Charge carrier localization and radiative processes in III-nitrides & perovskites

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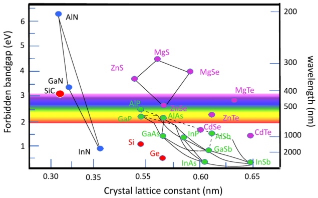
Context

Context for the study of nitride ternary alloys

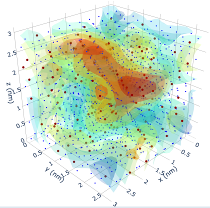
Nitrides = III-N semiconductors

Bandgap engineering

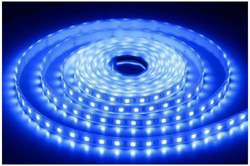


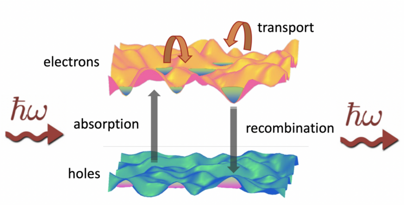
C. Weisbuch, Comptes Rendus Physique, Volume 19, Issue 3 (2018)

$In_x Ga_{1-x} N$

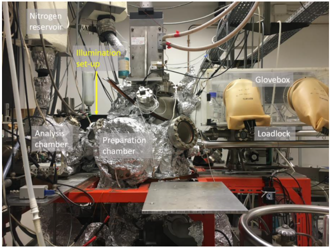


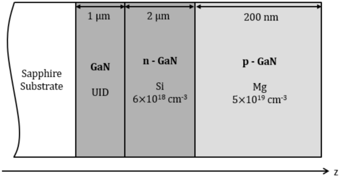
LED, lighting applications

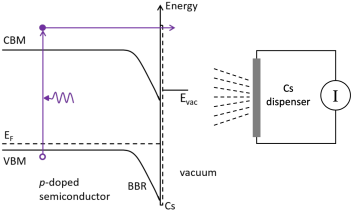




Photoemission experiment on GaN and InGaN





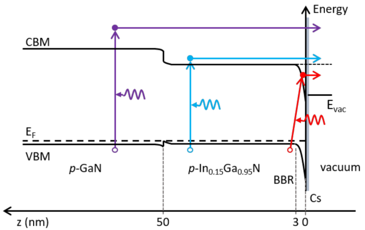


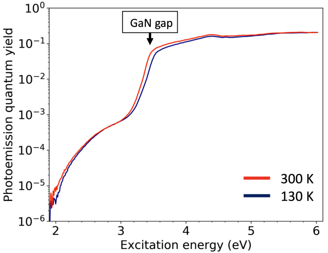
Three-step process

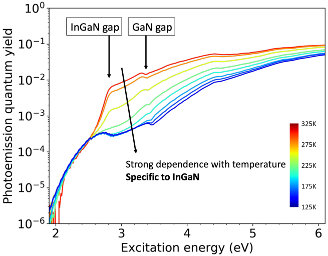
- Photon absorption, creation of e-h pair.
- Electron relaxation and transport in the conduction band.
- Electron transmission through the surface.

Freezing of electron transport in InGaN

Low energy photoemission experiment





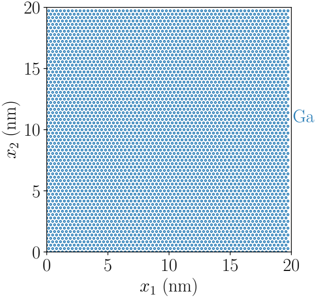


Proof for electron localization?

⋆ M. Sauty *et al*, Phys. Rev. Lett. **129**, 216602 (2022)

Effective mass model of disordered semiconductor alloys

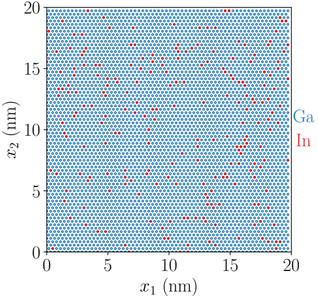
From ordered GaN...



Eigenstates in the conduction band of a periodic potential (Bloch waves)

$$\psi_\mu^{(c)}(\mathbf{r}) = \underbrace{u_c(\mathbf{r})}_{\text{cell function}} \underbrace{\exp(i\,\mathbf{k}_\mu \cdot \mathbf{r})}_{\text{plane wave}}$$

From ordered GaN... to disordered InGaN



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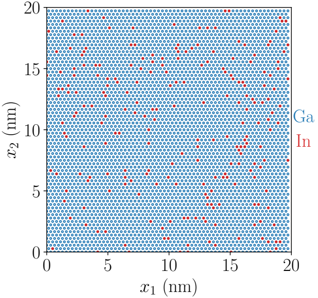
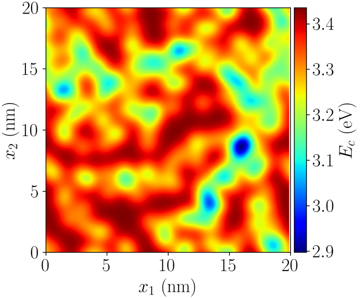




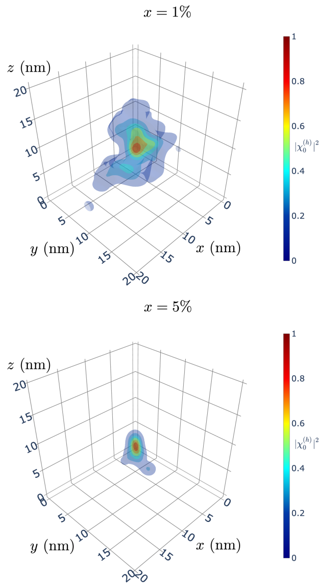
Eigenstates in the conduction band in the *effective mass approximation*

$$\psi_\mu^{(c)}(\mathbf{r}) = \underbrace{u_c(\mathbf{r})}_{\text{cell function}} \underbrace{\chi_\mu^{(c)}(\mathbf{r})}_{\text{envelope}} \quad \text{and} \quad \psi_\nu^{(v)}(\mathbf{r}) = u_v(\mathbf{r})\,\chi_\nu^{(v)}(\mathbf{r})$$

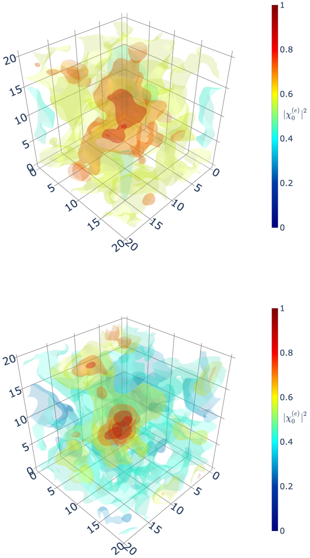
Schrödinger equation

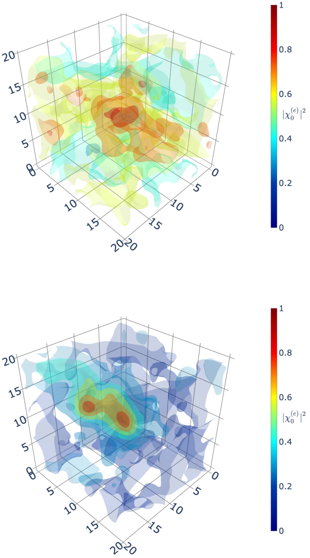
$$-\frac{\hbar^2}{2}\nabla\cdot\left[\frac{\nabla\,\chi_\mu^{(c)}}{m_c}\right] + E_c(\mathbf{r})\,\chi_\mu^{(c)} = E_\mu^{(c)}\,\chi_\mu^{(c)}$$

Examples of hole wave function ($In_xGa_{1-x}N$)

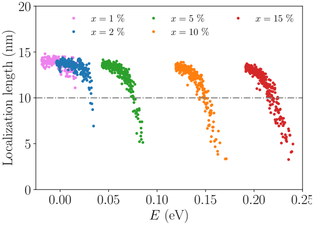


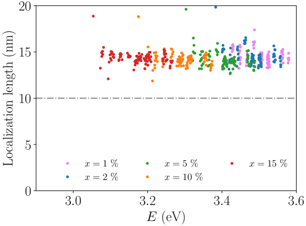
Examples of electron wave function ($In_xGa_{1-x}N$)



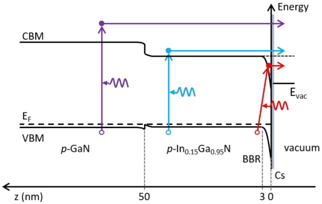


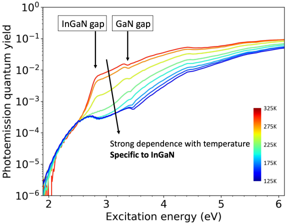
Localization length vs energy ($In_xGa_{1-x}N$)





Freezing of electron transport but delocalized electrons?
Electron-hole Coulomb interaction!





⋆ M. Sauty *et al*, Phys. Rev. Lett. **129**, 216602 (2022)

⋆ A. David and C. Weisbuch, Phys. Rev. Research 4, 043004 (2022)

Impact of alloy disorder on radiative properties & localization landscape

What is the localization landscape?

Original motivation: finding a bounding function to the eigenfunctions.

Schrödinger eigenvalue problem

$$-\frac{\hbar^2}{2m}\Delta\psi + V\psi = E\psi$$

Integral representation for ψ

$$\psi(\mathbf{r}) = \int G(\mathbf{r},\mathbf{r}')E\psi(\mathbf{r}')\,\mathrm{d}^d r'$$

Straightforward upper bound

$$|\psi(\mathbf{r})| \leq \int |G(\mathbf{r},\mathbf{r}')E\psi(\mathbf{r}')|\,\mathrm{d}^d r' \leq |E|\|\psi\|_\infty \int |G(\mathbf{r},\mathbf{r}')|\,\mathrm{d}^d r'$$

Hence

$$\frac{|\psi(\mathbf{r})|}{\|\psi\|_\infty} \leq |E|\mathcal{L}(\mathbf{r})$$

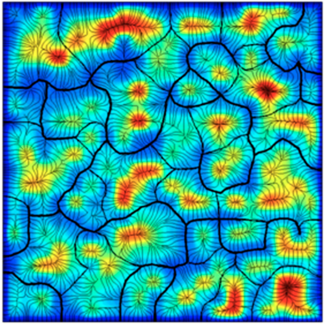
If $G \geq 0$, the landscape is easily obtained by solving

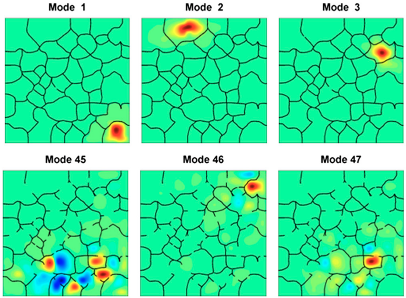
$$H\mathcal{L} = -\frac{\hbar^2}{2m}\Delta\mathcal{L} + V\mathcal{L} = 1$$

⋆ M. Filoche and S. Mayboroda, Proceedings of the National Academy of Sciences 109, 14761 (2012)

The landscape bounds the eigenstates

$$\frac{|\psi(\mathbf{r})|}{\|\psi\|_\infty} \leq |E|\mathcal{L}(\mathbf{r})$$





⋆ M. Filoche and S. Mayboroda, Proceedings of the National Academy of Sciences 109, 14761 (2012)

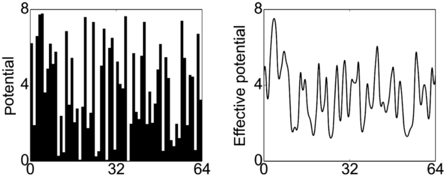
$$H\mathcal{L} = 1$$

Change of unknown function $\psi = \mathcal{L}\varphi$

$$-\frac{\hbar^2}{2m}\Delta(\mathcal{L}\varphi) + V\mathcal{L}\varphi = E\mathcal{L}\varphi$$

$$\vdots$$

$$\underbrace{-\frac{\hbar^2}{2m}\frac{1}{\mathcal{L}^2}\nabla\cdot\left[\mathcal{L}^2\nabla\varphi\right]}_{\text{Eff. kinetic energy}} + \underbrace{\frac{1}{\mathcal{L}}}_{\text{Eff. potential}}\varphi = E\varphi$$



⋆ D. N. Arnold *et al.* Phys. Rev. Lett. 116, 056602 (2016)

⋆ D. N. Arnold *et al.* SIAM J. Sci. Comput., 41(1), B69–B92 (2019)

Landscape based absorption coefficient

Absorption coefficient

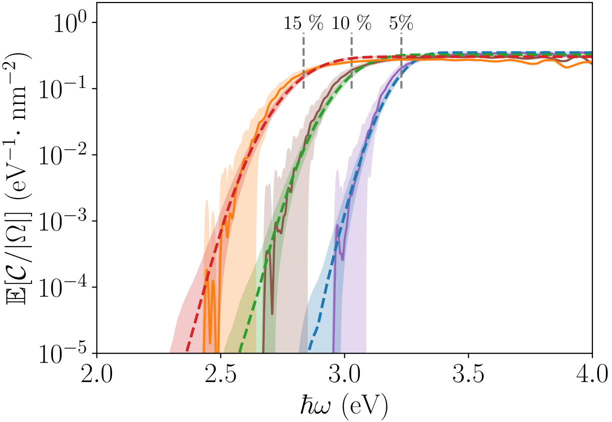
$$\alpha(\omega) \propto \sum_{\mu,\nu} \underbrace{\left| \langle \chi_\mu^{(c)} | \chi_\nu^{(v)} \rangle \right|^2}_{\text{coupling}} \underbrace{\delta\left(E_\mu^{(c)} - E_\nu^{(v)} - \hbar\omega\right)}_{\text{energy conservation}}$$

can be shown to be approximated by the *landscape based semi-classical approximation*

$$\alpha(\omega) \propto \int m_r^{3/2}(\mathbf{r}) \sqrt{\left(\hbar\omega - E_g^{(\text{eff})}(\mathbf{r})\right)_+} \, \mathrm{d}r$$

⋆ J.-P. Banon, P. Pelletier, C. Weisbuch, S. Mayboroda, M. Filoche, Phys. Rev. B **105**, 125422 (2022)

Simulated absorption curves

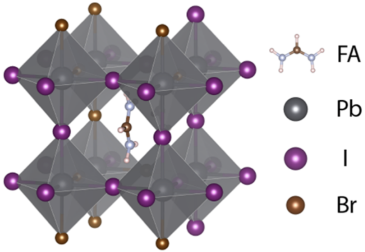


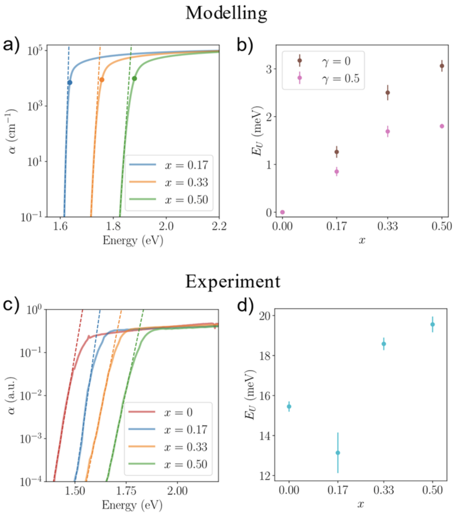
Normalized absorption coefficient spectra for 2D alloys of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{N}$ averaged over 100 realizations. Domain size 50 nm $\times$ 50 nm.

Computational speed-up ≈ 300.

$\star$ J.-P. Banon, P. Pelletier, C. Weisbuch, S. Mayboroda, M. Filoche, Phys. Rev. B **105**, 125422 (2022)

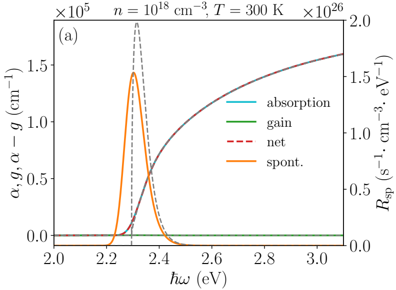
Absorption coefficient and Urbach tail in perovskites

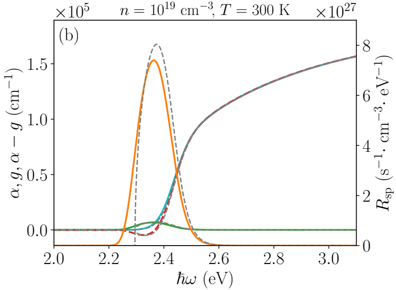




⋆ Y. Liu, *et al.*, ACS Energy Letters, 2023, 8, 1, 250–258.

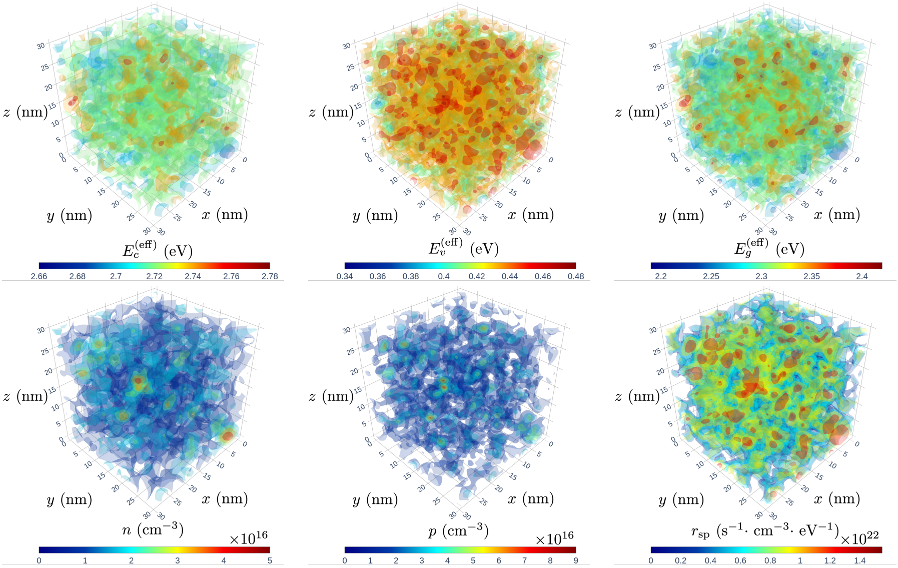
Absorption and light emission for $In_{0.3}Ga_{0.7}N$





How does it look like locally?

Parameters: $x = 0.3$, $T = 300$ K, $\bar{n} = \bar{p} = 10^{16}$ cm^{-3}



Contributors

Wigner-Weyl theory of absorption

M. Filoche

S. Mayboroda

P. Pelletier

Absorption in perovskites

R. Friend

Y. Liu

(De)localization in III-V vs III-N

J. Speck

C. Weisbuch

Evidence of localization in InGaN

J. Peretti

M. Sauty

Fundings: Work supported by the Simons foundation grant 601944.

Thank you for your attention.

A few references to know more about ...

... photoemission experiment in InGaN

⋆ M. Sauty, N. M. S. Lopes, J.-P. Banon, Y. Lassailly, L. Martinelli, A. Alhassan, Y. Chao Chow, S. Nakamura, J. S. Speck, C. Weisbuch, J. Peretti, Phys. Rev. Lett. **129**, 216602 (2022)

... the original paper on the localization landscape

⋆ M. Filoche and S. Mayboroda, PNAS **109**, 14761 (2012)

... the effective potential and the modified Weyl law for the IDOS

⋆ D. N. Arnold, G. David, D. Jerison, S. Mayboroda, and M. Filoche, Phys. Rev. Lett. **116**, 056602 (2016)

... absorption in disordered semiconductors

⋆ J.-P. Banon, P. Pelletier, C. Weisbuch, S. Mayboroda, M. Filoche, Phys. Rev. B **105**, 125422 (2022).

... energy landscape and absorption in lead mixed halide perovskites

⋆ Y. Liu, J.-P. Banon, K. Frohna, Y-H Chiang, G. Tumen-Ulzii, S. D. Stranks, M. Filoche, and R. H. Friend, ACS Energy Letters, 2023, 8, 1, 250–258.