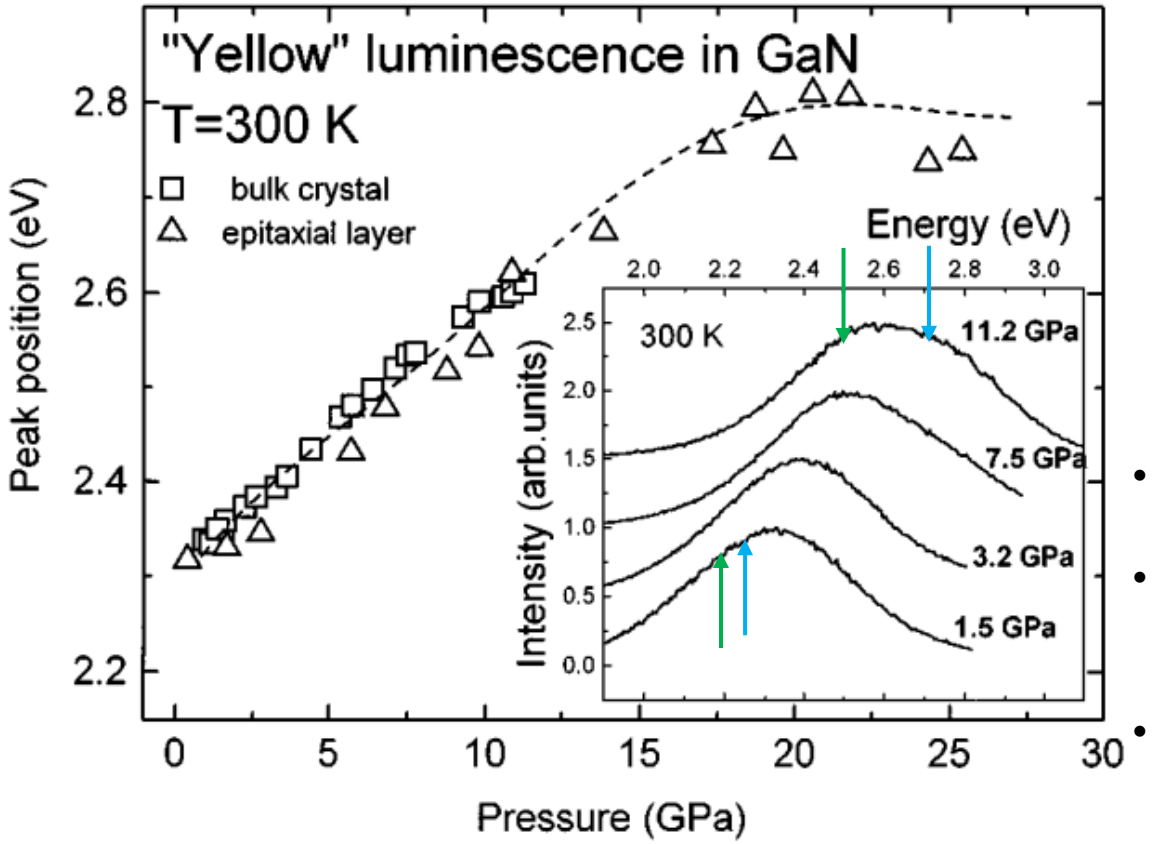


+1 at 0 eq. geometry



- Frank-Condon transition would be into VB
- We can calculate ZPL, but not the FCS
- Band-to-band Luminescence

Experimental Comparison



V_{Ga}
C_N

- Reasonable agreement for 0 Gpa and 12 Gpa
- Very good qualitative agreement for broadening
- Supports that multiple defects contribute

Conclusions

- LMCC method yields c-GaN bandgap within 10% of experiment
- 12 GPa frontier levels predicts band gap enlarged by 0.42 eV
  (in good agreement with experiment)
- V_{Ga} and C_N strong candidates for YL (in agreement with others)
- Surprising quantitative agreement with Lyons et al. for PL peaks
  - Different crystal phase
  - Different band gap
  - Cancelling differences
- V_{Ga} and C_N exhibit different values for dE_{PL}/dP
  - Values bracket experimental value for overall YL pressure shift
  - Qualitative agreement with PL broadening and shape change
- We do not predict same PL from CN-ON: +1 (0) peak buried in VB