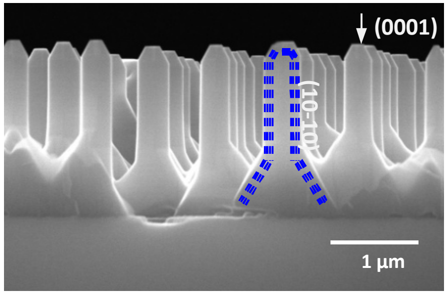
SSLS EFRC Research Challenges

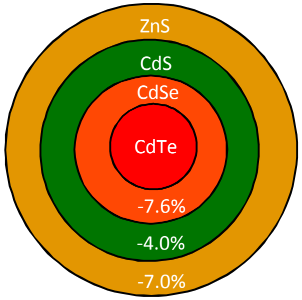
SAND2012-1777P

Materials Architectures

1: Nanowires (George Wang)



2: Quantum Dots & Phosphors (Jim Martin)



Light Emission Phenomena

3: Competing Rad & Non-Rad Processes (Mary Crawford)

Power-conversion efficiency · Joule efficiency · Injection efficiency · Internal quantum efficiency (ε_{IQE}) · Extraction efficiency

$$\varepsilon = \varepsilon_{Joule} \cdot \varepsilon_{inj} \cdot \frac{BN^2}{AN + BN^2 + CN^3 + \ldots} \cdot \varepsilon_{ext}$$

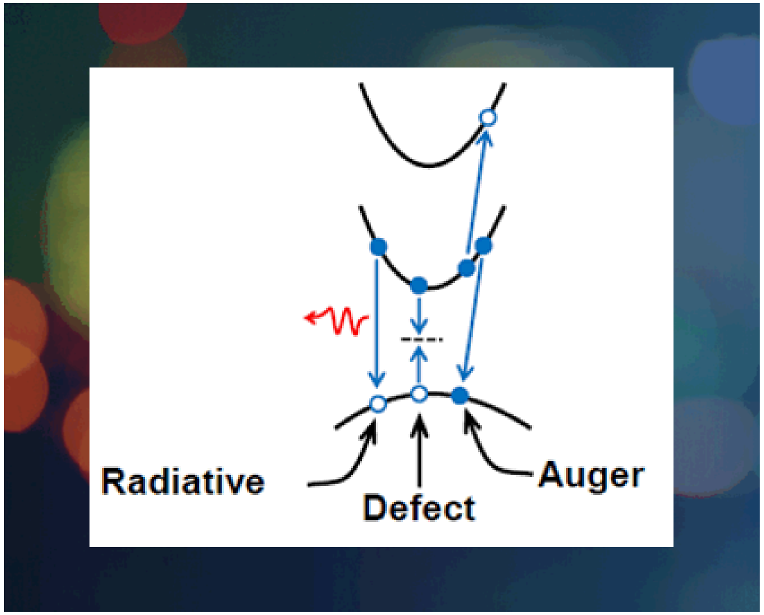
4: Defect-Carrier Interactions (Andy Armstrong)

5: Enhanced Spontaneous Emission (Igal Brener)

6: Beyond Spontaneous Emission (Art Fischer)

Research Challenge 3:
Competing Radiative and Non-Radiative Processes

Exploring the competing radiative and non-radiative recombination processes that limit the efficiency of light-emitting materials



Mary Crawford, **Weng Chow, and Daniel Koleske**
Sandia National Labs

E. Fred Schubert, David Meyaard, Guan-Bo Lin, Di Zhu, Qi Dai, and Jaehee Cho
Rensselaer Polytechnic Institute

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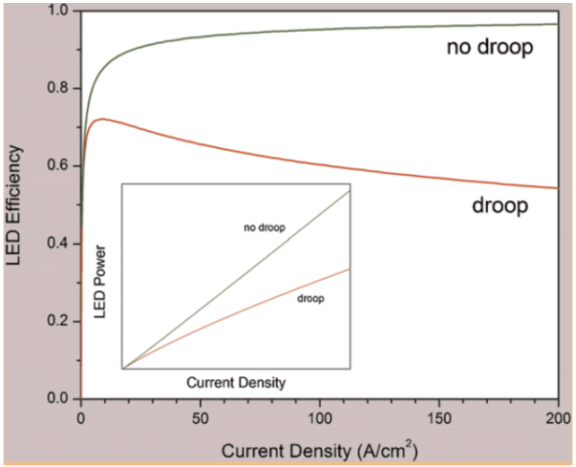
Motivation of this Research Challenge

High-level Goal: **Fundamental understanding of the *competition* between carrier recombination processes *that limit the efficiency of light-emitting materials***

Emphasis on → InGaN heterostructures

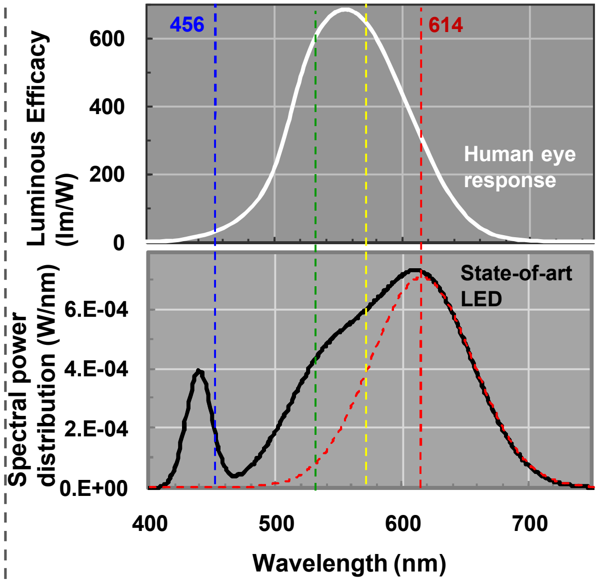
→ High carrier density regime

① ***Eliminate blue LED efficiency droop at high currents***

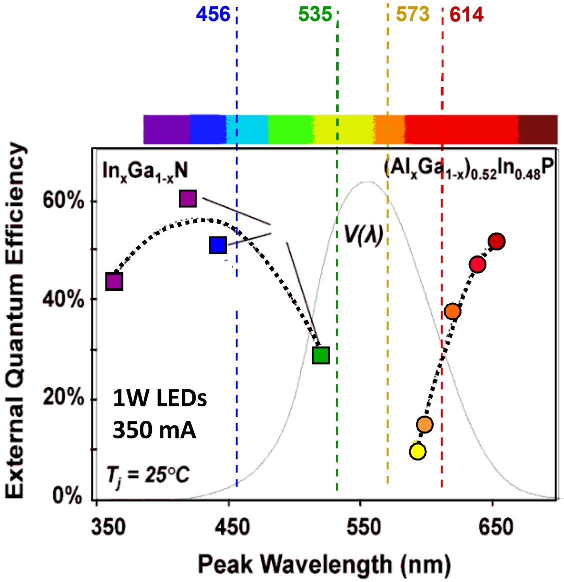


Piprek, PSSA, 2010

② ***Narrow-linewidth shallow-red emitter***



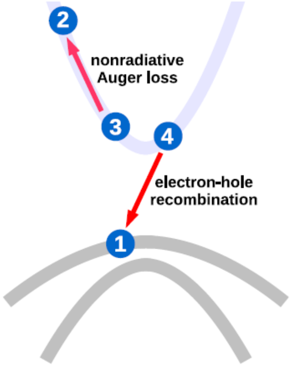
③ ***Fill in the green-yellow gap in LED efficiency***



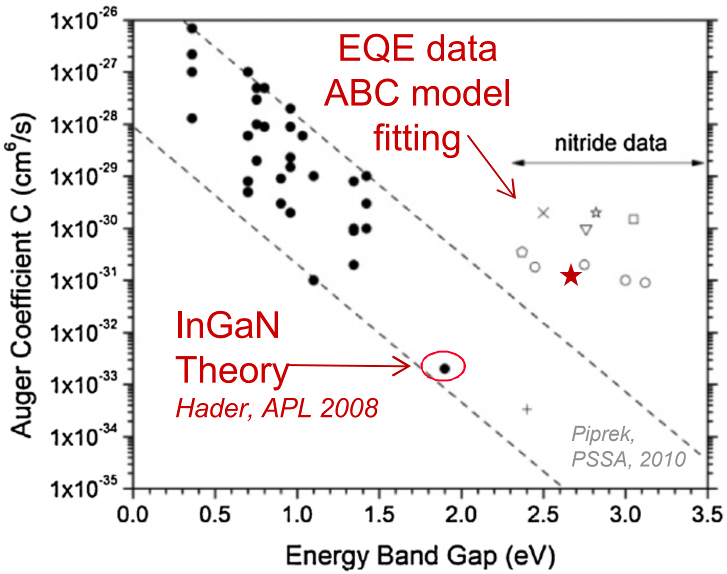
Courtesy of M. Krames, Philips-Lumileds

Proposed Mechanisms for Efficiency Droop

Direct Auger recombination



Pasenow, 2009

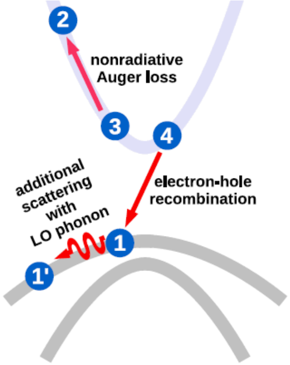


EQE data ABC model fitting

InGaN Theory

Hader, APL 2008

Phonon-assisted Auger recombination



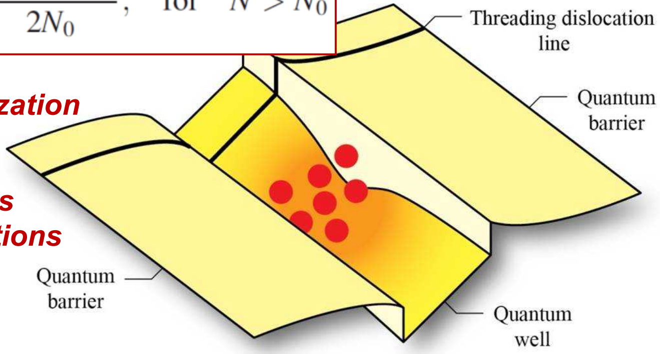
Van de Walle et al, APL 2010

Density-activated Defect Recombination

$$J_{DADR} = \begin{cases} 0, & \text{for} \quad N < N_0 \\ \dfrac{e n_w}{\tau_{DADR}} \dfrac{(N - N_0)^2}{2N_0}, & \text{for} \quad N > N_0 \end{cases}$$

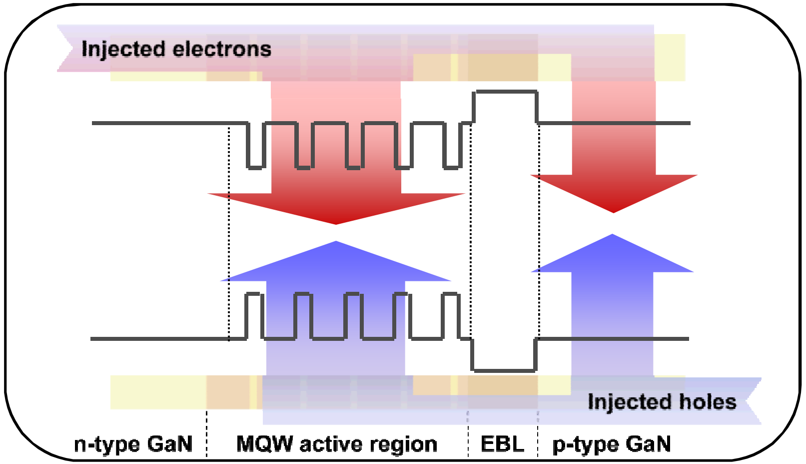
Carrier localization

Indium fluctuations
Well width fluctuations

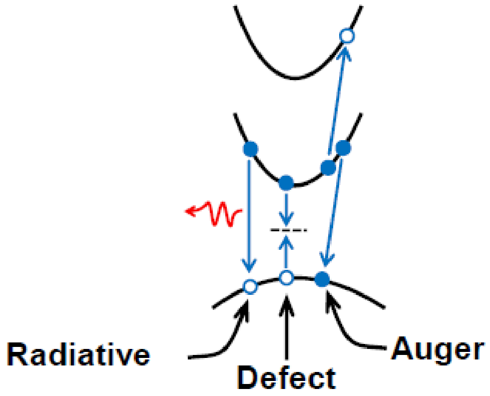


Hader et al, APL 2011

Carrier capture / leakage



EFRC Focus and Distinct Contributions



Power-conversion efficiency

Injection efficiency

Internal quantum efficiency (ε_{IQE})

$$\varepsilon = \varepsilon_{Joule} \cdot \varepsilon_{inj} \cdot \frac{BN^2}{AN + BN^2 + CN^3 + \ldots} \cdot \varepsilon_{ext}$$

Defects *Radiative* *Auger +…*

- Focus on *competing recombination processes,* not a single process (e.g., Auger)
- *Advancement beyond ABC model*: bandstructure, momentum-resolved carrier distributions, direct calculation of radiative recombination, non-equilibrium phenomena
- Extensive experimental and modeling studies on the *role of carrier transport*
- New studies on the critical impact of *defect recombination* on peak IQE and efficiency droop, considering distinct defect properties of InGaN materials
- Future work: exploration of *direct dynamical measurement of Auger processes*

Technical Presentations

I. Research Highlights

- **Development and application of a microscopic model to study efficiency limitations of InGaN LEDs (*Crawford*)**
- **Analysis of carrier transport contributions to efficiency droop (*Schubert*)**

II. Future Work *(Crawford)*

Limitations of the ABC Model

LED rate-equation model

$$\frac{dN}{dt} = \frac{J}{ed} - AN - BN^2 - CN^3$$

Defects

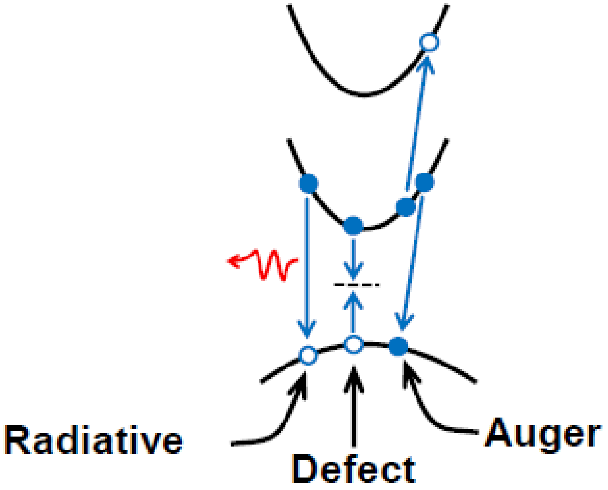
Spontaneous emission

Auger?

Internal quantum efficiency $= \dfrac{BN^2}{AN + BN^2 + CN^3}$

"ABC model"—no carrier injection

Radiative and Non-radiative Processes



Model Shortcomings:

a) True density dependence not simple N^m

b) Carrier injection/capture ignored

c) Non-equilibrium effects ignored

d) Large number of fitting parameters

Goal:

Develop improved model to gain insight into mechanisms that limit LED efficiency

Application of a Microscopic Model to Study Efficiency Limitations of InGaN LEDs

W. Chow, SNL

$$\varepsilon = \varepsilon_{Joule} \cdot \varepsilon_{inj} \cdot \frac{BN^2}{AN + BN^2 + CN^3 + \ldots} \cdot \varepsilon_{ext}$$

Key Advances:

→Replace total "N" with momentum–resolved distributions of electrons and holes (QW & barrier)

→ *implement bandstructure directly into model*

→Add carrier-carrier and carrier–phonon interactions

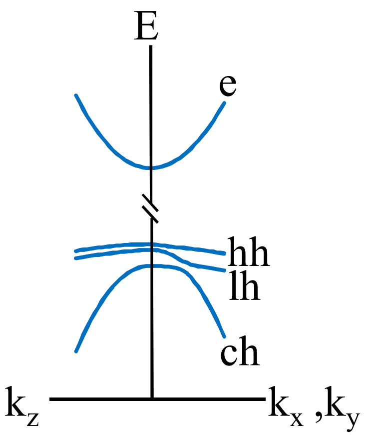
→ *enables more accurate carrier distribution vs. current and temperature*

→Calculate radiative contribution directly from bandstructure & carrier distributions

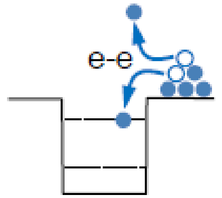
→ *B(n,T), avoids constant B parameter*

$$N = \sum_{k} n_{e(h),k}$$

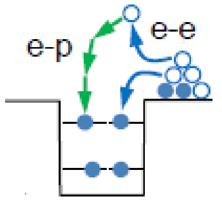
Bandstructure



Carrier-carrier (100fs)



Carrier-phonon (1ps)



Chow et al, Appl. Phys. Lett. 97, 121105 (2010)

"ABC model"

$$\frac{dN}{dt} = \frac{J}{ed} - AN - BN^2 - CN^3$$

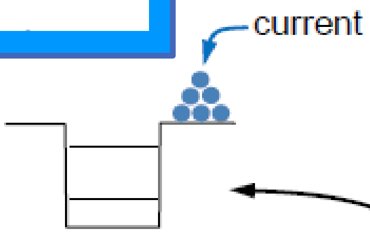
Defects

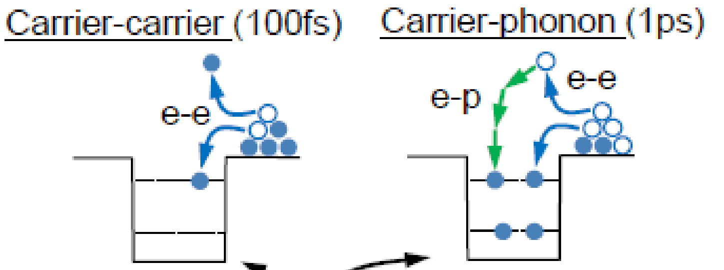
Spontaneous emission

Auger?

Summary of new model and equations

New Model





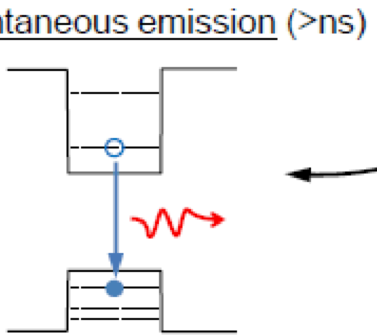
e or *h*

Barrier:

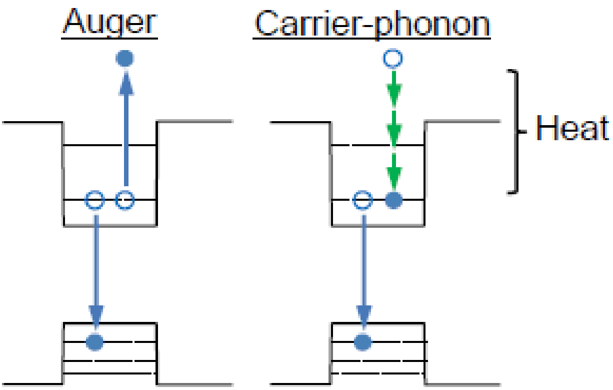
$$\frac{dn^b_{\sigma,k}}{dt} = \overbrace{\frac{J(t)}{eN^p_\alpha} f\left(\varepsilon^b_{\sigma k}, \mu^p_\alpha, T_p\right)\left(1 - n^b_{\alpha,k}\right)} - A_b n^b_{\sigma,k} + \left.\frac{\partial n^b_{\sigma,k}(t)}{\partial t}\right|_{col}$$

QW:

$$\frac{dn_{\sigma,\alpha_\sigma,k_\perp}}{dt} = \underbrace{-n_{\sigma,\alpha_\sigma,k_\perp} \sum_{\alpha_{\sigma'}} \frac{\wp^2_{\alpha\beta k}\Omega^3_k}{\pi\varepsilon\hbar c^3} n_{\sigma',\alpha_{\sigma'},k_\perp}} - A n_{\sigma,\alpha_\sigma,k_\perp} + \left.\frac{\partial n_{\sigma,\alpha_\sigma,k_\perp}}{\partial t}\right|_{col}$$

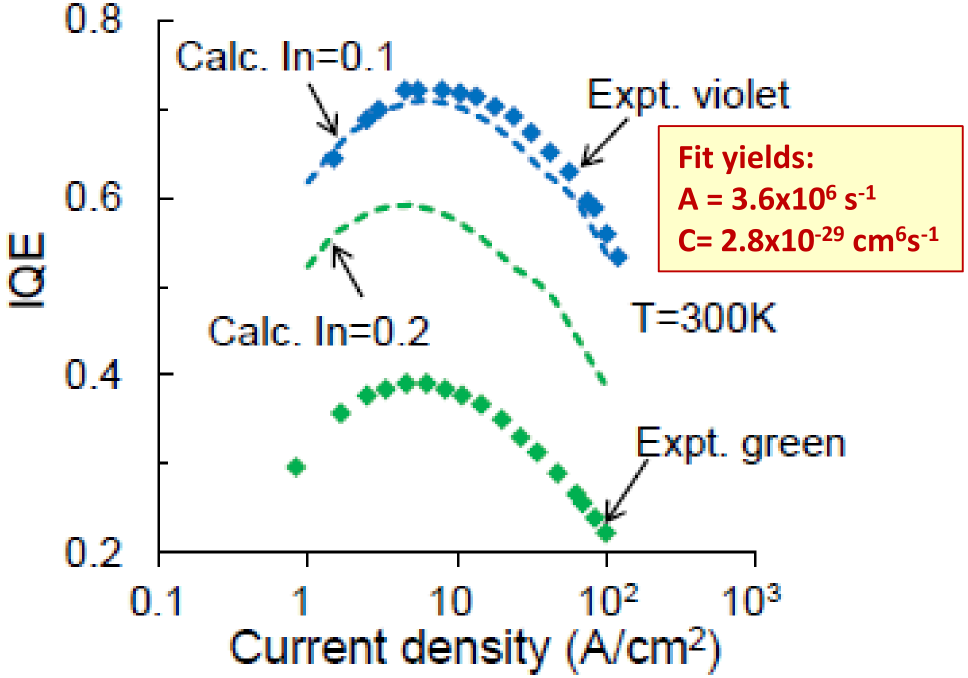


$\tau \sim 1/CN^2$



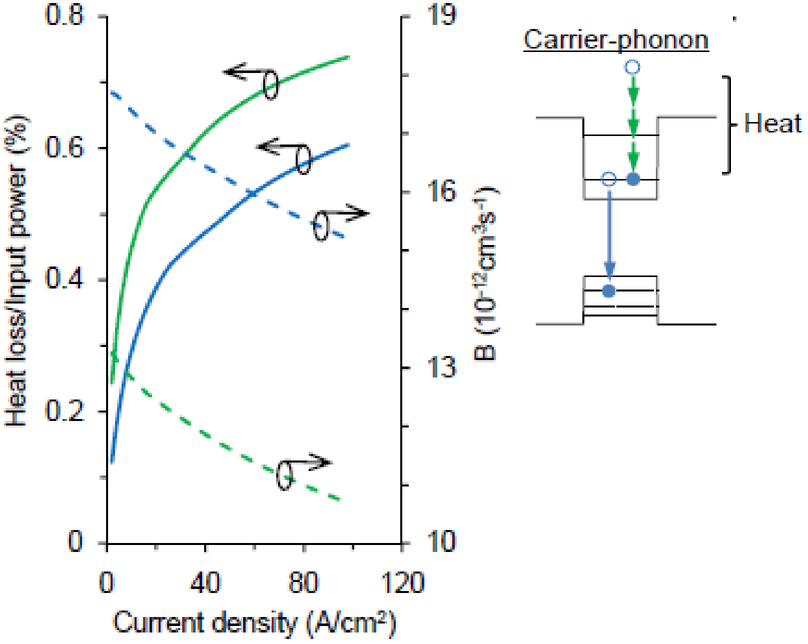
Contributions to LED Efficiency Dependence on Wavelength

IQE of Commercial Violet and Green InGaN LEDs



Fit yields:
A = 3.6x10^{6} s^{-1}
C= 2.8x10^{-29} cm^{6}s^{-1}

Contributions to lower green LED efficiency



$$B_{eff} = N^{-2} \sum_{k_\perp} \left|\wp_{k_\perp}\right|^2 \Omega_{k_\perp}^3 (\pi\varepsilon\hbar c^3)^{-1} n_{e,k_\perp} n_{h,k_\perp}$$

Model insights:

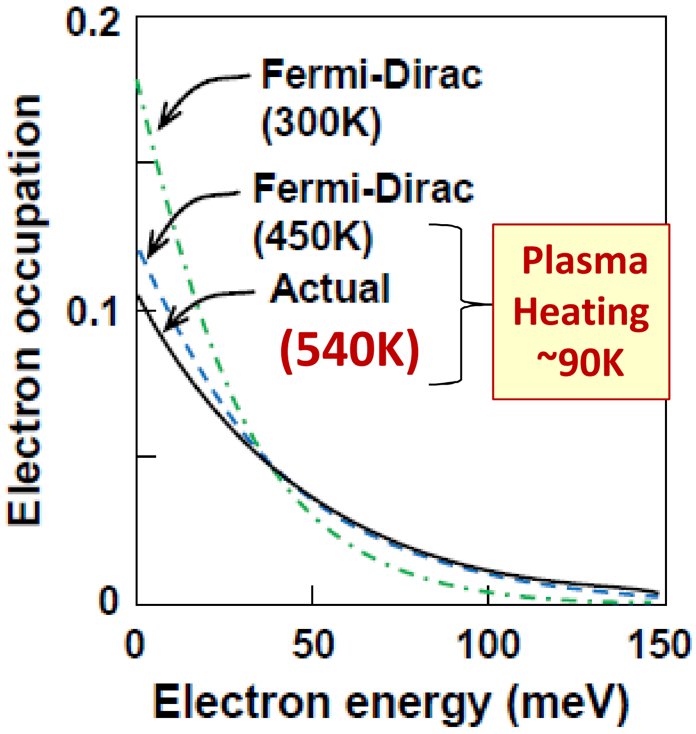
→ ~1/3 of difference between violet & green LED efficiency due to intrinsic contributions

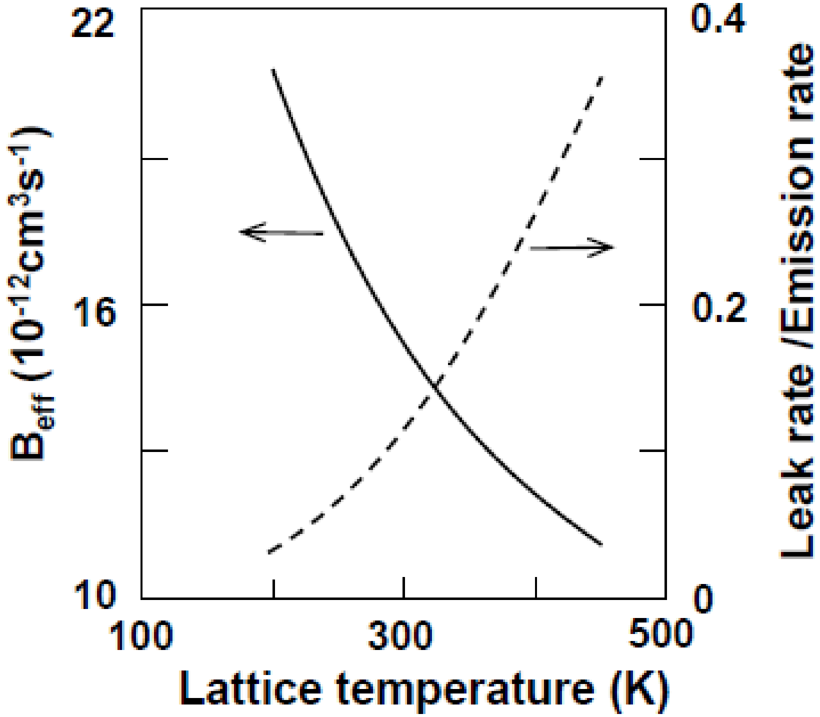
→ enhanced heat loss (phonons), reduced effective B coefficient, for green LED

Expt:Krames et al, J. Display Technology 3, 1551 (2007)

Contributions to LED Efficiency Dependence on Temperature

Assumptions: In $_{0.1}$Ga$_{0.9}$N QW LED (violet) J = 100 A/cm^2 T= 450K





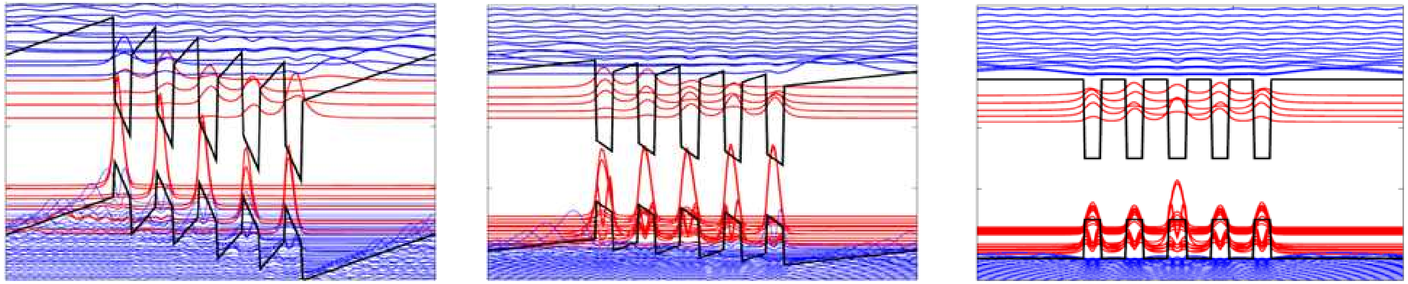
- Model quantifies plasma heating at high current densities (100 A/cm^2)
- Identifies contributions to lower efficiency at elevated temperatures:

→ carrier leakage out of QWs, reduced B_{eff}

Additional Modeling Efforts and Intra-EFRC Synergies

Excitation-dependent Bandstructure Influences on IQE and Efficiency droop

$N \sim 2 \times 10^{13}$ cm^{-2} → *$N \sim 7 \times 10^{13}$ cm^{-2}* → *Flat-band limit*



Poster Weng Chow

Research Challenge 4: Multi-level Defect Contributions to Efficiency Droop

LED rate-equation model

A(n)

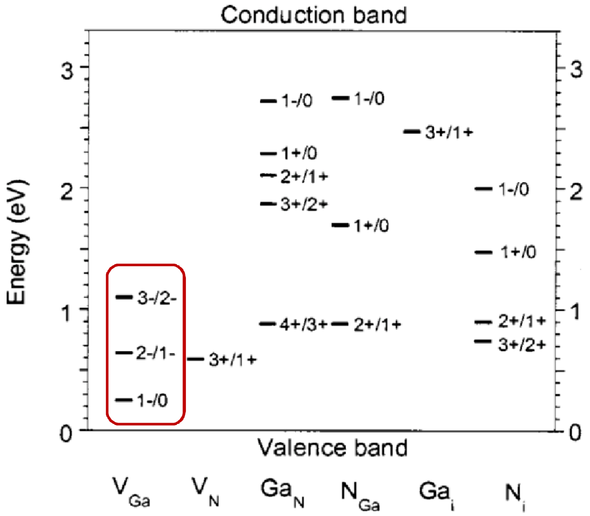
$$\frac{dN}{dt} = \frac{J}{ed} - AN - BN^2 - CN^3$$

Defects

Spontaneous emission

Auger?

Internal quantum efficiency $= \dfrac{BN^2}{AN + BN^2 + CN^3}$

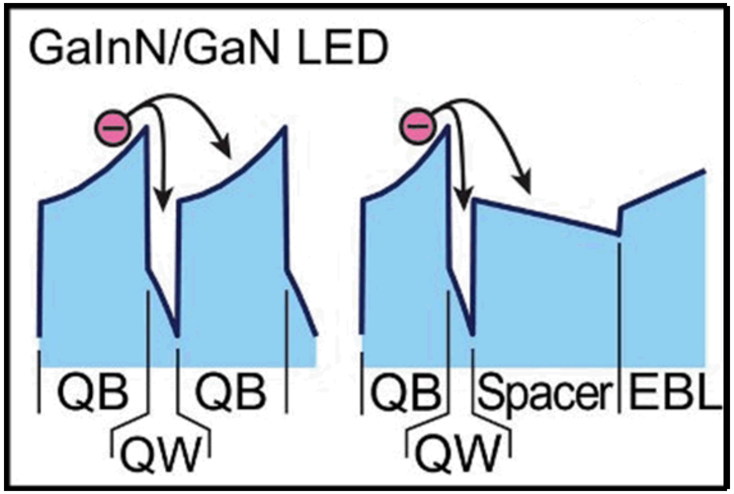


Presentation & Poster Normand Modine Andrew Armstrong

Limpijumnong and Van de Walle, PRB 2004.

Research Challenge 3:
Competing Radiative and Non-Radiative Processes

Analysis of Carrier Transport Contributions to Efficiency Droop of InGaN LEDs



E. Fred Schubert, **David Meyaard, Guan-Bo Lin,**
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Outline

Current research:

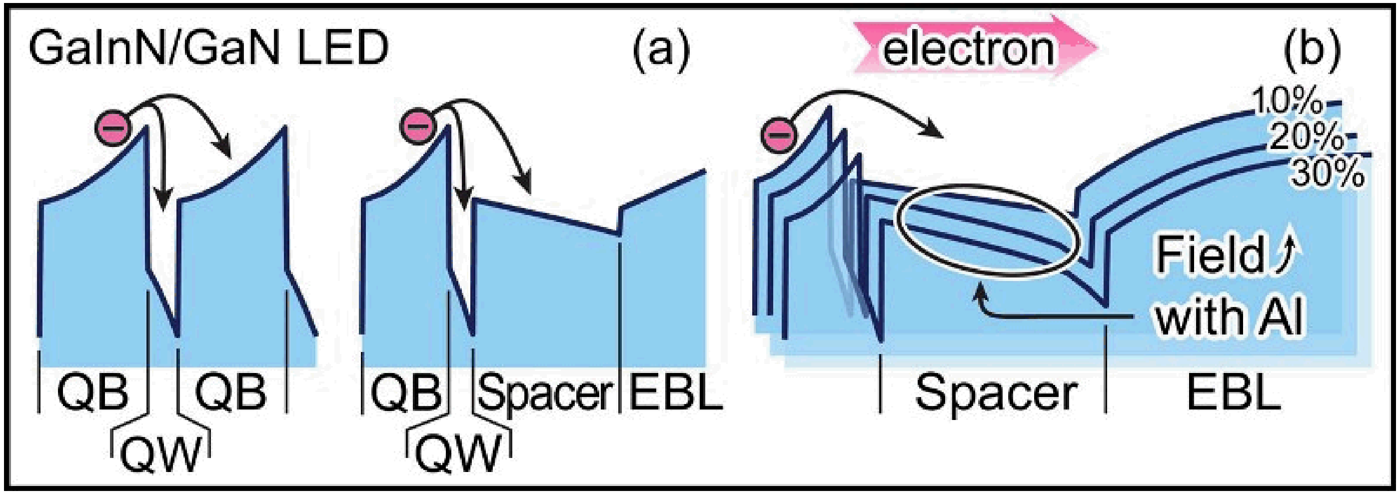
- Transport in the active region of MQW structure
- Confirmation of experimental results by Genetic Algorithm

Current and future research:

- Analytic model for efficiency droop
- Implementation of parameters suggested by model

Transport of carriers in the MQW active region

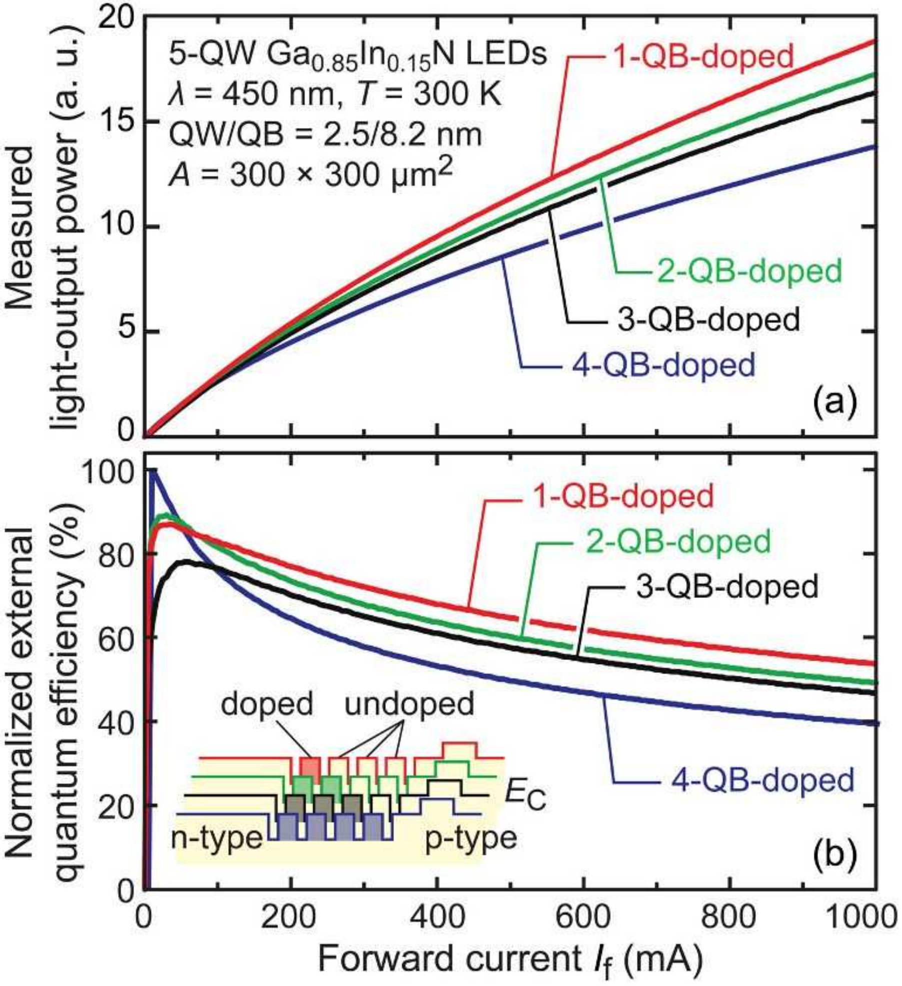
- **Known problem with GaInN LEDs**
  - Injection efficiency has been estimated in the range of 50% – 95%
  - Carrier capture hindered by small QW widths
  - Carrier escape possible due to polarization fields



- **Doping of the quantum barriers (QBs)**
  - ➔ Control of distribution of carriers within MQW

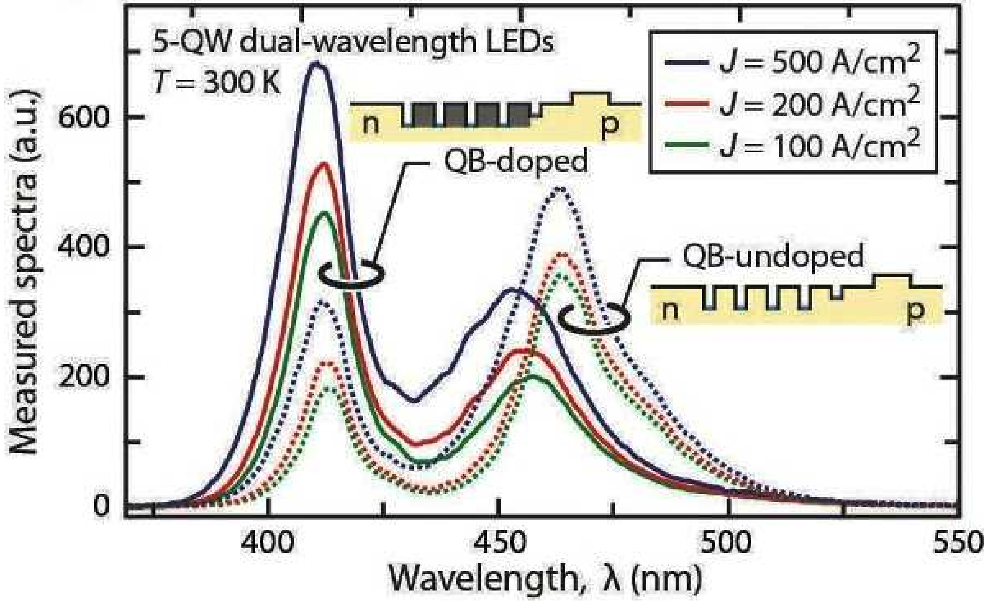
Series of GaInN LED samples and results

- Light output is strong function of QB doping
- Irrespective of the mechanism causing the efficiency droop
- Lower carrier concentration in the QWs are beneficial
  - ***Leakage***: The fewer carriers are in the last-grown QW, the less leakage
  - ***Delocalization, Auger***: The more uniform the carrier distribution, the lower the droop



Series of GaInN LED samples and results

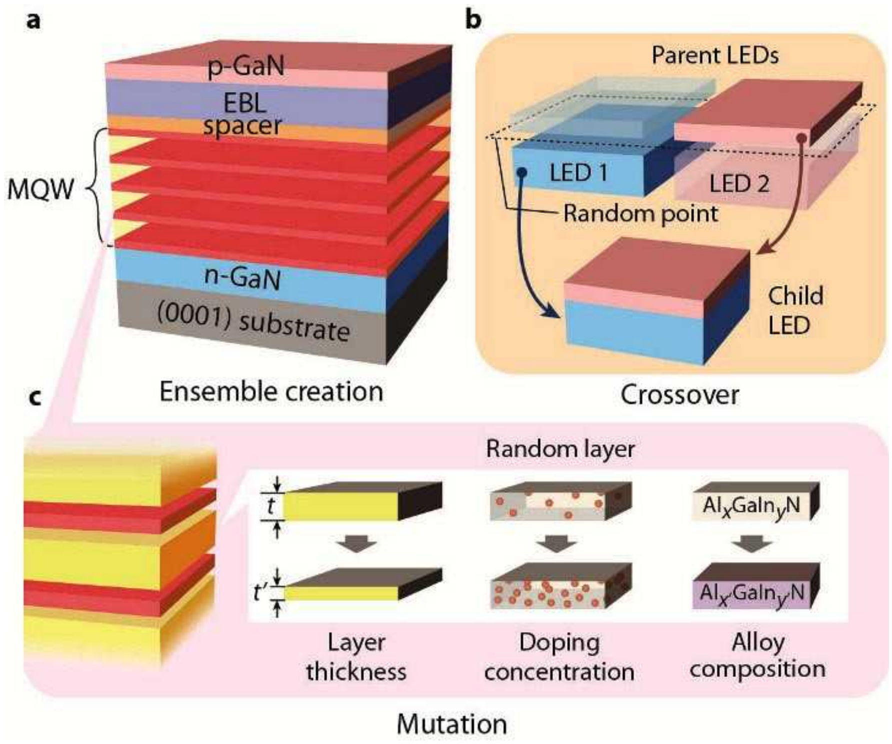
- Verification of carrier distribution by employment of blue and violet MQW structure



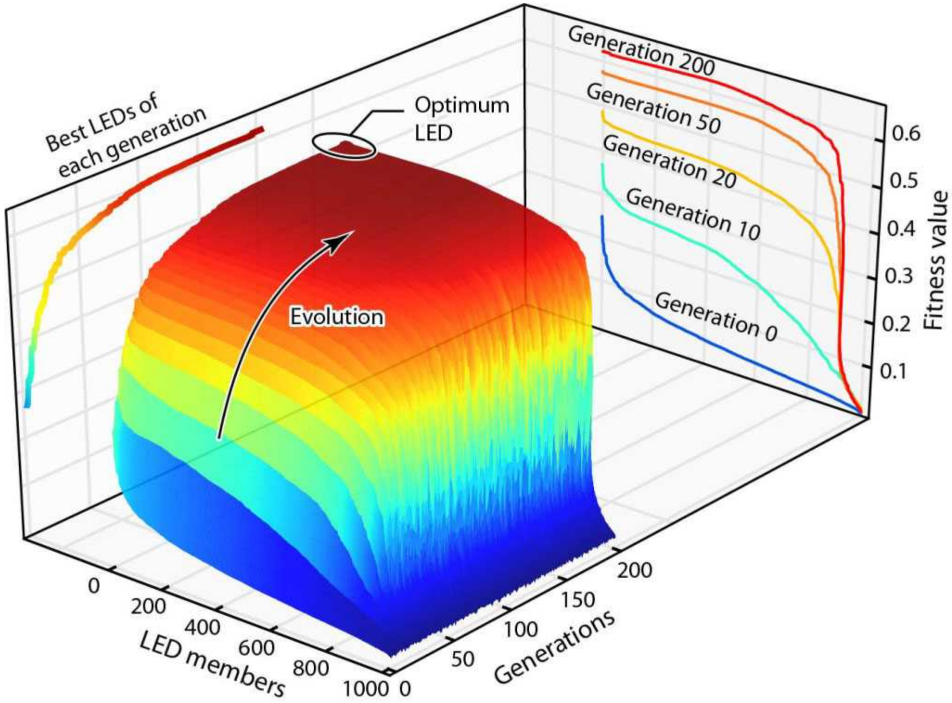
- Violet-to-blue ratio is a measure of the carrier distribution among the QWs

Genetic Algorithm for LEDs

- Crossover
- Mutation

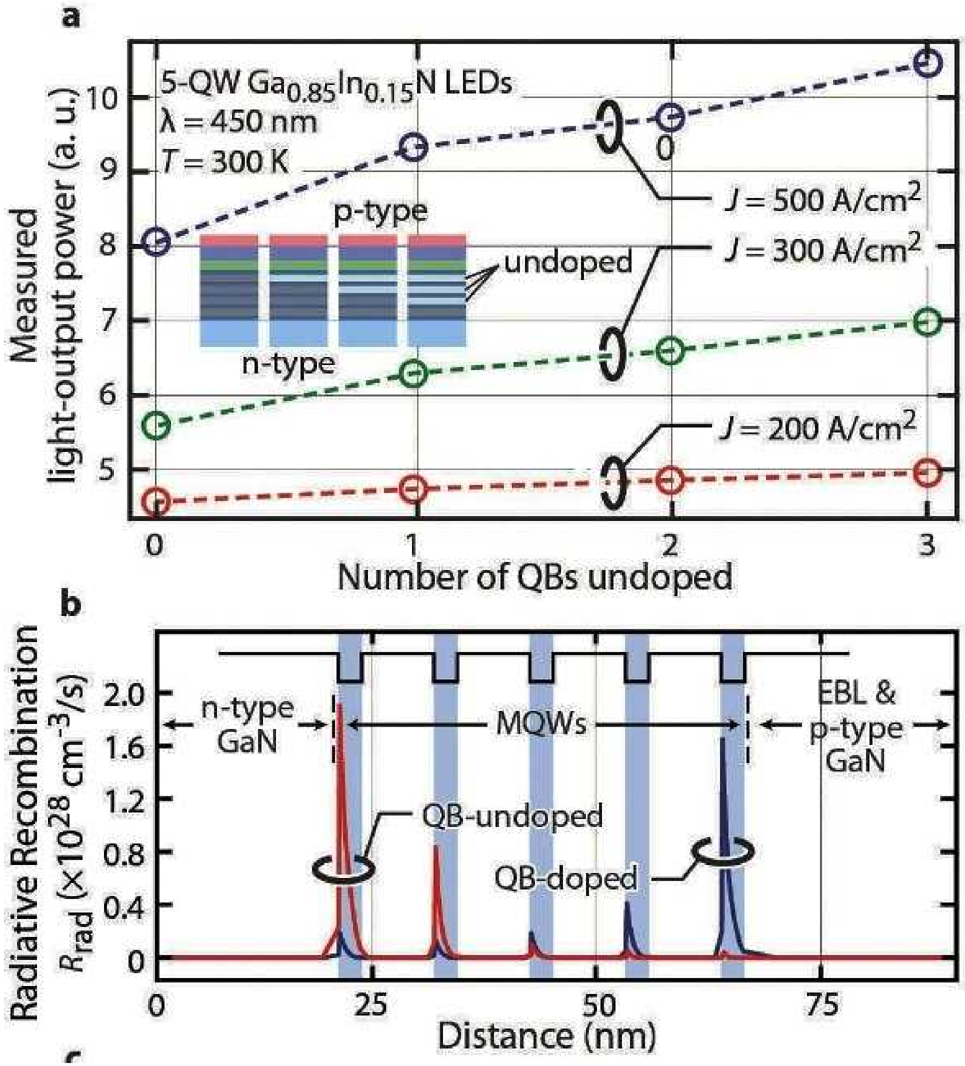


- Evolution of fitness



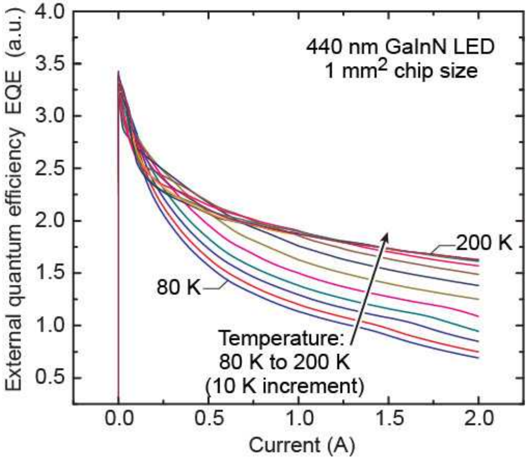
Genetic Algorithm

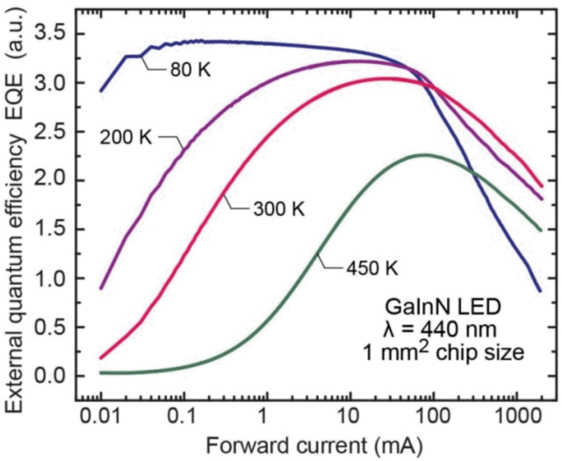
- Genetic Algorithm approach
  - Free parameter: Doping of QBs
- Result
  - Doping in QBs affects
    - Carrier distribution
    - Light-output power at high currents



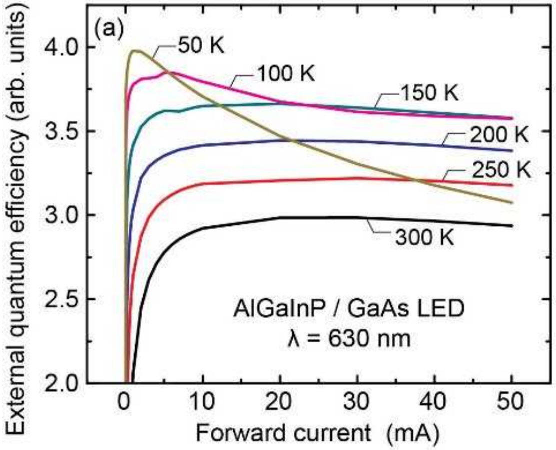
Droop is general problem in LEDs: GaInN and AlGaInP

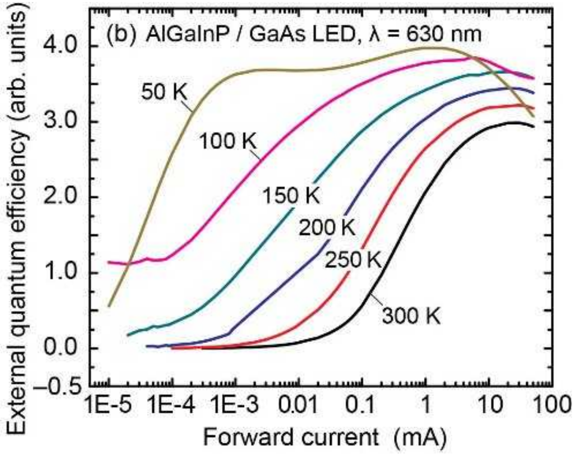
InGaN LED:





AlGaInP LED:





Results are remarkable: Different from InGaN, AlGaInP has:

- No indium composition fluctuations
- No dislocations
- No polarization fields
- No narrow active region (38 QWs ➔ low carrier concentration)

➔ Droop in LEDs is a more general problem than initially thought

Analytic Model for Efficiency Droop

- Conventional ***high-injection condition*** (Shockley, 1950):

$$\Delta n_p(0) << p_{p0} \qquad \text{Onset of high injection: } \Delta n_p(0) = 0.1 \times p_{p0}$$

- In the presence of a large mobility difference, the high-injection condition needs to be generalized

$$\Delta n_p(0) \quad << \quad p_{p0}\ (\mu_p / \mu_n)$$

- Electron leakage in low-injection regime

$$J_{\text{Diffiusion}} = \frac{e\, D_n\, \Delta n_p(0)}{L_{\text{p-GaN}}}$$

- Electron leakage driven by electric field in high-injection regime

$$J_{\text{Drift}} = e\, \mu_n\, \Delta n_p(0)\, E = e\, D_n \frac{e}{kT} \Delta n_p(0) \frac{J_{\text{Total}}}{\sigma_p}$$

- Droop is significant when $J_{\text{Drift}} = J_{\text{Diffusion}}$

$$J_{\text{Total}}\Big|_{\text{Droop}} = \frac{\sigma_p}{L_{\text{p-GaN}}} \frac{kT}{e} = \frac{p_{p0}\ \mu_p}{L_{\text{p-GaN}}} kT$$

The droop *C* and *D* coefficient

- Condition for Onset of droop:

$$J_{\text{Onset-of-droop}} = 0.1 \times J_{\text{Total}}\Big|_{\text{Droop}} = 0.1 \times \frac{p_{\text{p0}}\,\mu_{\text{p}}}{L_{\text{p-GaN}}}\,kT$$

  - Consistent with *T* dependence of onset of droop in GaInN and AlGaInP LEDs

- Recombination can be described by
  - $R = An + Bn^2 + Cn^3 + f(n)$

- Analysis shows
  - $f(n) \propto n_{\text{QW}}^3$ near efficiency peak
  - $f(n) \propto n_{\text{QW}}^4$ beyond efficiency peak where droop is significant

$$C = \frac{\alpha\,\mu_{\text{n}}}{p_{\text{p0}}\,\mu_{\text{p}}} \times B \qquad D = \left(\frac{\alpha\,\mu_{\text{n}}}{p_{\text{p0}}\,\mu_{\text{p}}}\right)^2 \times B \qquad \text{where } \alpha = \Delta n_{\text{p}}(0)\,/\,n_{\text{QW}}$$

Note: $C \propto p_{\text{p0}}^{-1}$ and $D \propto p_{\text{p0}}^{-2}$

P-type doping

- Improvement of p-type doping (p_{p0}) from 5 x 10^{17} to 2 x 10^{18} cm^{-3} …
  - … would reduce *C* by factor of 4
  - … would reduce *D* by factor of 16

- Needed: Improvement of p-type doping
  - InGaN layers
    - Preliminary results show excellent improvement in p-type doping properties
  - ZnO injection layers
    - Preliminary results show excellent performance of green LEDs

Conclusions

We demonstrated:

- Transport in the active region affects the efficiency droop

We developed:

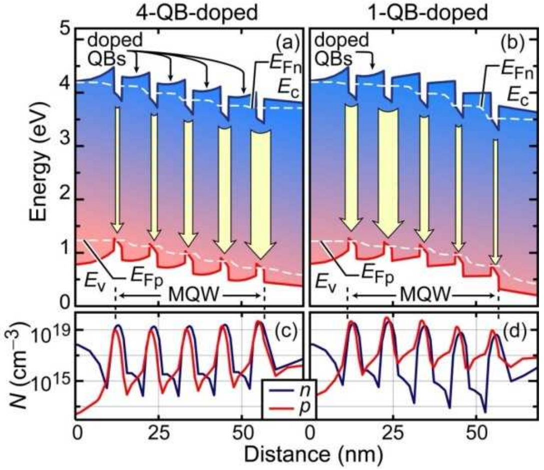
- Analytic model for efficiency droop
- Model allows clear predictions

Based on model, proposed future directions are

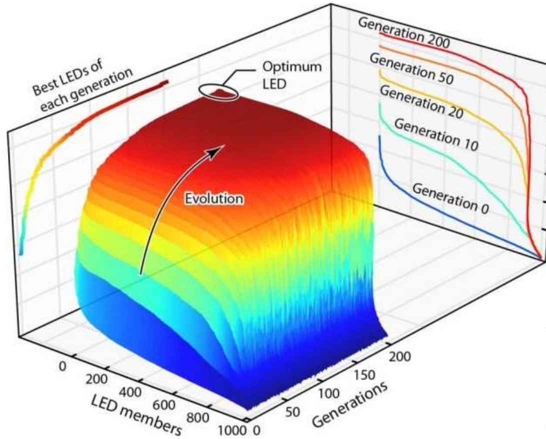
- Enhancement of p-type doping by use of GaInN
- Alternative materials, such as ZnO, for enhanced p-type doping

RPI-led studies on carrier transport and modeling

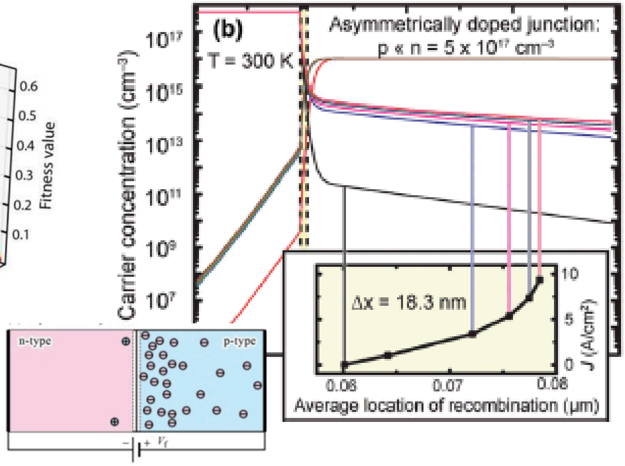
Impact of barrier doping on carrier distribution ***D. Zhu et al.***



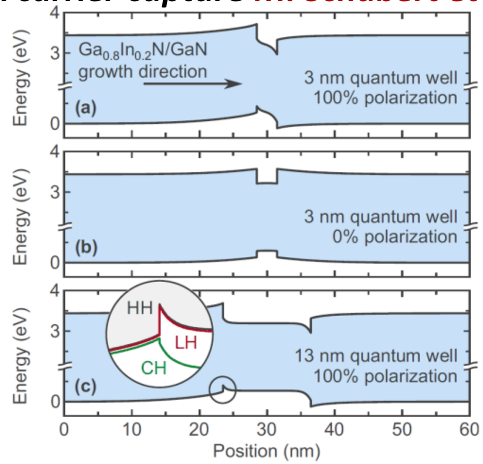
Genetic algorithms for innovative, High-efficiency LED designs ***D. Zhu et al.***



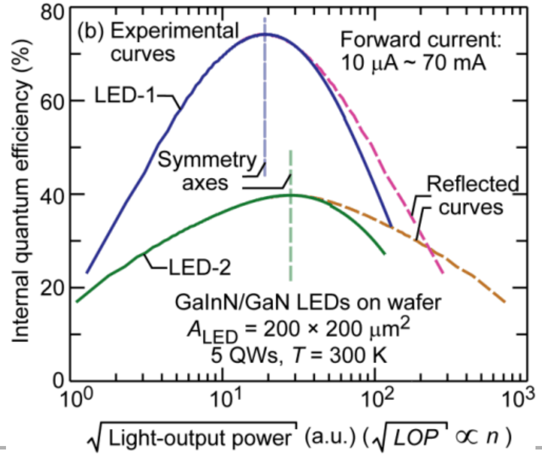
Carrier transport asymmetry leading to efficiency droop ***D. Meyaard et al.***



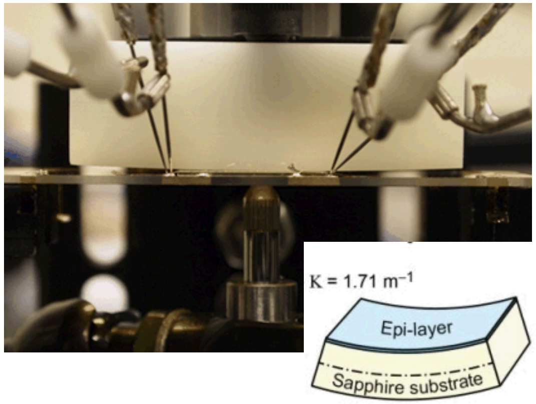
Effect of well width and polarization on carrier capture ***M. Schubert et al.***



Asymmetry of LED IQE curves and implications for efficiency droop ***Q. Dai et al.***



Manipulation of polarization fields via wafer bending ***J. Xu et al.***



Publications

1. Dai, Qi; Shan, Qifeng; Wang, Jing; Chhajed, Sameer; Cho, Jaehee; Schubert, E. Fred; Crawford, Mary H.; Koleske, Daniel D.; Kim, Min-Ho; and Park, Yongjo Carrier recombination mechanisms and efficiency droop in GaInN/GaN light-emitting diodes, *Applied Physics Letters* **97**, 133507 (2010).
2. Chow, Weng W.; Crawford, Mary H.; Tsao, Jeffrey Y.; and Kneissl, Michael Internal efficiency of InGaN light-emitting diodes: Beyond a quasiequilibrium model, *Applied Physics Letters* **97**, 121105 (2010).
3. Zhu, D.; Noemaun, A. N.; Schubert, Martin F.; Cho, J.; Schubert, E. Fred; Crawford, Mary H.; and Koleske, Daniel D. Enhanced electron capture and symmetrized carrier distribution in GaInN light-emitting diodes having tailored barrier doping, *Applied Physics Letters* **96**, 121110 (2010).
4. Shan, Qifeng; Dai, Qi; Chhajed, Sameer; Cho, Jaehee; and Schubert, E. Fred Analysis of thermal properties of GaInN light-emitting diodes and laser diodes, *Journal of Applied Physics* **108**, 084504 (2010).
5. Chhajed, Sameer; Cho, Jaehee; Schubert, E. Fred; Kim, Jong Kyu; Koleske, Daniel D.; Crawford, Mary H. Temperature-dependent Light-output characteristics of GaInN light-emitting diodes with different dislocation densities, *Physica Status Solidi A* **208**, 947 (2011).
6. Dai, Qi; Shan, Qifeng; Cho, Jaehee; Schubert, E. Fred; Crawford, Mary H.; Koleske, Daniel D.; Kim, Min-Ho; and Park, Yongjo On the symmetry of efficiency-versus-carrier-concentration curves in GaInN/GaN light-emitting diodes and relation to droop-causing mechanisms, *Applied Physics Letters* **98**, 033506 (2011).
7. Chow, Weng W. Modeling excitation-dependent bandstructure effects on InGaN light-emitting diode efficiency, *Optics Express* **19**, 21818 (2011).
8. Xu, Jiuru; Schubert, Martin F.; Zhu, Di; Cho, Jaehee; Schubert, E. Fred; Shim, Hyunwook; and Sone, Cheolsoo Effects of polarization-field tuning in GaInN light-emitting diodes, *Applied Physics Letters* **99**, 041105 (2011).

Publications (cont'd) and Presentations

9. Meyaard, David S.; Shan, Qifeng; Dai, Qi; Cho, Jaehee; Schubert, E. Fred; Kim, Min-Ho; and Sone, Cheolsoo ON THE TEMPERATURE DEPENDENCE OF ELECTRON LEAKAGE FROM THE ACTIVE REGION OF GAINN/GAN LIGHT-EMITTING DIODES, *Applied Physics Letters* **99**, 041112 (2011).
10. Meyaard, David S.; Lin, Guan-Bo; Shan, Qifeng; Cho, Jaehee; Schubert, E. Fred; Shim, Hyunwook; Kim, Min-Ho; and Sone, Cheolsoo ASYMMETRY OF CARRIER TRANSPORT LEADING TO EFFICIENCY DROOP IN GAINN BASED LIGHT-EMITTING DIODES, *Applied Physics Letters, **99**,* 251115 (2011).
11. Lin, Guan-Bo; Schubert, Martin F.; Cho, Jaehee; Schubert, E. Fred; and Kim, Hyungkun A COMPLEMENTARY MATCHING TECHNIQUE TO REDUCE THE VARIANCE OF OPTICAL AND ELECTRICAL PROPERTIES OF LIGHT-EMITTING DIODES, *Journal of the Society for Information Display* **19**, 431 (2011).
12. Zhu, Di; Schubert, Martin F.; Xu, Jiuru; Cho, Jaehee; Schubert, E. Fred; Crawford, Mary H.; Koleske, Daniel D. GENETIC ALGORITHM FOR INNOVATIVE DESIGNS IN HIGH EFFICIENCY III-V NITRIDE LIGHT-EMITTING DIODES, *Applied Physics Express, 5, 012102* (2012).

10 Invited Presentations & 2 Plenary Presentations

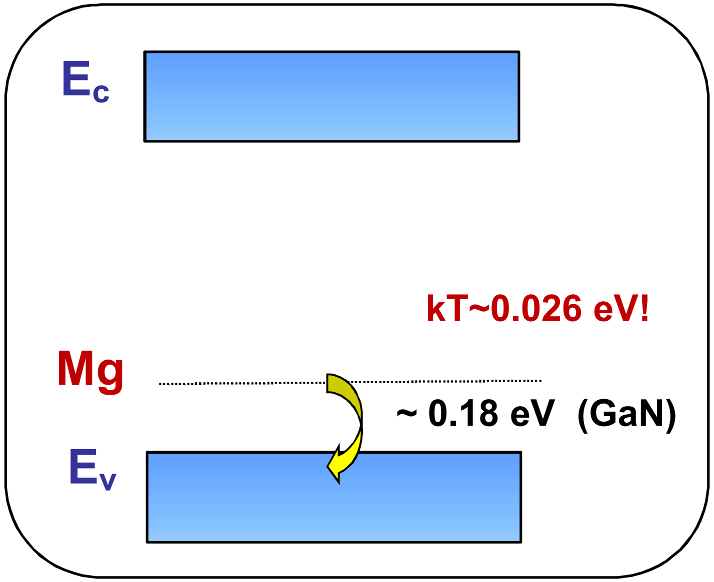
- *SPIE Photonics West,*
- *Materials Research Society Meeting,*
- *American Physical Society Meeting,*
- *Conference on Lasers and Electro-Optics (CLEO),*
- *Nano-Energy Workshop,*
- *Renewable Energy and the Environment Conference (OSA),*
- *American Vacuum Society*

Future Work: investigation of alternative p-type materials for improved carrier transport and efficiency trends

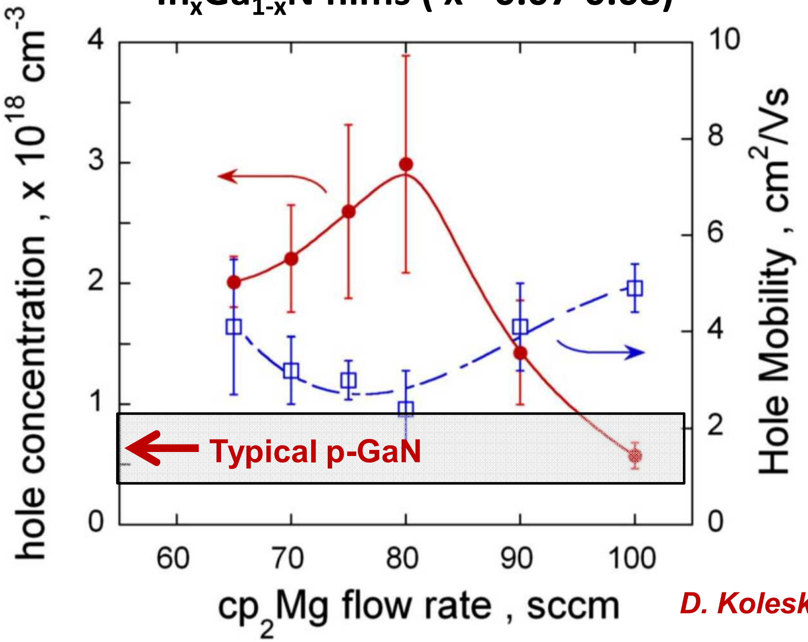
Prediction of Analytic Model (RPI)

- Improvement of p-type doping (p_{p0}) from 5 x 10^{17} to 2 x 10^{18} cm^{-3} …
  - … would reduce *C* by factor of 4
  - … would reduce *D* by factor of 16

Increasing Mg activation energy with bandgap for III-N alloys



Electrical Performance of p-type $In_xGa_{1-x}N$ films (x= 0.07-0.08)



D. Koleske (SNL)

Future Work: Advancement of microscopic recombination model

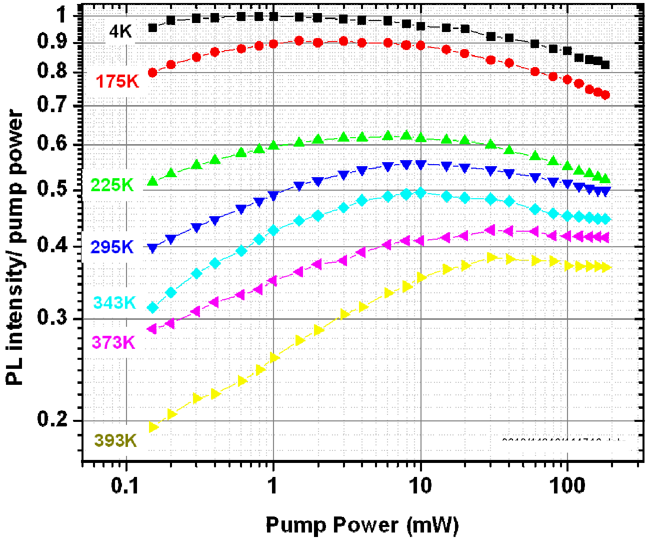
I. Model validation: IQE vs. temperature

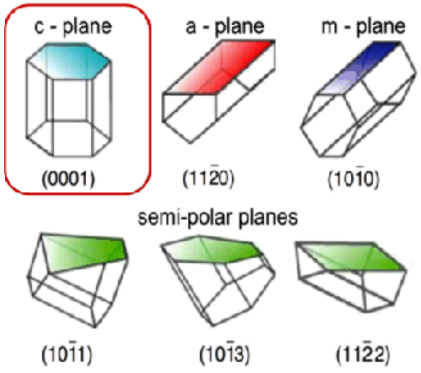
- Fit experimental IQE trends vs. temperature

II. Extend model to compute emission spectra

- Determine carrier distributions (carrier density and plasma temperature) in experiments
- **Challenge:** incorporate many-body Coulomb effects

InGaN QW IQE via optical pumping

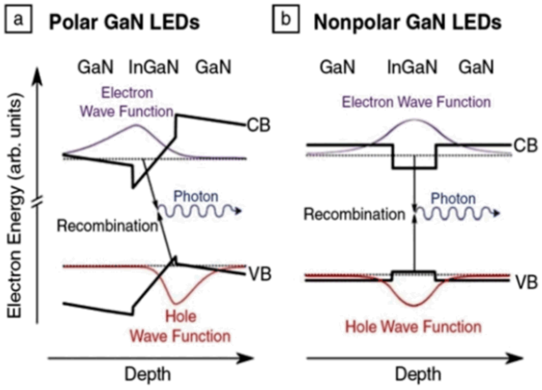




Speck & Chichibu, MRS Bulletin 34, 304 (2009)

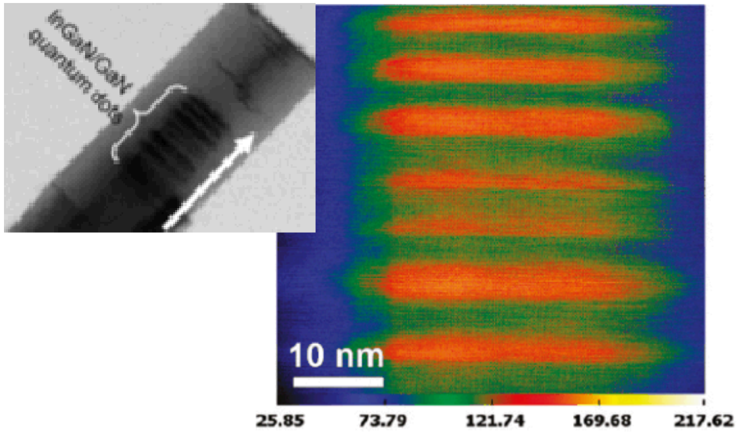
III. Compare polar versus non-polar LEDs

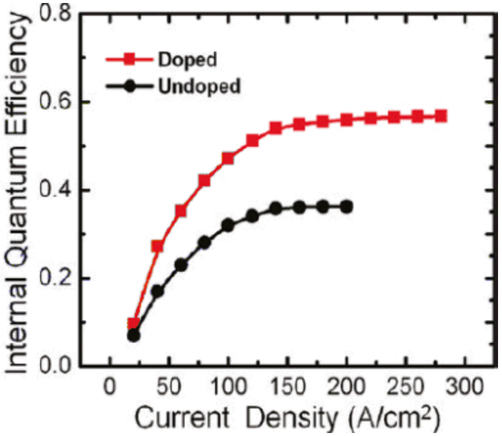
Challenge: Extend model to account for increased bandstructure asymmetries with arbitrary crystal orientations



New Direction: Efficiency studies of nanowires and other lower dimensional structures*

* Collaboration with Nanowire and Defect-Carrier Interactions Research Challenges





Recent publications suggest little or no efficiency droop in nanowire LED structures

Nguyen et al. Nanolett. 2011
McGill University

Nanowire features:
- no extended defects
- polar or nonpolar QW orientations
- improved hole injection with InGaN barriers (strain accommodation)
- Auger recombination?

Experimental exploration of these features will include:

→ IQE studies via optical and electrical excitation (including time-resolved PL)
→ Cathodoluminescence for spatial luminescence studies,
→ Defect spectroscopy studies of ***point defects*** in NWs and NW LED structures

New Direction: Direct, Dynamical Measurement of Auger Recombination Processes in InGaN*

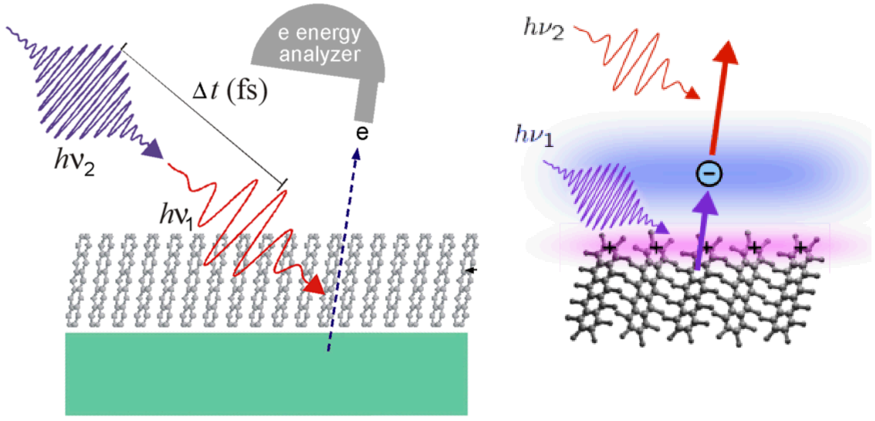
Challenges to direct observation of Auger:

- Involvement of very high energy states in wide bandgap materials
- Ultrafast relaxation of carriers from excited states

Novel Approach: *Collaboration with Prof. Xiaoyang Zhu, UT Austin (EFRC:CST)

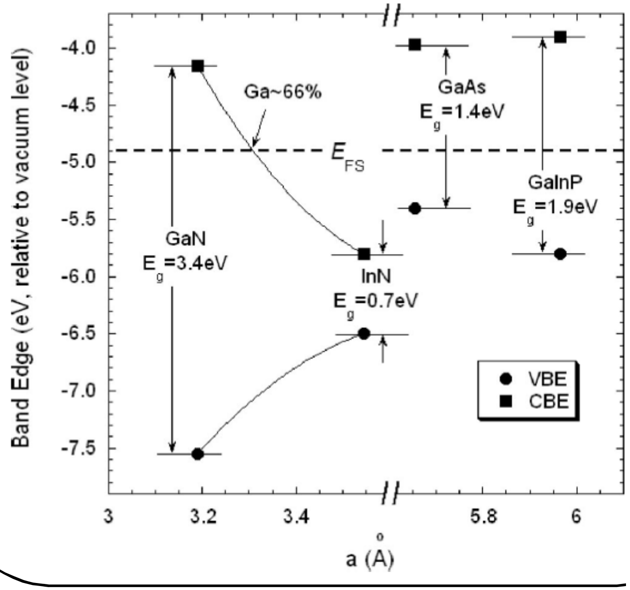
- Time-resolved ***photoemission*** studies of excited state populations (Auger electrons)

Femtosecond two-photon photoemission spectroscopy



Tracking evolution of excited state populations with sub-picosecond time resolution

Band-edge Relative to Vacuum Level for $In_xGa_{1-x}N$



Li et al.
PRB 2005

Summary:

I. Research Highlights

- **Development of a microscopic model to study efficiency limitations of InGaN LEDs**
    - Revealed intrinsic contributions to efficiency vs. wavelength trends (e.g., green-yellow gap)
    - Revealed dominant contributions to LED efficiency vs. temperature trends
    - Excitation dependence bandstructure influences on LED efficiency (POSTER)
    - Multi-level defect contributions to efficiency droop (POSTER)

- **Analysis of carrier transport contributions to efficiency droop**
    - Insights into the role of barrier doping on carrier distributions and efficiency droop
    - Genetic algorithm for design of high-efficiency LEDs
    - Analytic model for efficiency droop, quantifying the benefit of improved p-type doping
    - Numerous additional modeling and experimental studies of carrier transport effects

II. Future Work

- Enhanced p-type materials for transport asymmetry studies
- Recombination model validation and extension to arbitrary crystal orientations
- New Directions: nanowire efficiency studies and Auger recombination studies via photoemission spectroscopy (UT Austin)