Pressure evolution of localized nitrogen cluster states in GaAsN alloys

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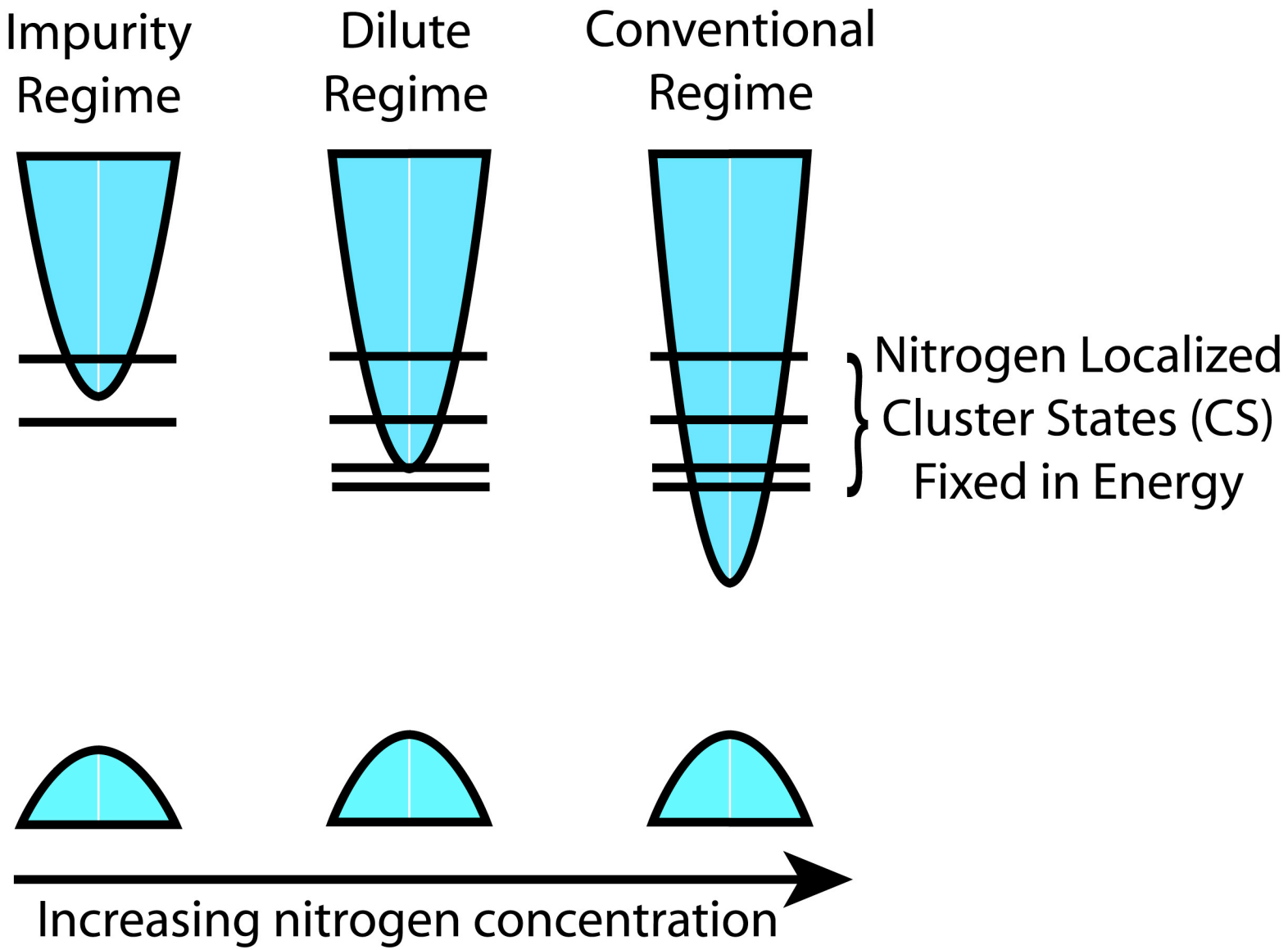
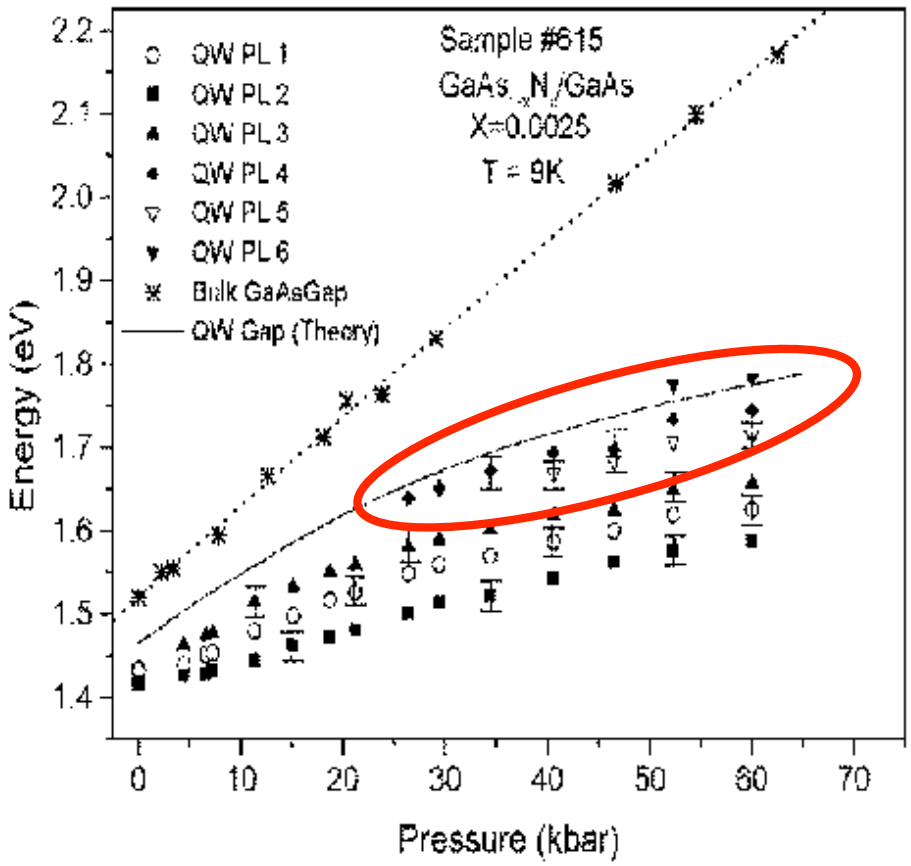
Localized-Delocalized Transition in GaAsN

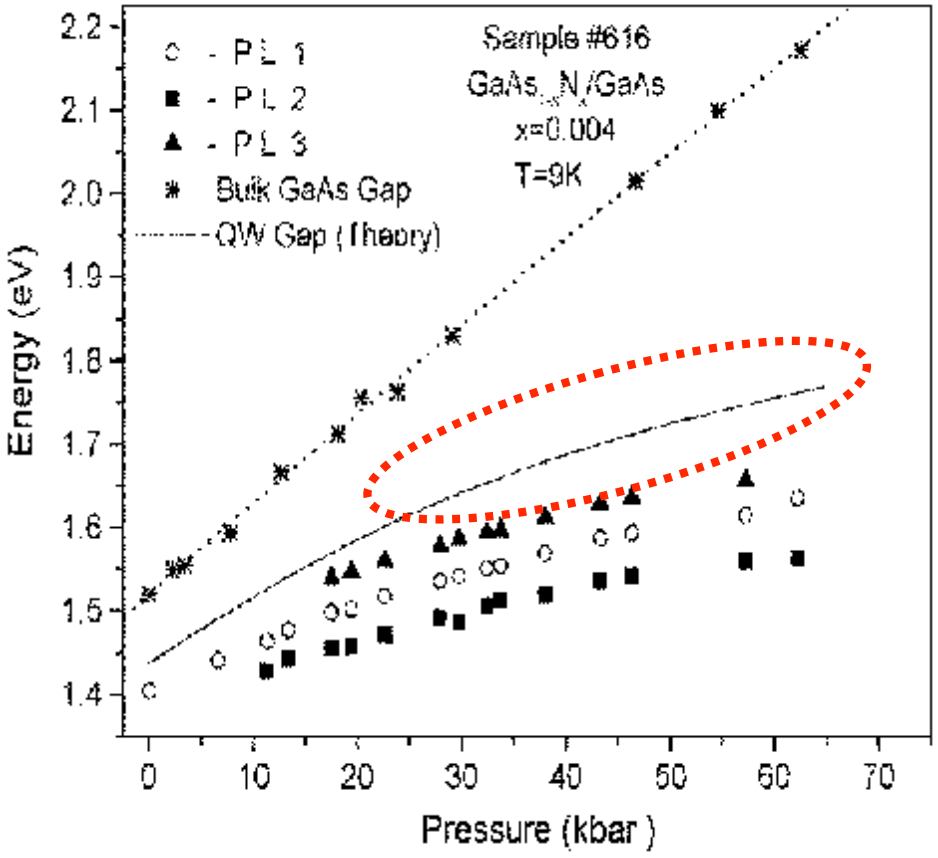
Kent and Zunger PRL **86** 2613 (2001)



Experimental Pressure & Composition Evolution

Courtesy B. Weinstein (Buffalo) ICPS2002, APS2002



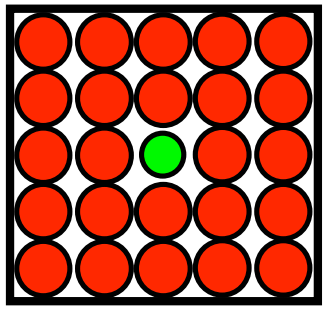


Pressure coefficient of CS reduced compared to bulk
Disappearance of highest energy CS!

How do the nitrogen CS evolve with pressure and composition?

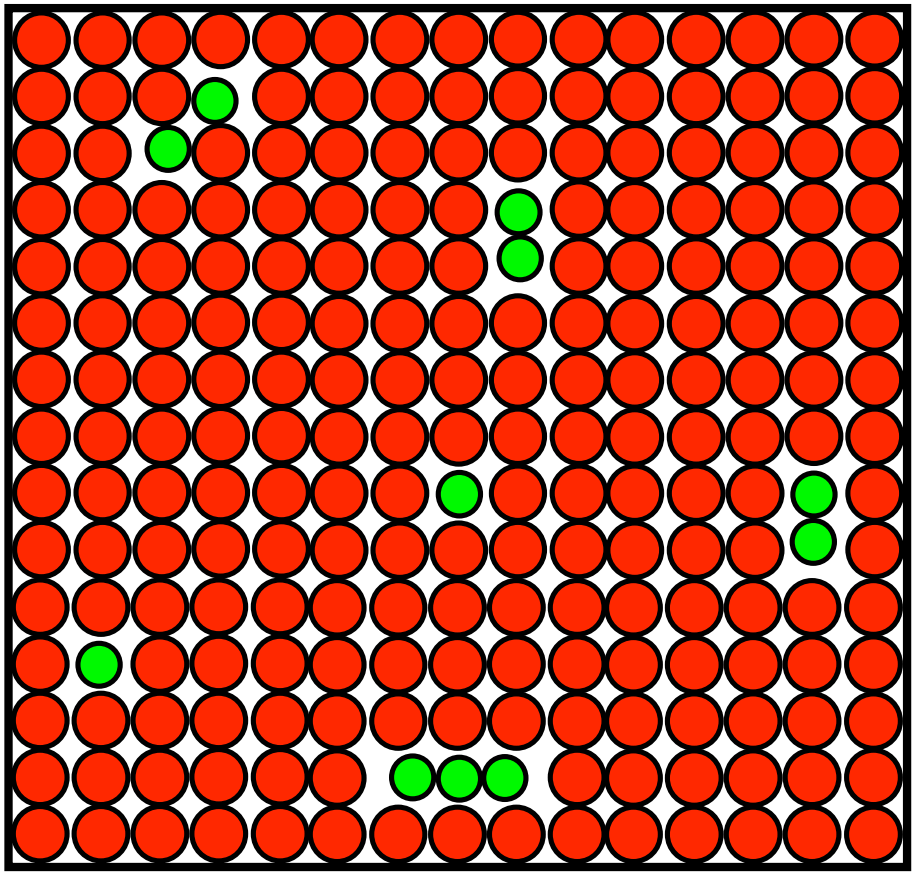
Computational Modeling of Dilute Alloys

Kent and Zunger PRB **64** 115208 (2001)



Small Supercell Approach

Large Supercell Approach

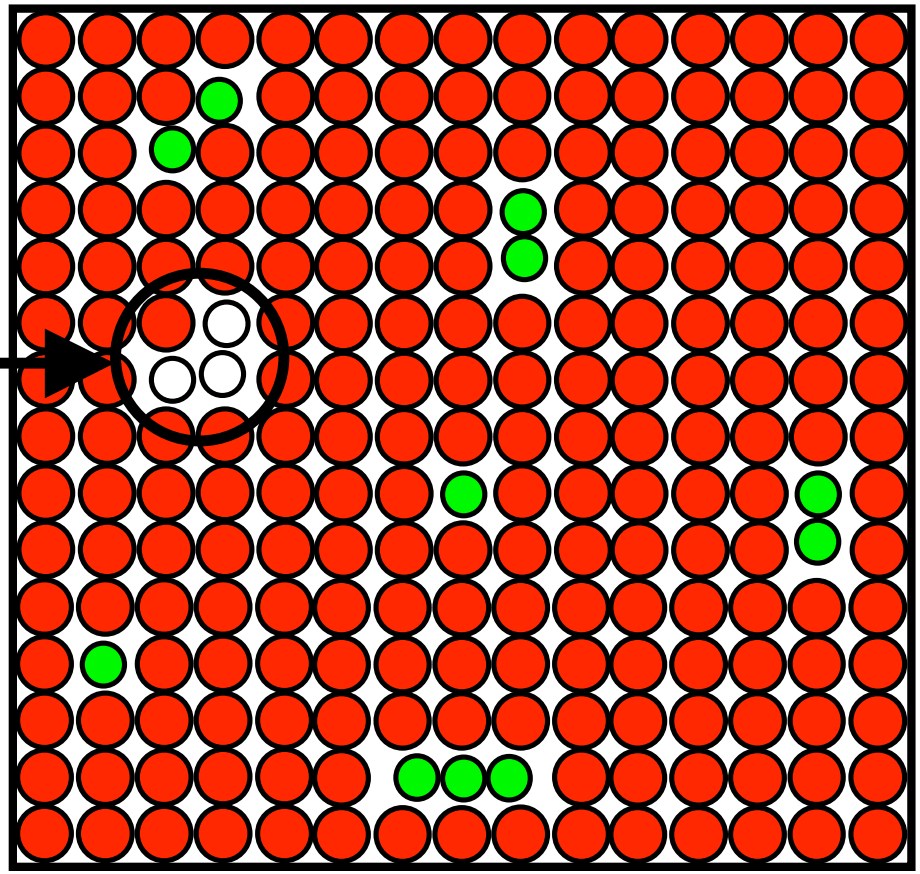


- Use large supercells (10^3-10^6 atoms) containing many nitrogens
- Statistically average properties of many random configurations
- Use VFF for structural relaxation
- Use Empirical pseudopotential method for wavefunctions

Computational Modeling

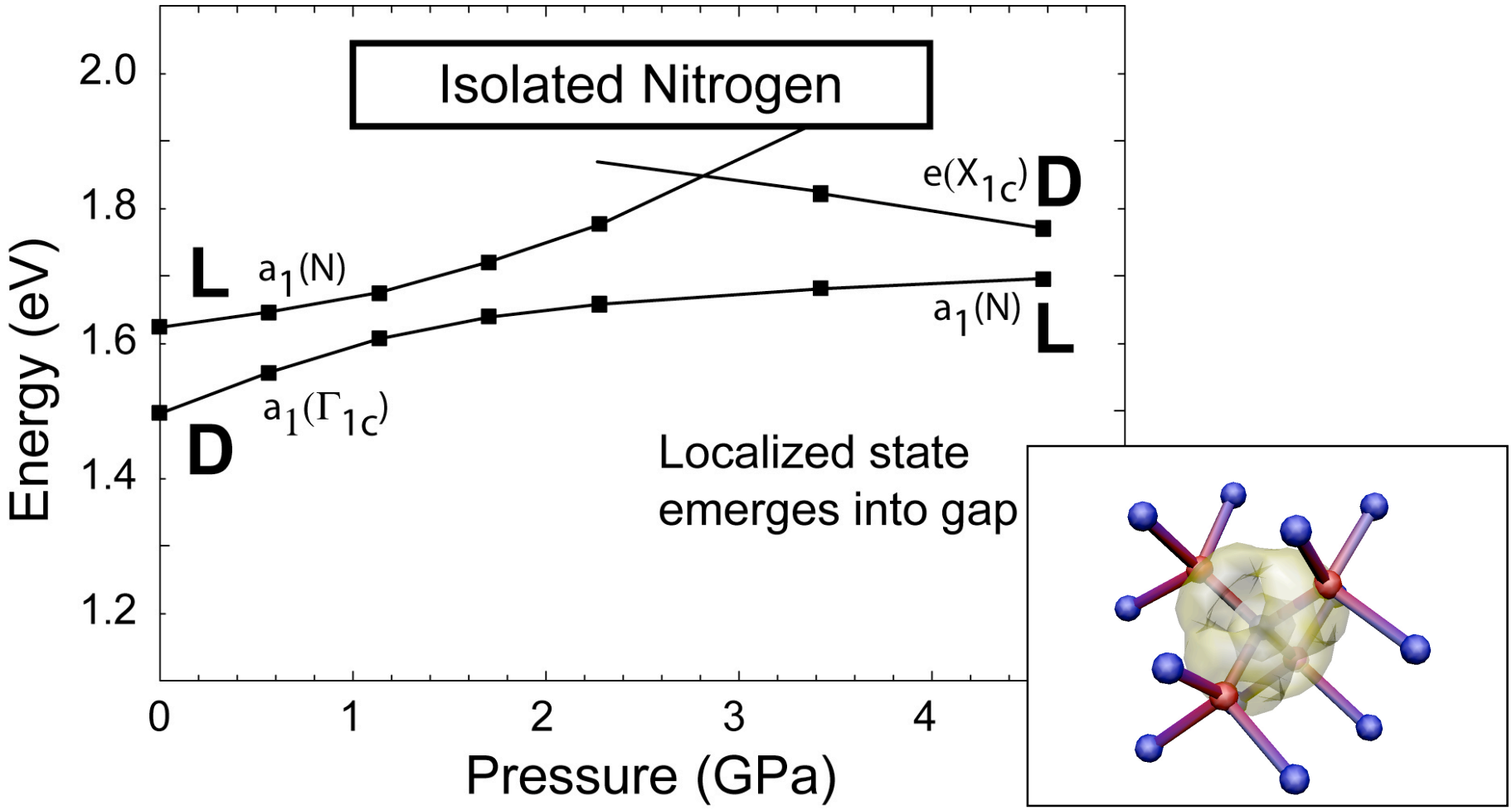
1. Create nitrogen cluster in dilute alloy

2. Follow localized state with pressure & nitrogen concentration



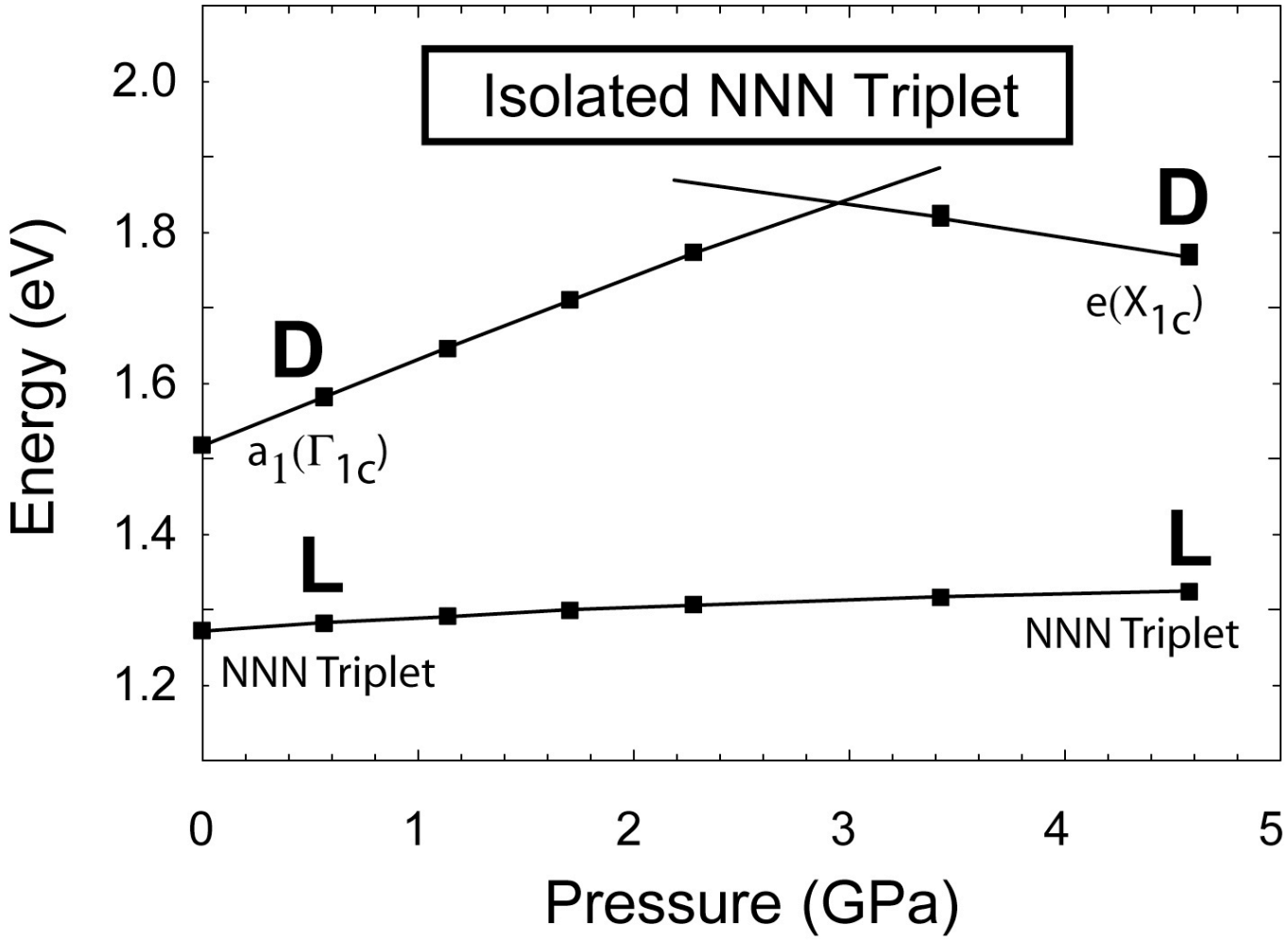
64000 atom supercells

Isolated nitrogen in bulk GaAs



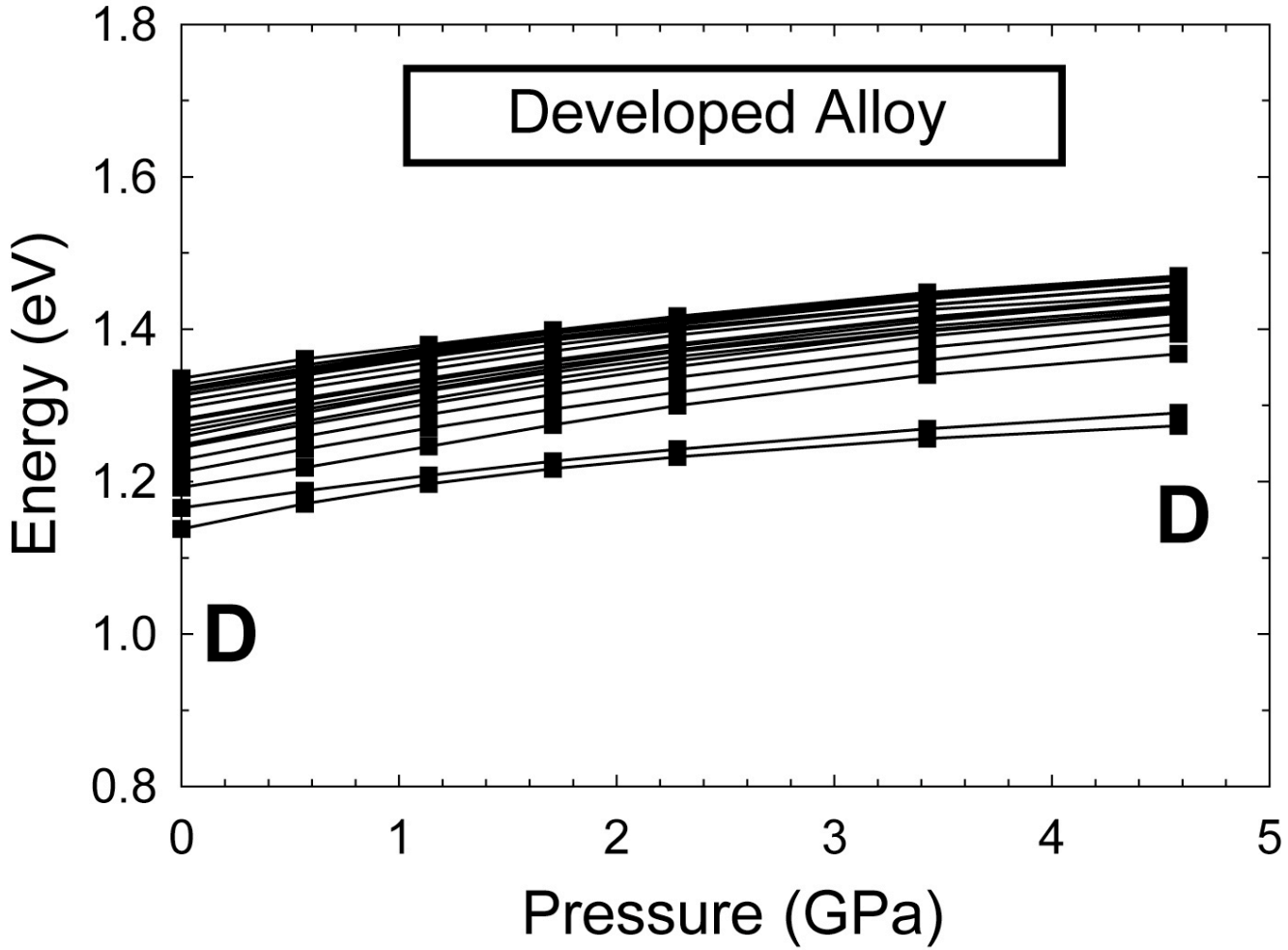
L=Localized D=Delocalized

Nitrogen triplet in Bulk GaAs



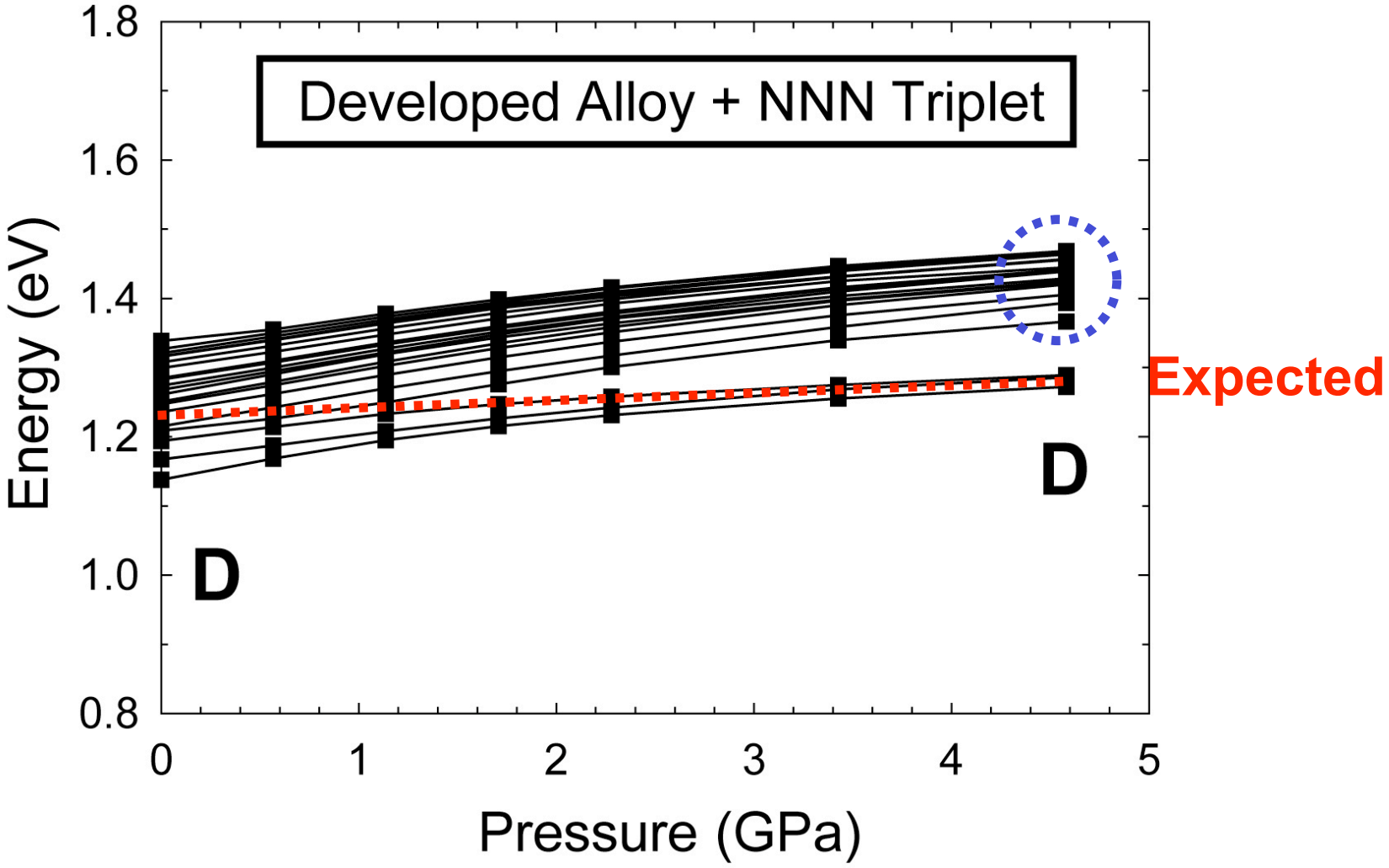
Localized state remains in gap
Low pressure coefficient

Developed alloy

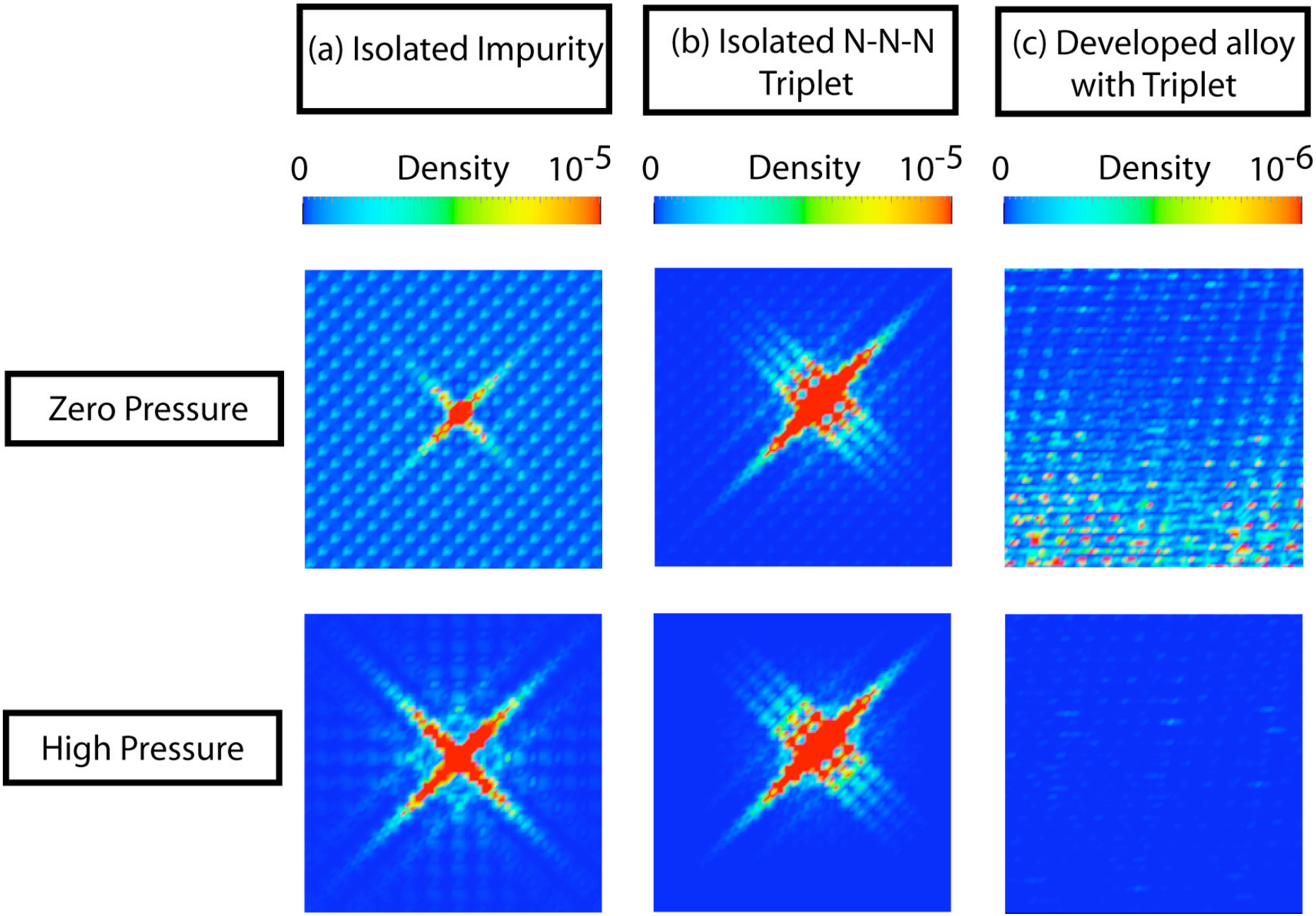


Delocalized states at band edges

Developed alloy with triplet



Delocalized states at band edges
Triplet not within gap or at band edge, as expected



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Summary

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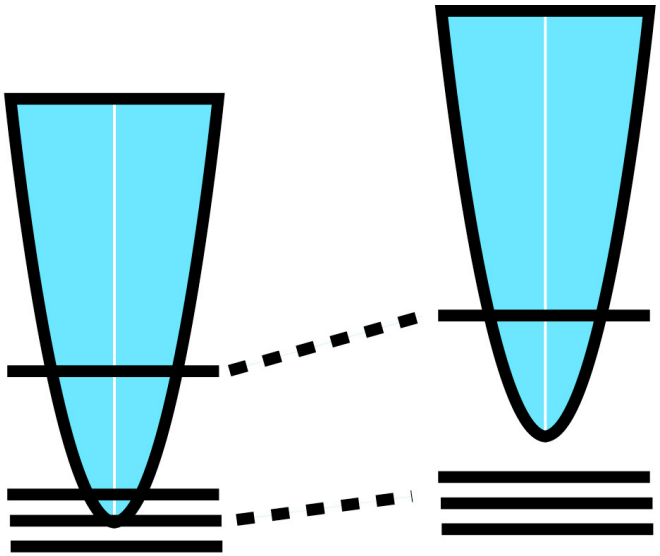

Highest energy CS
Increased pressure coeff.

Lowest energy CS
Small pressure coeff.

Increasing Pressure
Fixed nitrogen concentration

Model Comparison

Band anticrossing Good fit of band edge bowing
No cluster states (CS)

Impurity band Predicts broadening of localized states
→ ***Pressure*** exposes either broad impurity band, or nothing
→ ***No discrete CS to expose***

Atomistic model (This work)
Conduction states overtake discrete CS
Some hybridization at higher pressures
Consistent with experiment

Summary

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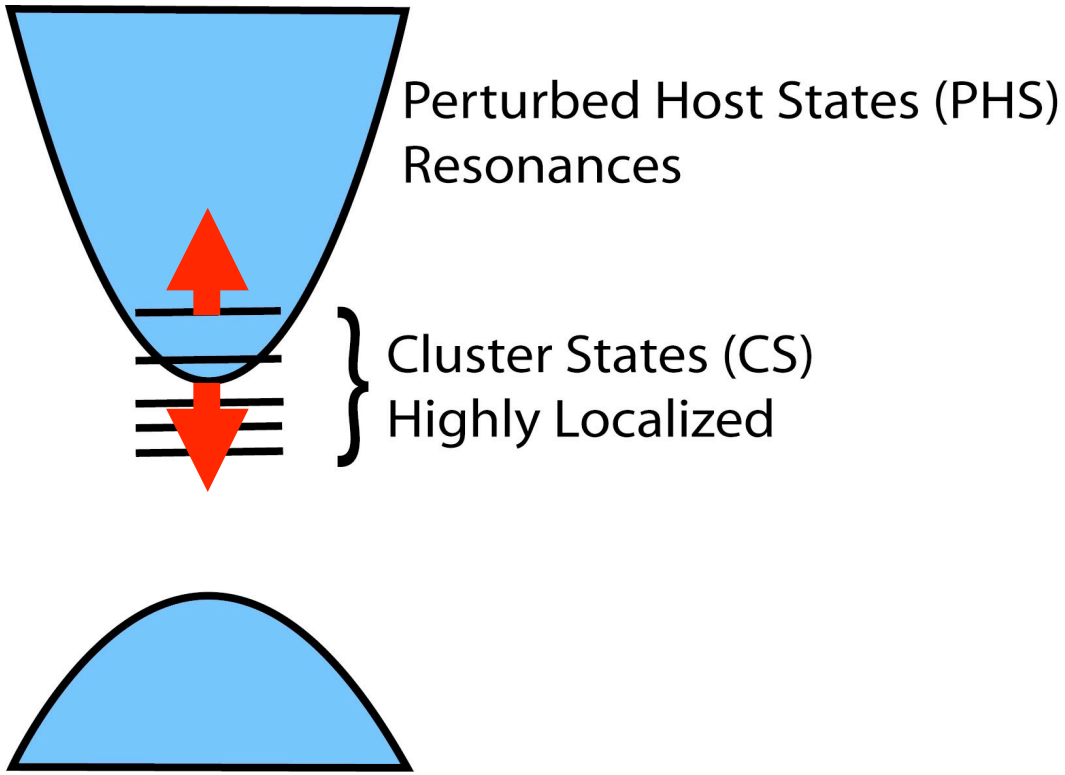
In dilute GaAsN

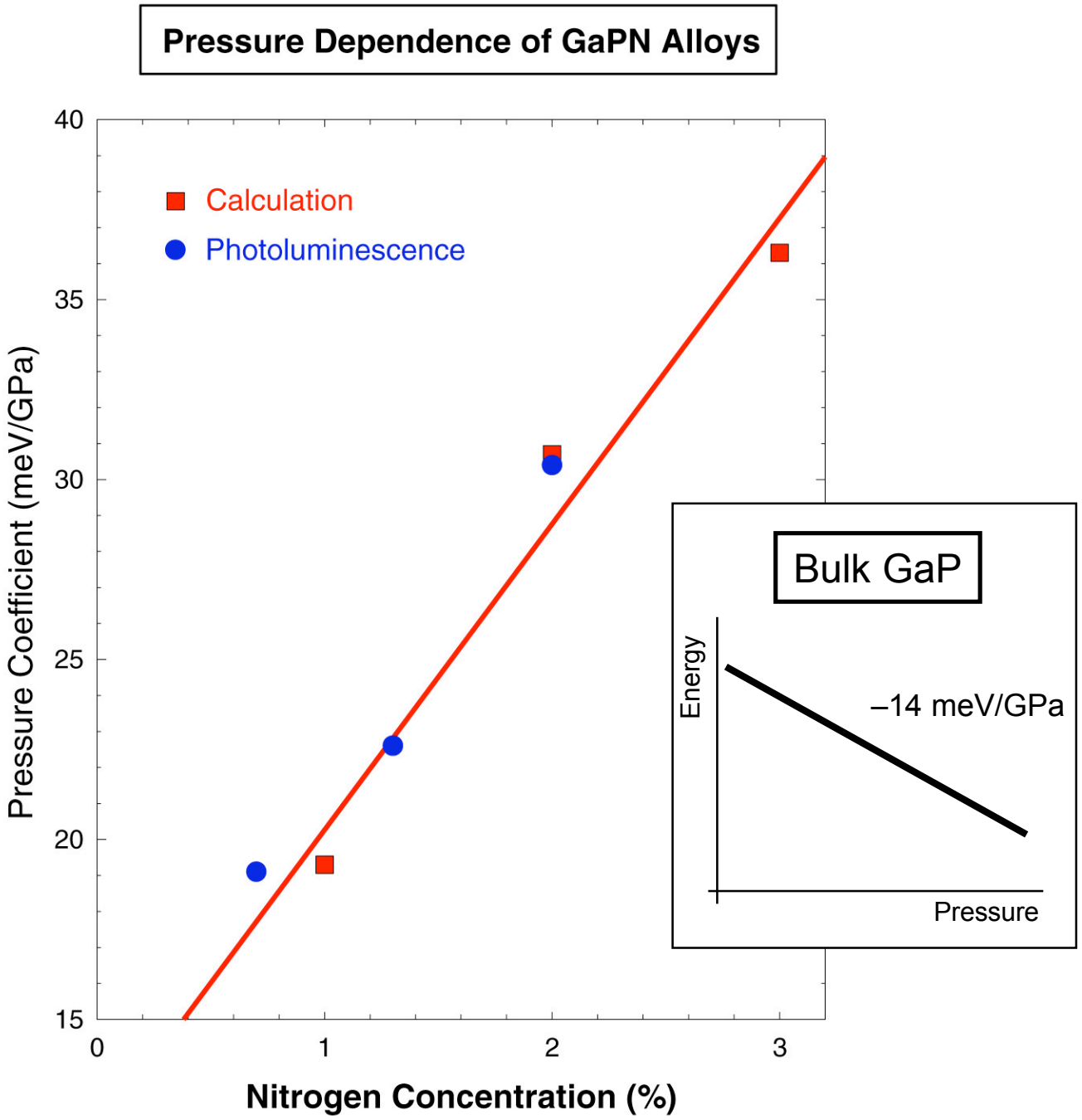
- Deep (low energy) CS emerge into gap with pressure
- Shallowest CS *can* hybridize
  - Pressure coefficient increased
  - Do not emerge into gap

Kent and Zunger Appl. Phys. Lett. **82** 370 (2003)

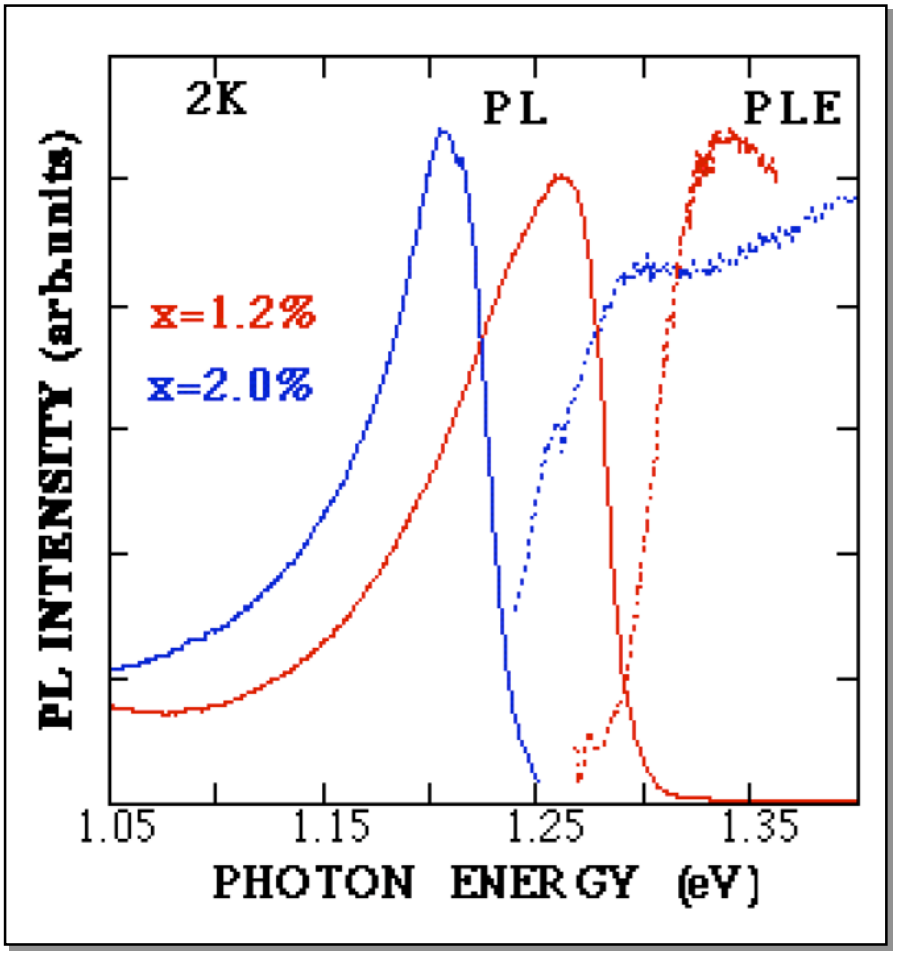
Band gap reduction

Anticrossing/repulsion between band edge and localized states drives band gap down

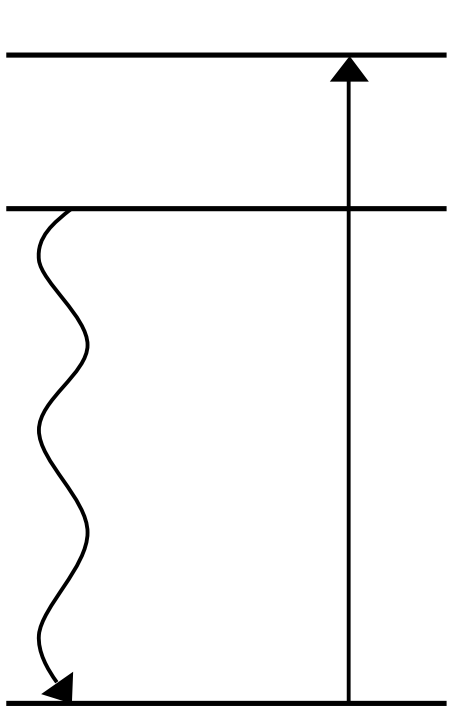



Pressure Dependence of GaPN Alloys
Calculation
Photoluminescence
Pressure Coefficient (meV/GPa)
Nitrogen Concentration (%)
40
35
30
25
20
15
0
1
2
3
Bulk GaP
Energy
–14 meV/GPa
Pressure

Red Shift of PL *vs* PLE



I. A. Buyanova *et al.* MRS IJNSR **6** 2 (2001)



- Emission from localized minority states

- Absorption to majority states