LEHIGH UNIVERSITY
Institute of Functional Materials and Devices (I-FMD)

Bulk and Thin-Film Single Crystal Growth of **Next-Generation Superhard, Electronic & Optical Materials**

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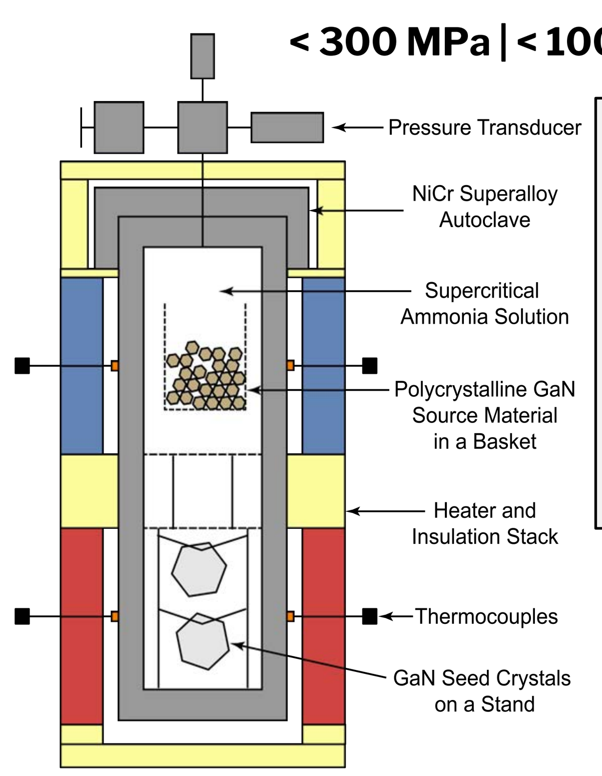
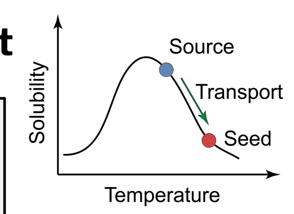
Solvothermal Methods

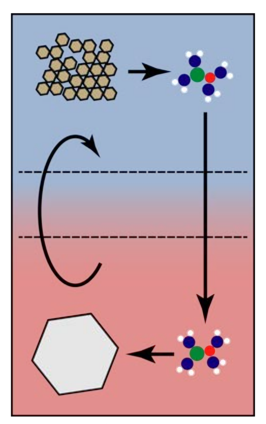
Ammonothermal Method

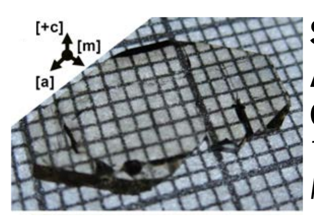
< 300 MPa | < 1000 °C | supercritical NH_3 solvent



Due to use of *ammonia* as a solvent, we can synthesize nitrogen-containing materials (*nitrides, carbonitrides, oxynitrides, etc.*) under relatively mild thermodynamic or kinetically-limited conditions to sizes < 1 inch.







Single Crystal Ammonothermal Gallium Nitride
$T_{melt} \approx 4000$ °C
$P_{melt} \approx 12$ GPa

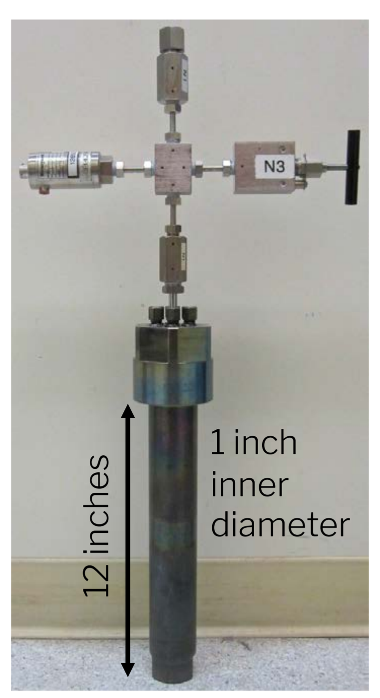
Suihkonen, S., Pimputkar, S., et al. Adv. Elec. Mat. 3 (2017) 1600496. 10.1002/aelm.201600496

MATS Facilities & Autoclaves

> 6 autoclaves in parallel | 2 independent setups | 1" inner diameter autoclaves

Our crystal growth facility is *unique in the world* due to our ability to pursue ammonothermal single crystal growth in *novel, extremely corrosion resistant Mo-alloy*, in traditional NiCr superalloy (right) or in SS autoclaves.

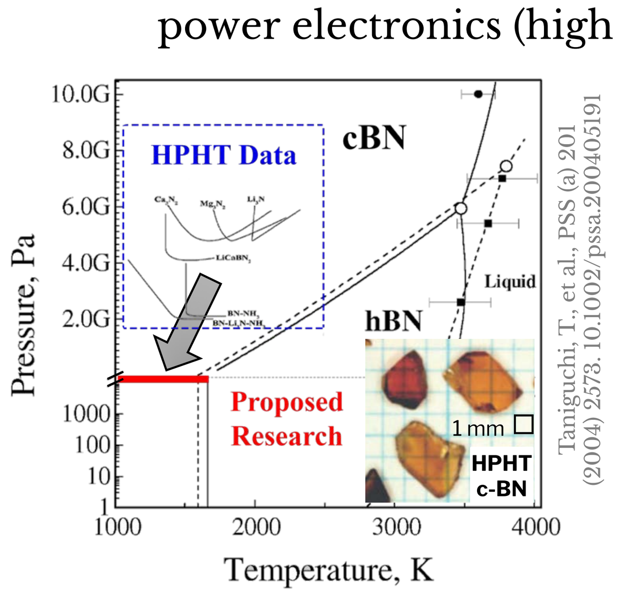
Operations require an oxygen-free environment and hence the use of *hermetically sealed, inert atmosphere, 1-inch thick steel-walled* containment vessels is mandated (left).



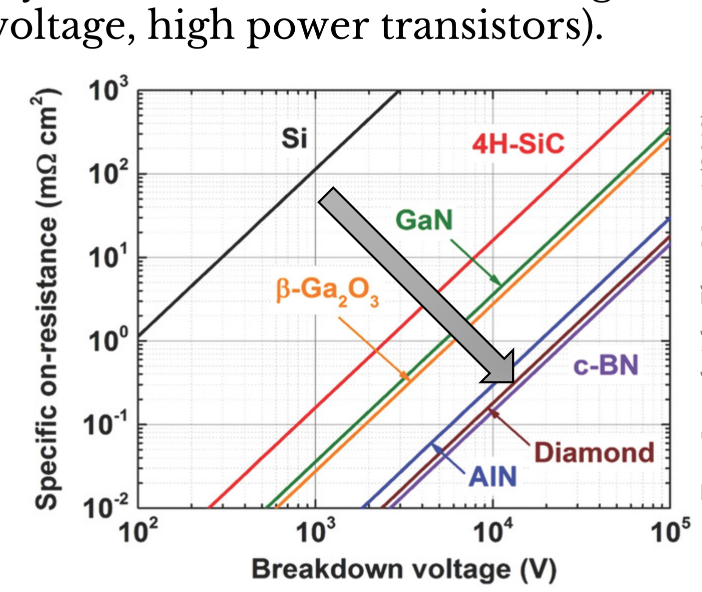
Materials: e.g. cubic BN

ultra-hard | ultrahigh–thermal conductivity | superior electronic properties

'Low' temperature and pressure synthesis will result in > 1-inch boules suitable for use in mechanical systems (abrasives, heat sinking) and power electronics (high voltage, high power transistors).



Taniguchi, T., et al., PSS (a) 201 (2004) 2573. 10.1002/pssa.200405191



Tsao, J., et al. Adv. Elec. Mat. 4 (2018) 1600501. 10.1002/aelm.201600501

Vision

novel equipment development | crystalline materials | nitrides & related materials | (ultra) wide bandgap semiconductor

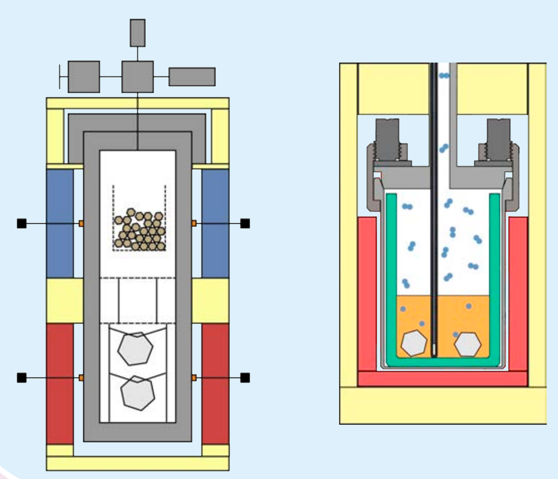
As a group, we develop new growth systems to produce crystalline materials which are currently challenging to synthesize or scale in size. We are particularly interested in pursuing materials which have *'extreme' properties* or are of particular value to enable *highly efficient energy conversion* (solar energy, electrical energy) or *optoelectronic device* of exceptional efficiency due to optimized pairing and tailoring of the substrate with the device layers.

We are *vertically integrated*, allowing us to develop next-gen devices on *tailored substrates* in *innovative growth systems*

Boule Growth

material properties | lattice constant

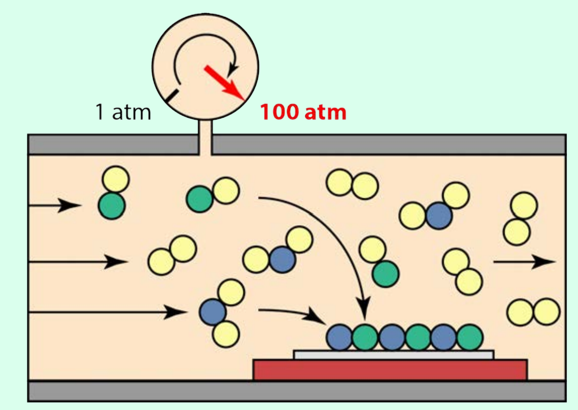
Grow large, *single* crystals for their desired *intrinsic* properties or to access novel substrates with *targeted* lattice constants.



Thin Films

alloy compositions | doping | phases

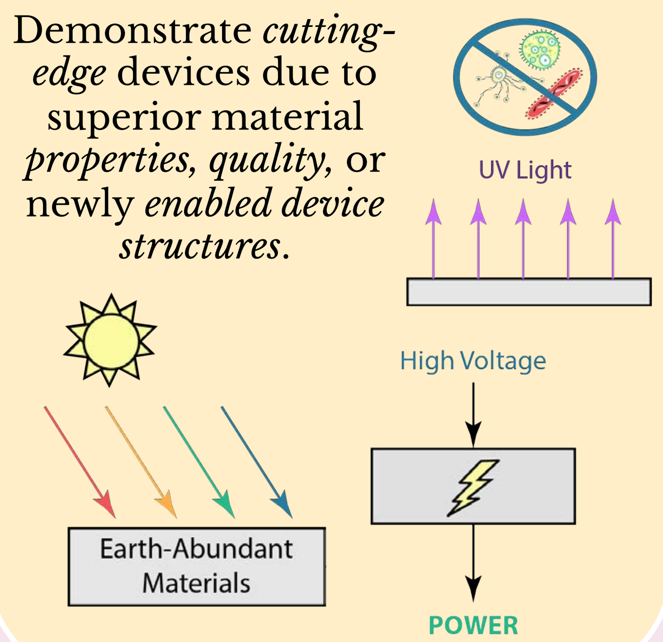
Use *in-house* produced (or external) substrates to demonstrate *new* materials with *superior properties* as thin films using novel precursors.



Devices

efficiency | novelty | performance

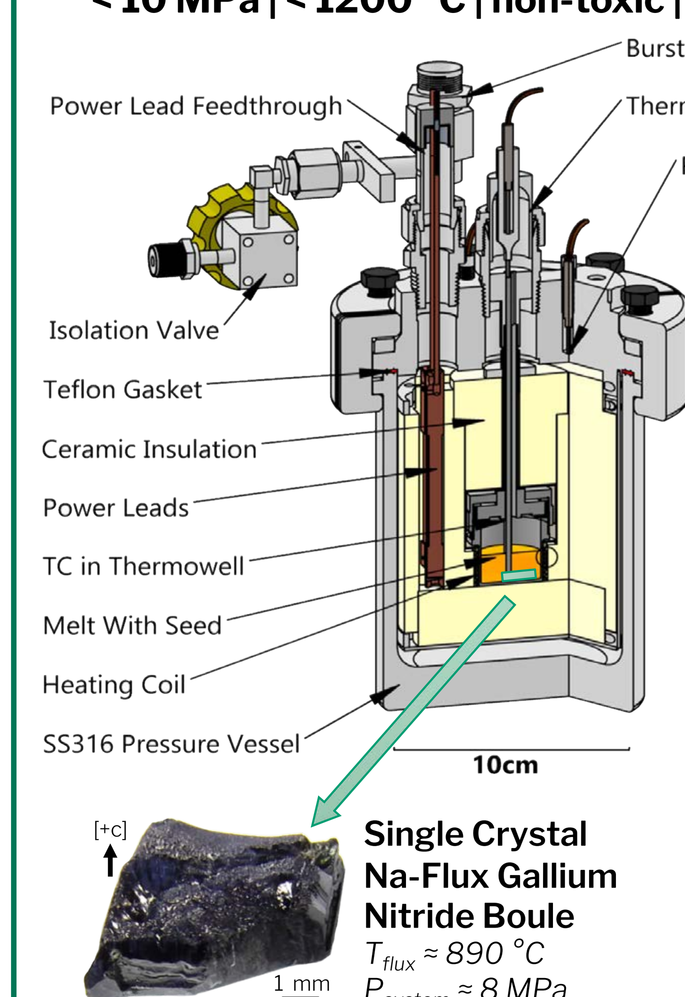
Demonstrate *cutting-edge* devices due to superior material *properties, quality*, or newly *enabled device structures*.



Flux Methods

Alkali metal Fluxes

< 10 MPa | < 1200 °C | non-toxic | high nitrogen solubility



Single Crystal Na-Flux Gallium Nitride Boule
$T_{flux} \approx 890$ °C
$P_{system} \approx 8$ MPa

Exposing *molten alkali metals* (e.g. Na) to high pressure gases, e.g. N_2, leads to *solubility of, e.g. N,* into the melt.

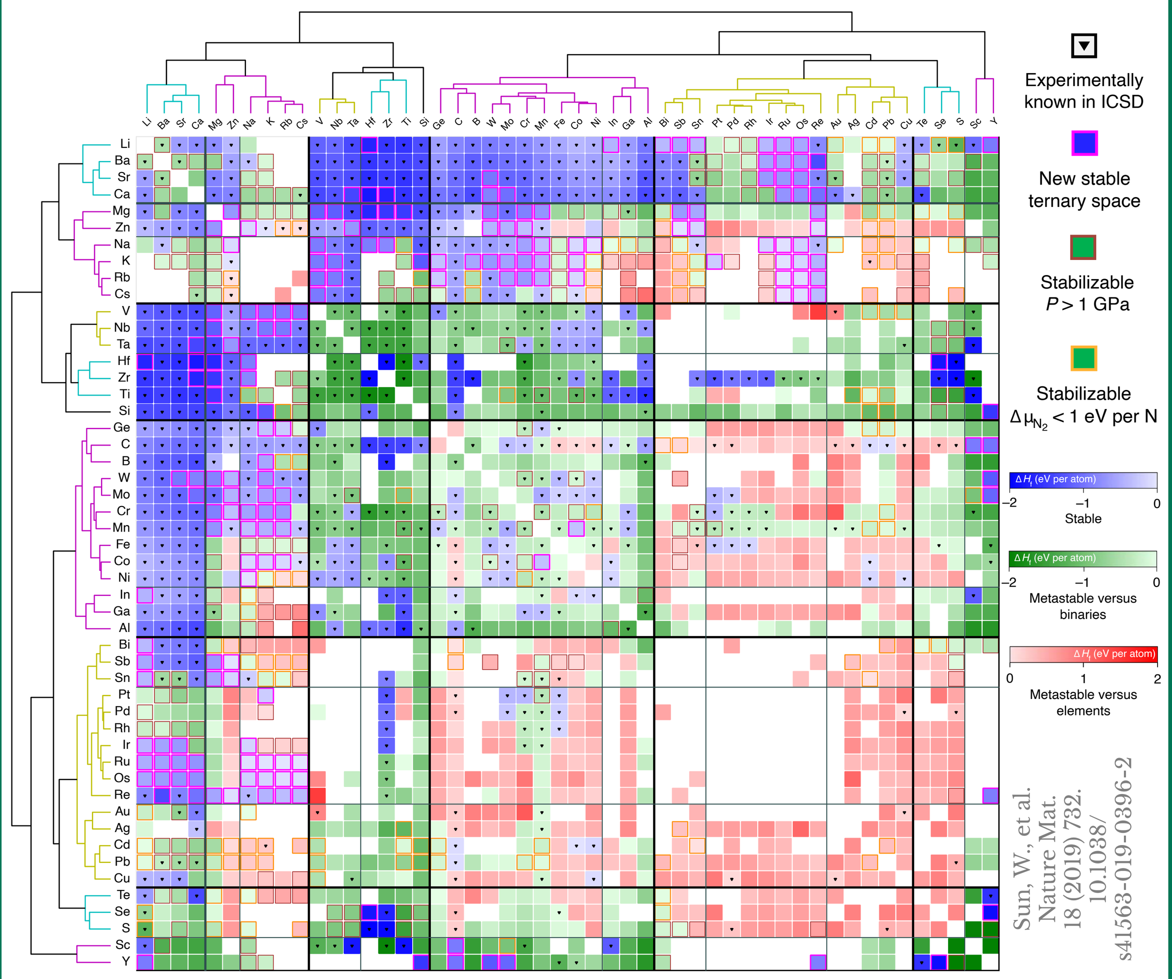
Desired crystals can be synthesized by tailoring the melt composition (e.g. addition of Ga) and driving growth via application of *gas pressures in excess of their equilibrium vapor pressures* or via *temperature gradients.*

Dollen, P., Pimputkar, S., et al. J. Cryst. Growth 456 (2016) 58. 10.1016/j.jcrysgro.2016.07.044;
Dollen, P., Pimputkar, S., et al. J. Cryst. Growth 456 (2016) 67. 10.1016/j.jcrysgro.2016.08.018

Materials: e.g. Ternary Nitrides

unexplored & new material systems | fundamental science

447 *stable* ternary nitrides predicted, only 213 demonstrated to-date. Requires *new approaches* to synthesize.



Sun, W., et al. Nature Mat. 18 (2019) 732. 10.1038/s41563-019-0396-2

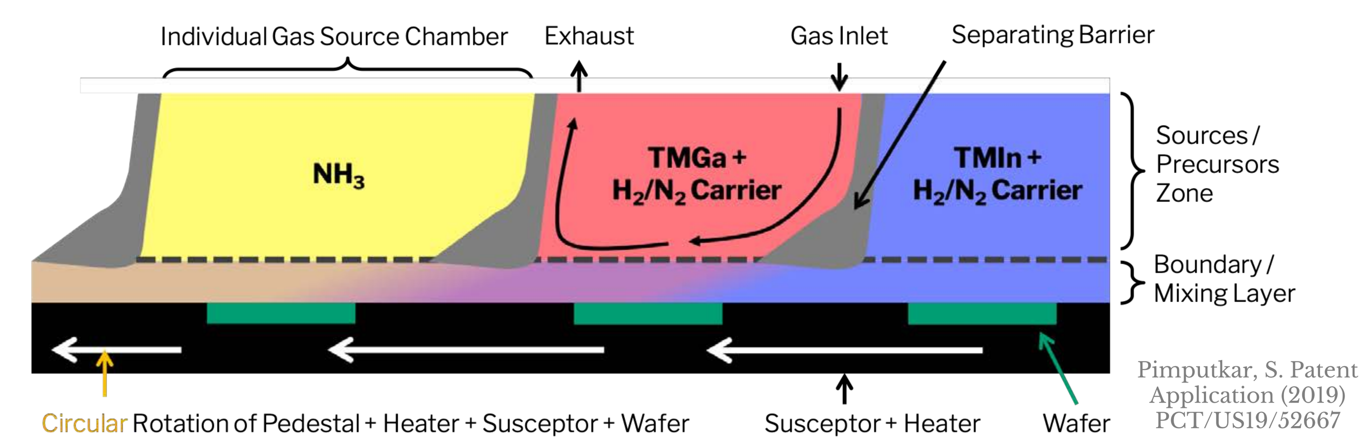
Chemical Vapor Deposition

HPS-CVD

< 100 atm | < 1300 °C | scalable design | grow decomposition-limited materials

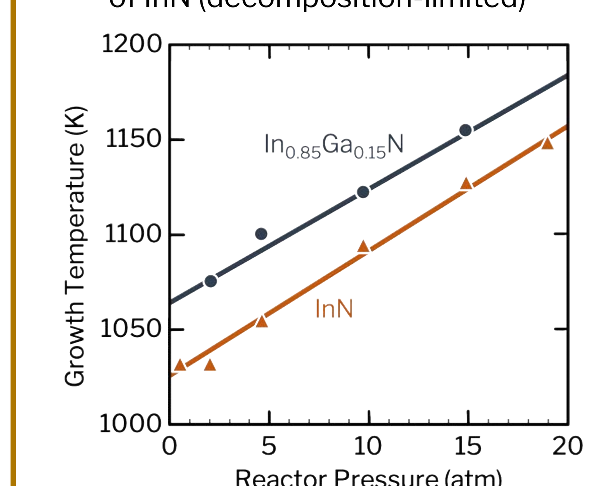
A novel *high pressure spatial chemical vapor deposition* (HPS-CVD) tool is being developed to synthesize thin films of decomposition-limited materials by growing at pressures up to 100 atm, for the first time.

Elevated pressures allow for *higher growth temperatures* (yielding high quality materials), *increased alloy constituent compositions* (via reduced decomposition), *increased impurity concentrations* (improved doping or desired element incorporation), and use of *pressure-stabilized precursors*.



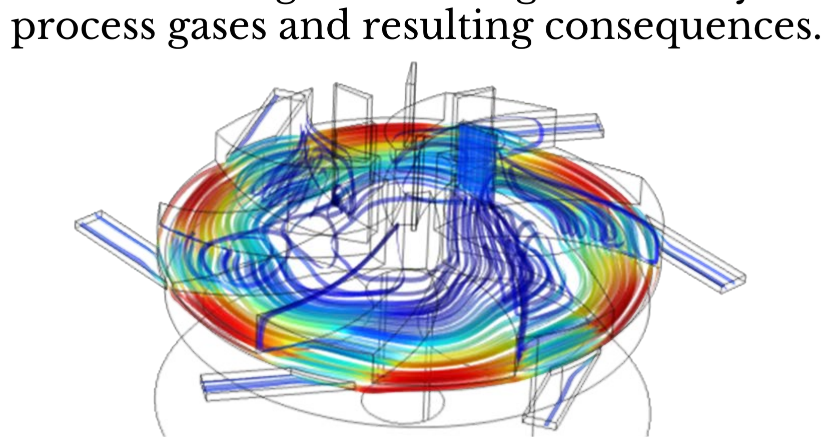
Pimputkar, S. Patent Application (2019) PCT/US19/52667

Proof-of-concept: HP-CVD growth of InN (decomposition-limited)



Dietz, N. Final Technical Report (2015). AFOSR FA9550-10-1-0097. 10.21236/ada624591

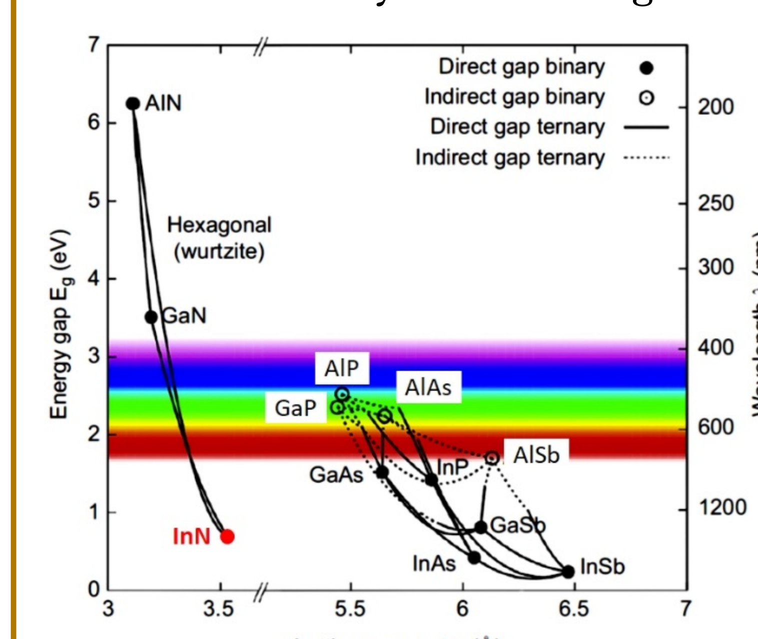
Designs are being optimized using *CFD/thermal computational methods* (COMSOL/ANSYS) to demonstrate ability to *overcome known challenges* due to higher density of process gases and resulting consequences.



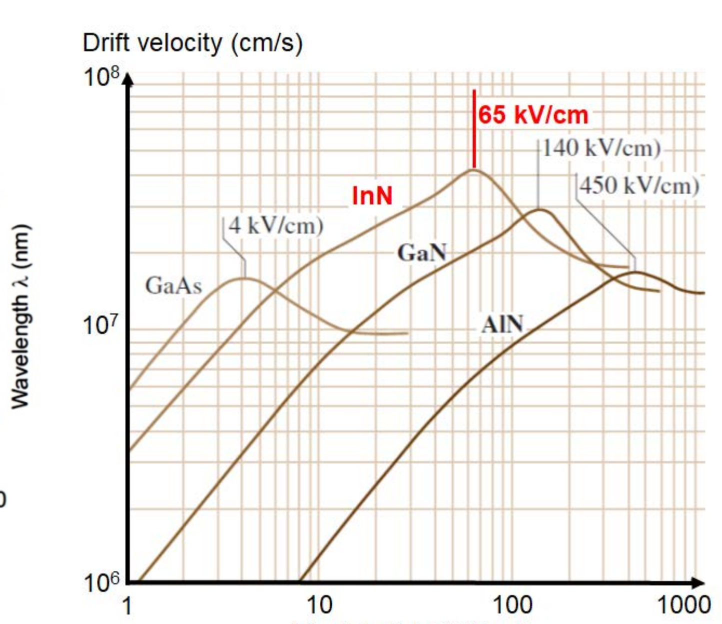
Materials: e.g. Group III-N

UV-IR emitters | high frequency/power electronics | UWBG semiconductors

High In-content III-N alloys can reveal the true potential of this semiconductor family via accessing the *entire solid solution region of AlInGaN.*



Modified from: Caro Bayo, M. Á., Ph.D. Thesis, University College Cork (2013), https://cora.ucc.ie/handle/10468/1344



Kasap, S. & Capper, P. 'Springer Handbook of Electronic and Photonic Materials', (2007). 10.1007/978-0-387-29185-7