Perovskite Light Emitting Diodes: Efficiency roll-off & Sub-E_g operation

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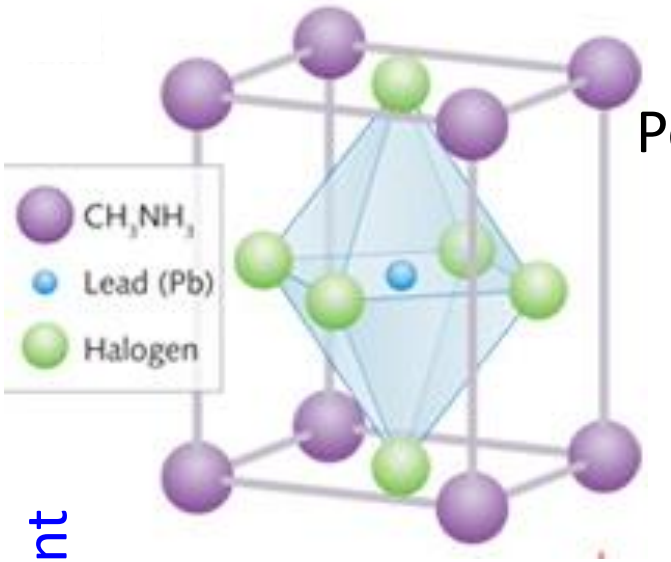
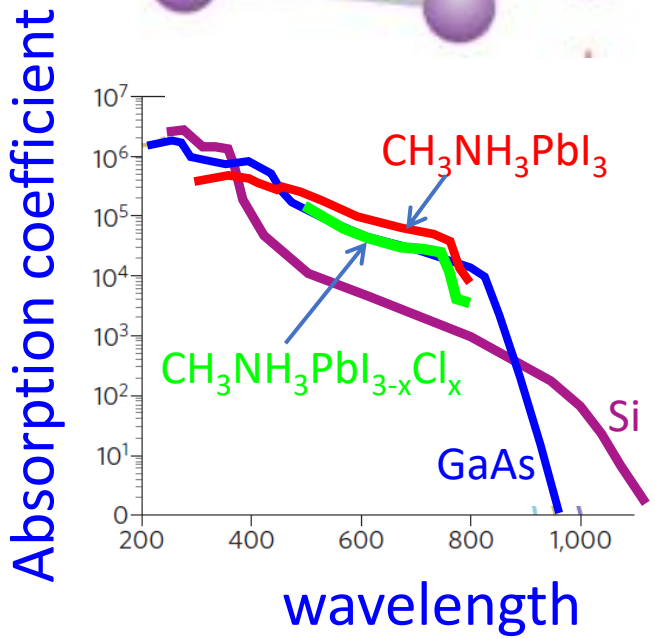
Perovskite optoelectronics



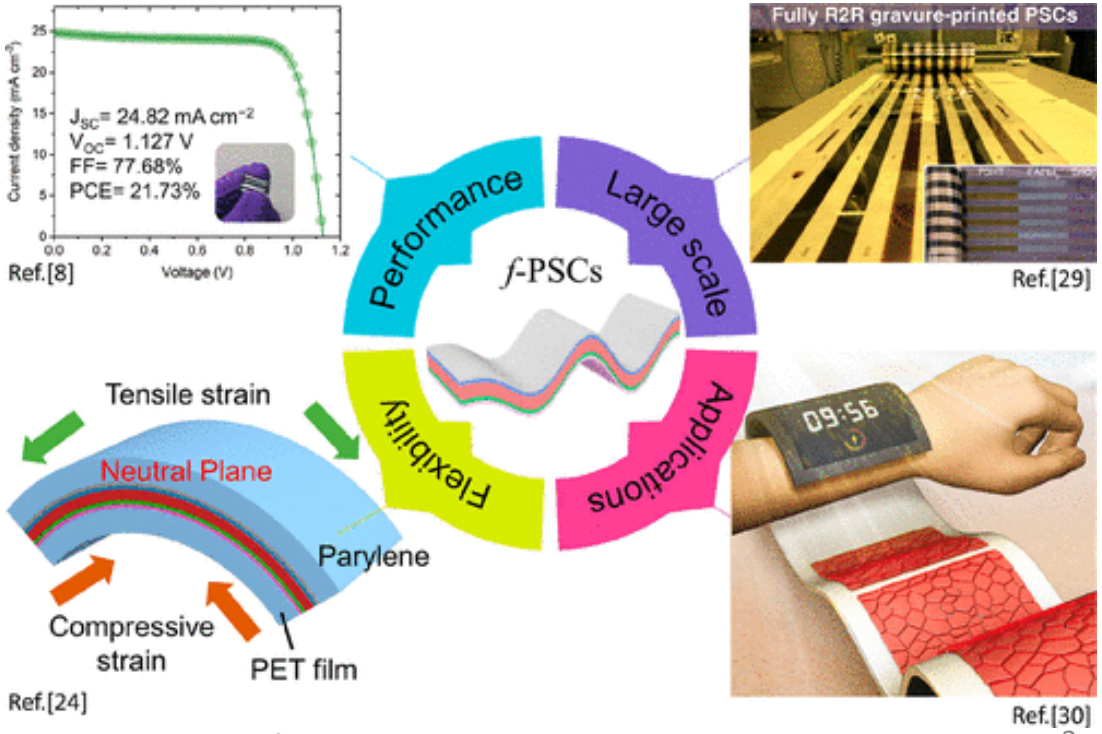
Perovskite : **ABX_3**

$CH_3NH_3PbBr_3$

$CH_3NH_3PbI_3$

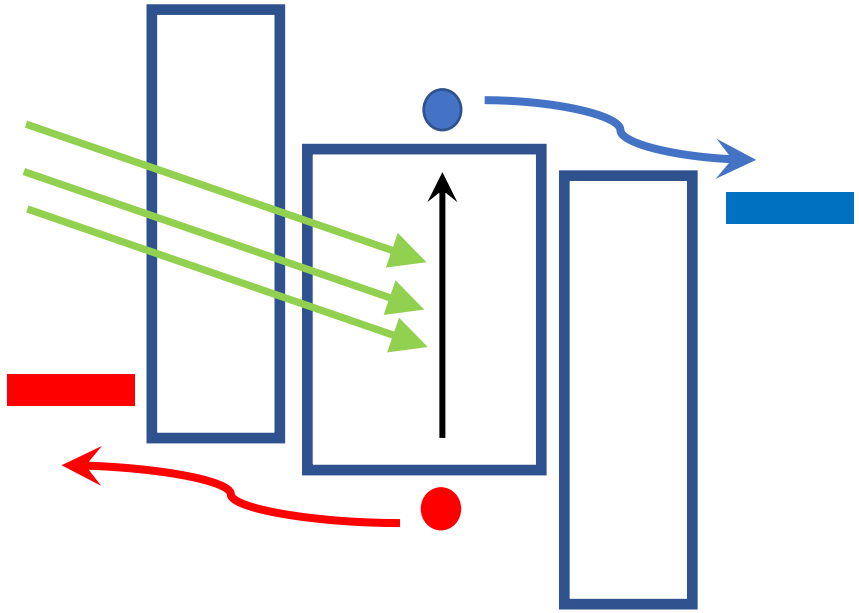


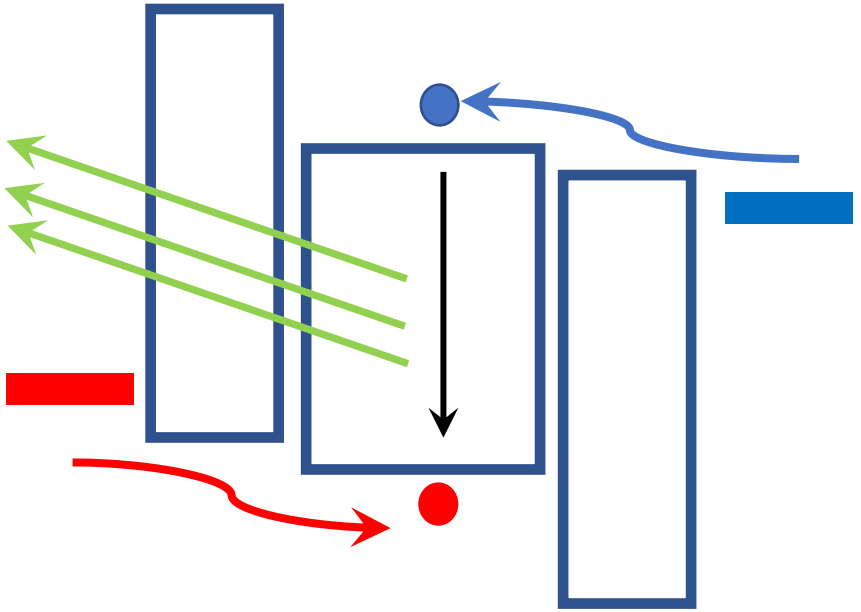
M. A. Green, et al., *Nature Photonics,* 2014.



Gao et al., ACS energy Letters 2022

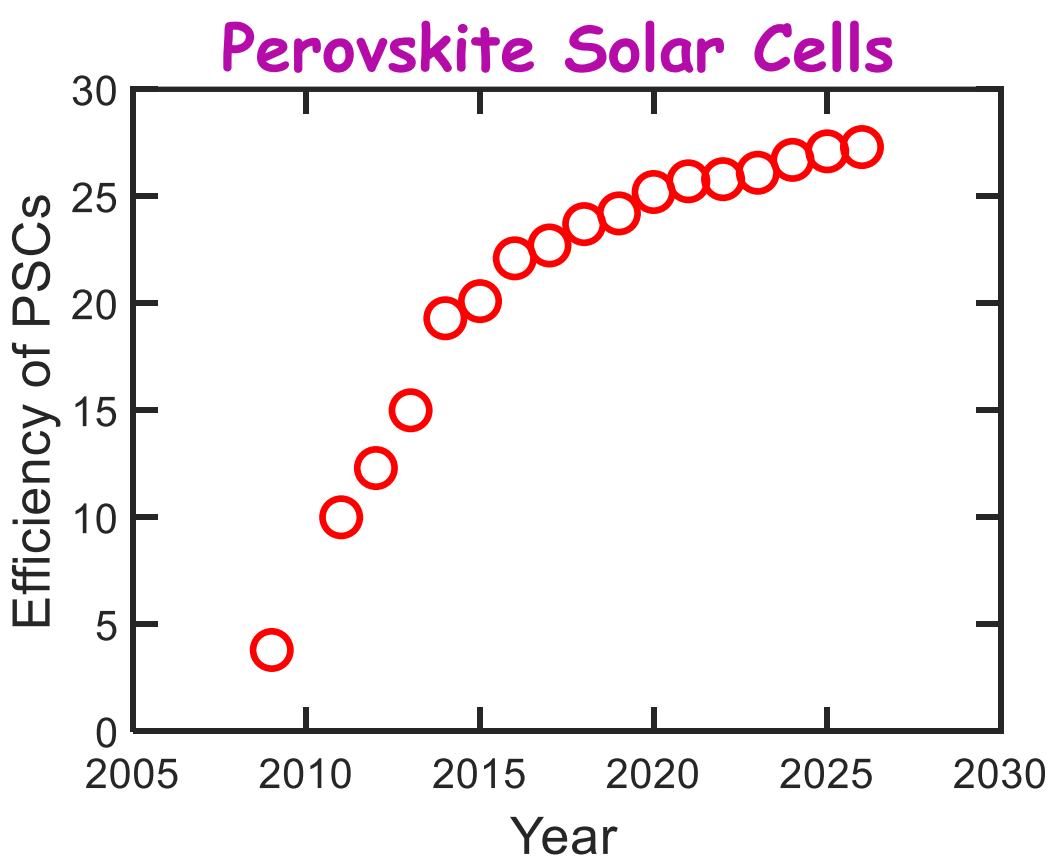
Optoelectronic devices

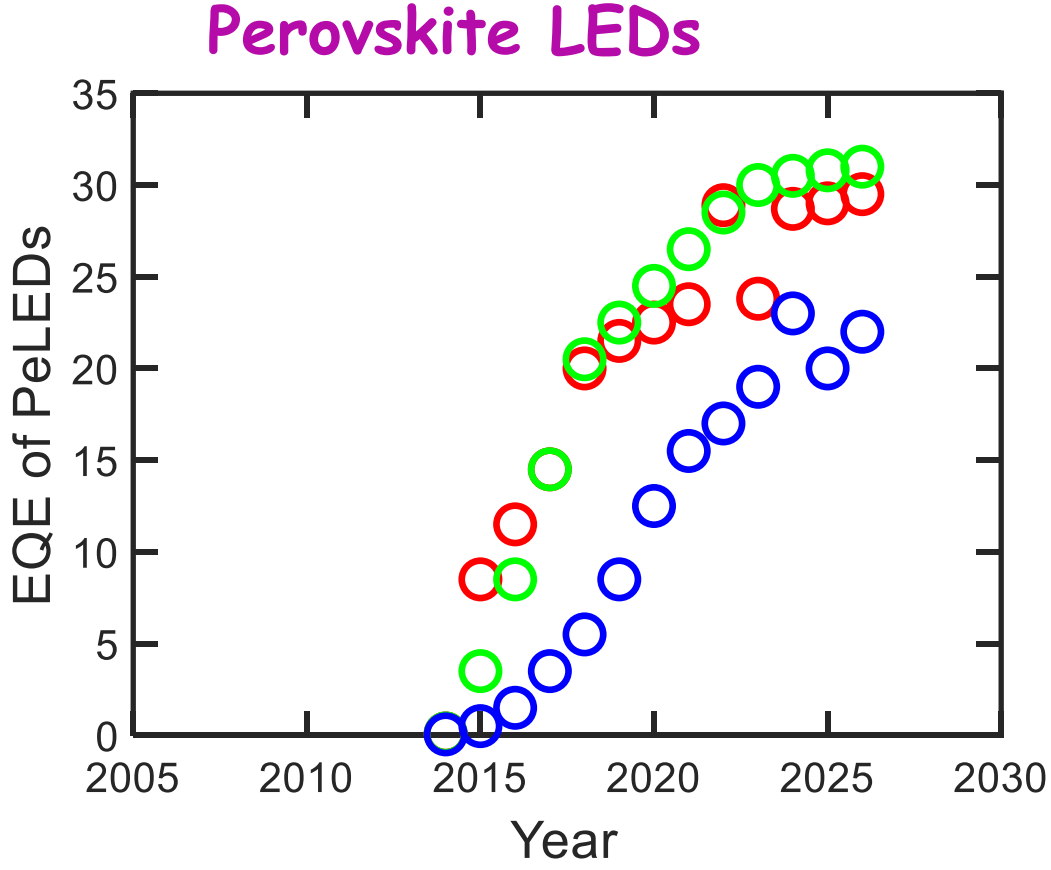




Carrier recombination is the most important phenomena…

Efficiency evolution..





From the literature…

Article

Acceleration of radiative recombination for efficient perovskite LEDs

Li et al., Nature 2024

https://doi.org/10.1038/s41586-024-07460-7

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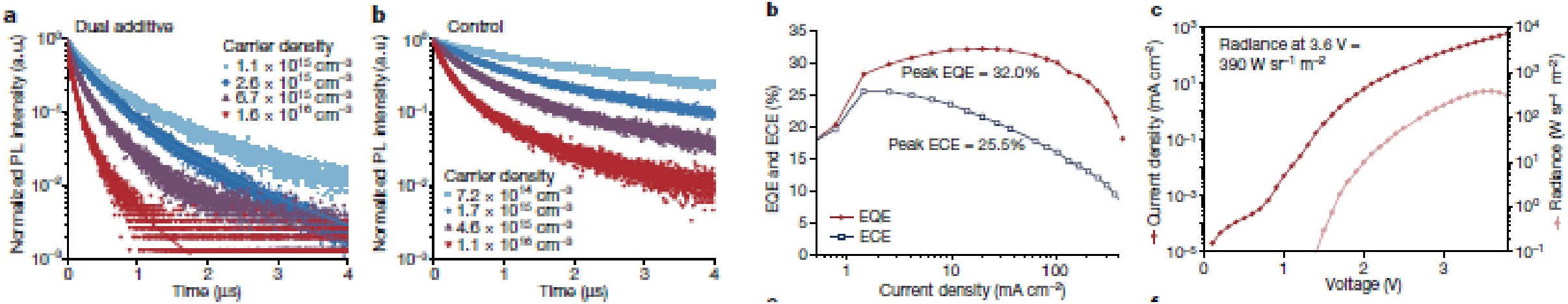
Mengmeng Li[1,2,13], Yingguo Yang[3,13], Zhiyuan Kuang[1,13], Chenjie Hao[1], Saixue Wang[1], Feiyue Lu[1], Zhongran Liu[4], Jinglong Liu[1], Lingjiao Zeng[1], Yuxiao Cai[1], Yulin Mao[5], Jingshu Guo[6], He Tian[4], Guichuan Xing[6], Yu Cao[2,7], Chao Ma[1], Nana Wang[1], Qiming Peng[1], Lin Zhu[1] ✉, Wei Huang[1,7,8,9,10] ✉ & Jianpu Wang[1,11,12] ✉



Why are these devices so good?
What are the performance limiting phenomena?
How much the performance could be improved?

Modeling of Perovskite solar cells and LEDs

$$\varepsilon \frac{\partial E}{\partial x} = q\left(p - n + N_{ion} + N_D^+ - N_A^-\right)$$

$$\frac{\partial n}{\partial t} = \frac{1}{q}\frac{\partial J_n}{\partial x} + R - G$$

$$\frac{\partial p}{\partial t} = -\frac{1}{q}\frac{\partial J_p}{\partial x} + R - G$$

$$\frac{\partial N_{ion}}{\partial t} = -\frac{1}{q}\frac{\partial J_{ion}}{\partial x} + G_{ion}$$

Electronic and Ionic transport

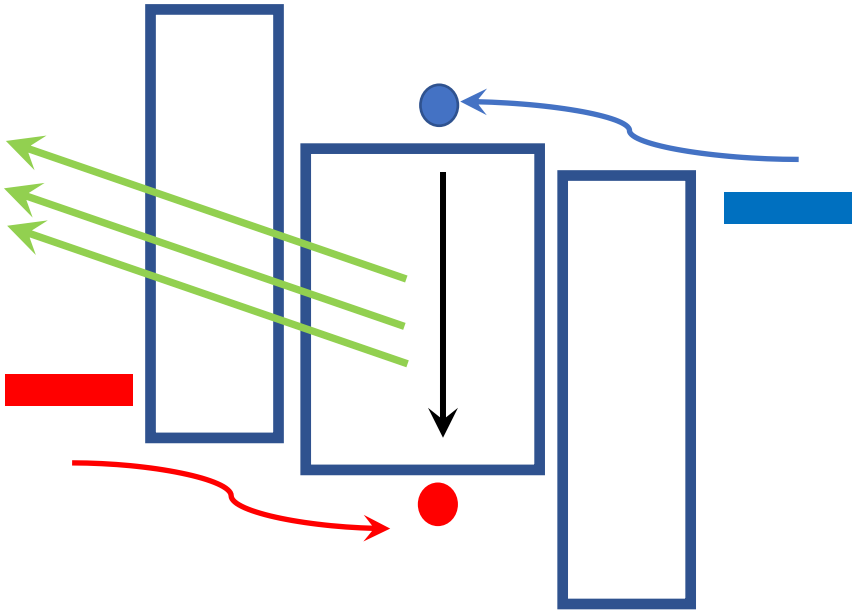
Recombination:
Bulk: Band-Band, trap assisted SRH, Auger,
Interface recombination

Generation:
Light intensity and absorption coefficient

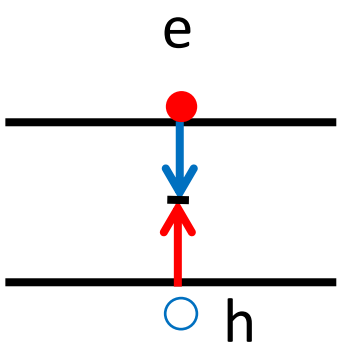
Numerical method:
Scharfetter-Gummel discretization,
Newton method for self-consistent solution
BDF2 time integration
Ions confined to perovskite

ACS Nano 2017, JPCC 2018, JAP 2019, JAP 2021,
ACS Energy Letters 2023, 2025, 2026, JAP 2025, ACS AEM 2025

LED Basics



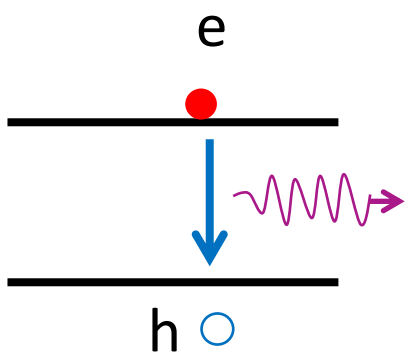
Trap assisted SRH



$k_1 n$

monomolecular

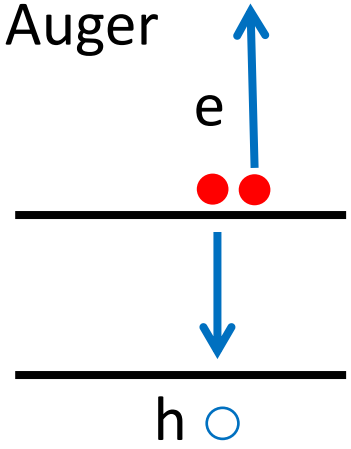
Radiative



$k_2 n^2$

bimolecular

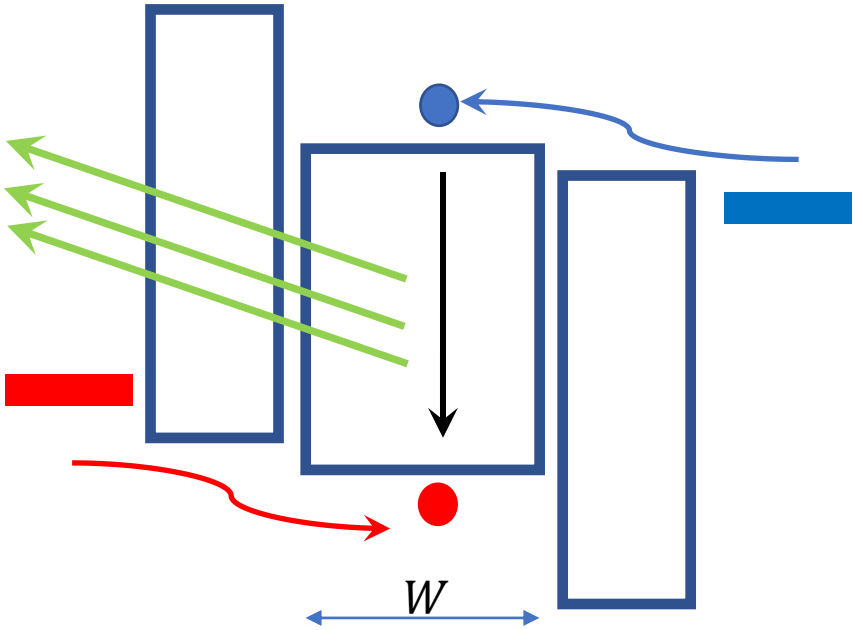
Auger



$k_3 n^3$

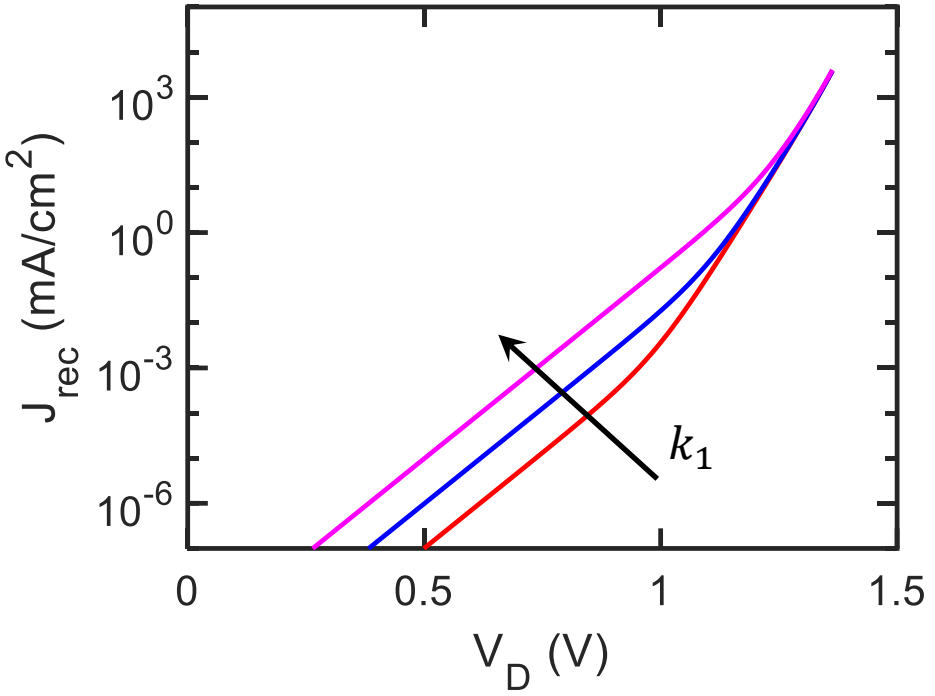
Auger

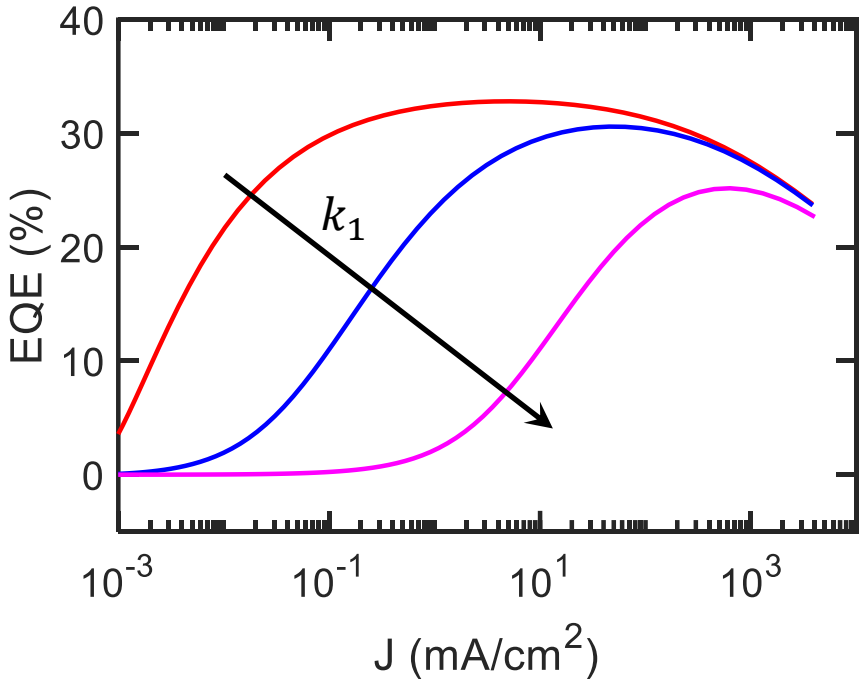
LED Basics



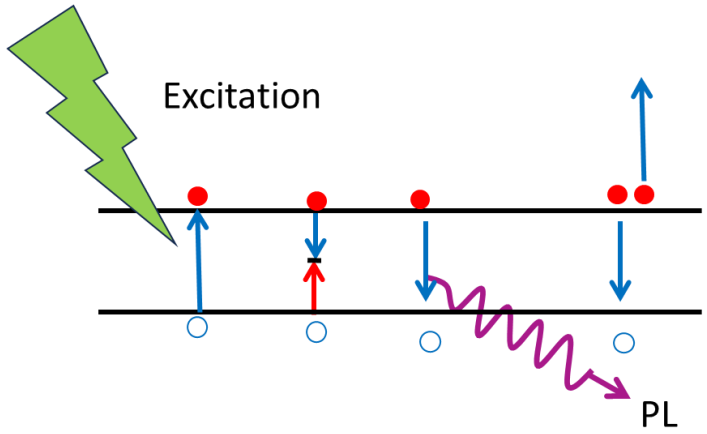
$$J = qW(k_1 n + k_2 n^2 + k_3 n^3)$$

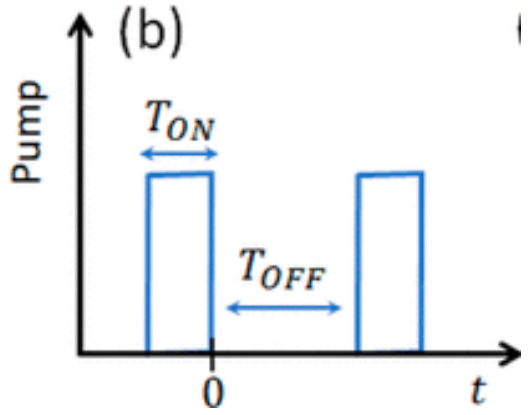
$$EQE = \frac{k_2 n^2 W \times \eta_{OC}}{J/q}$$

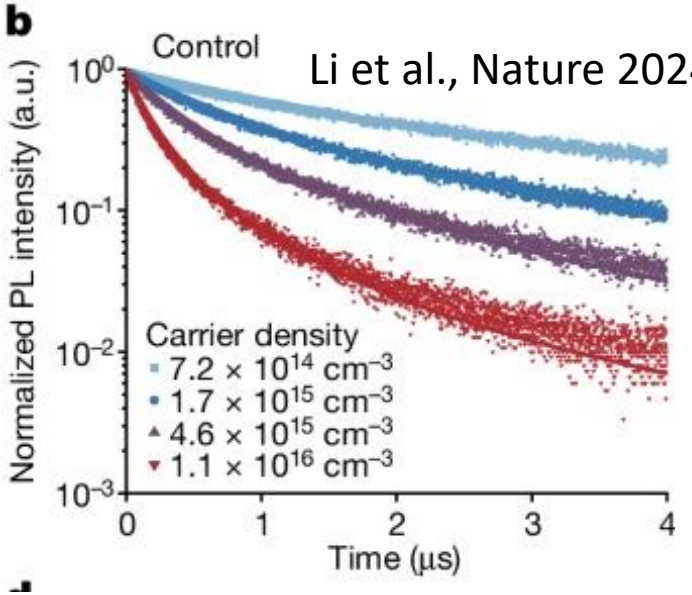




Recombination parameters from PL

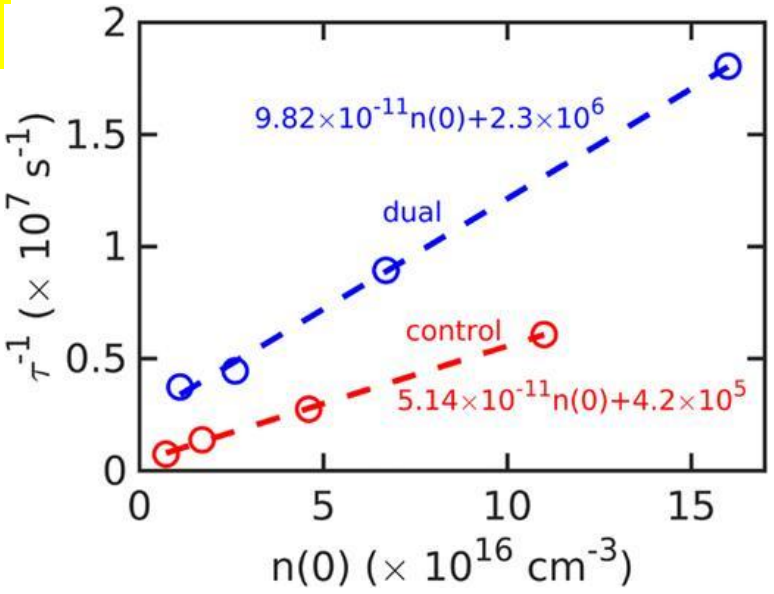


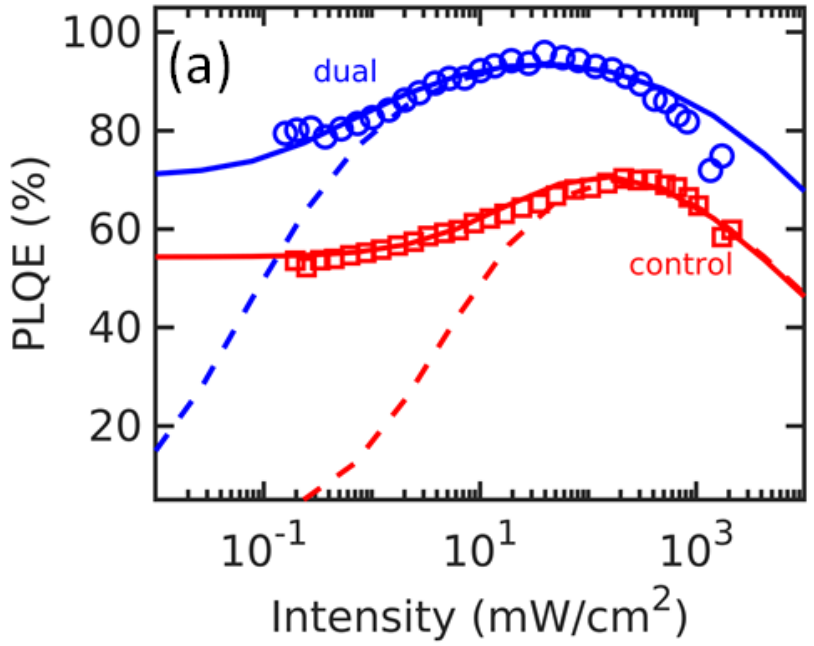




Li et al., Nature 2024

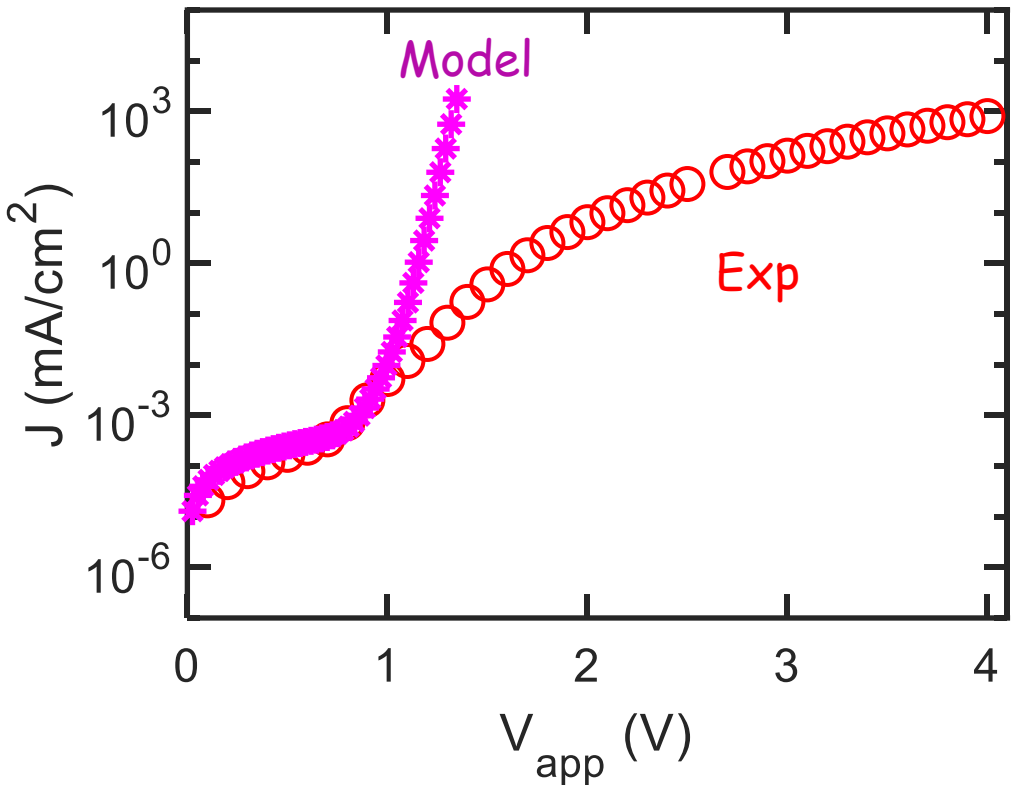
$$\frac{\partial n}{\partial t} = -k_1 n - k_2 n^2 - k_3 n^3$$



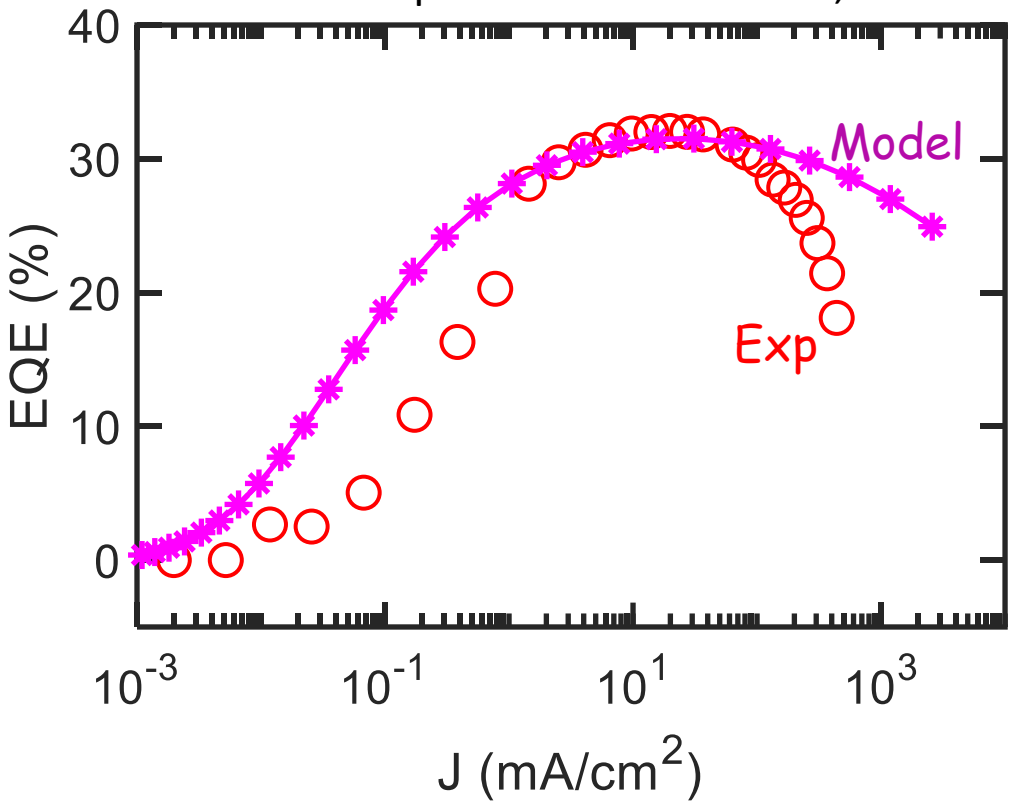


JAP, 2025

Model vs.experiments



Exp results from Li et al., Nature 2024

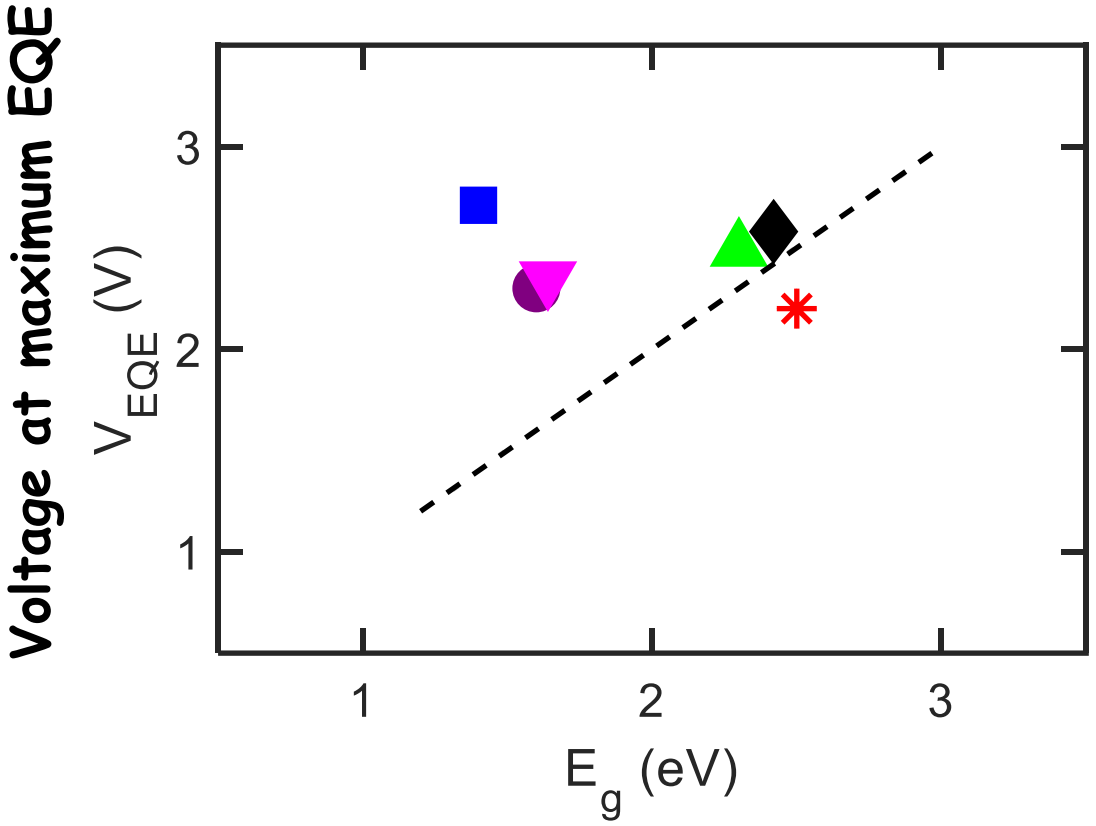


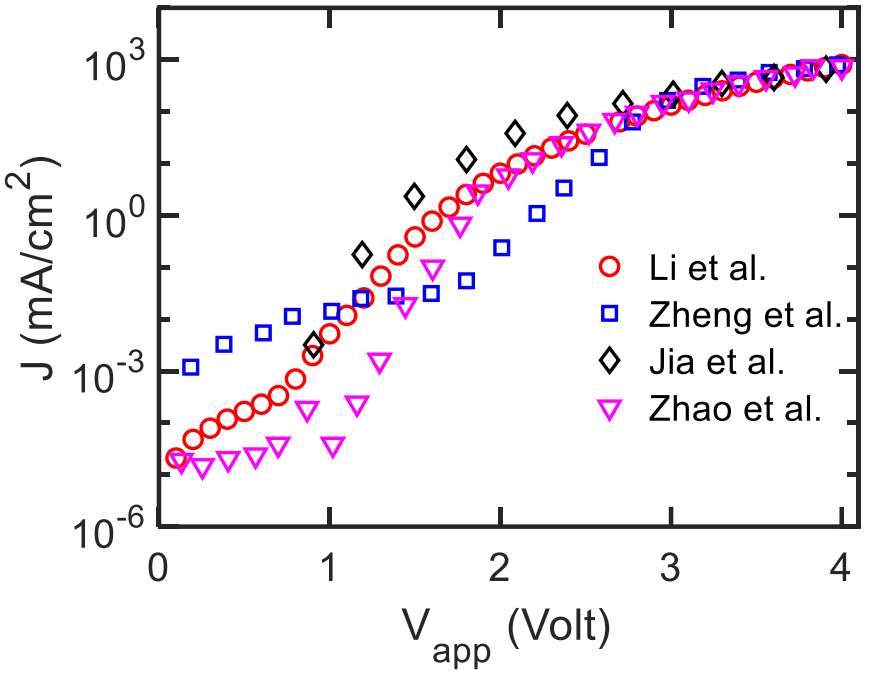
$$J = qW(k_1 n + k_2 n^2 + k_3 n^3)$$

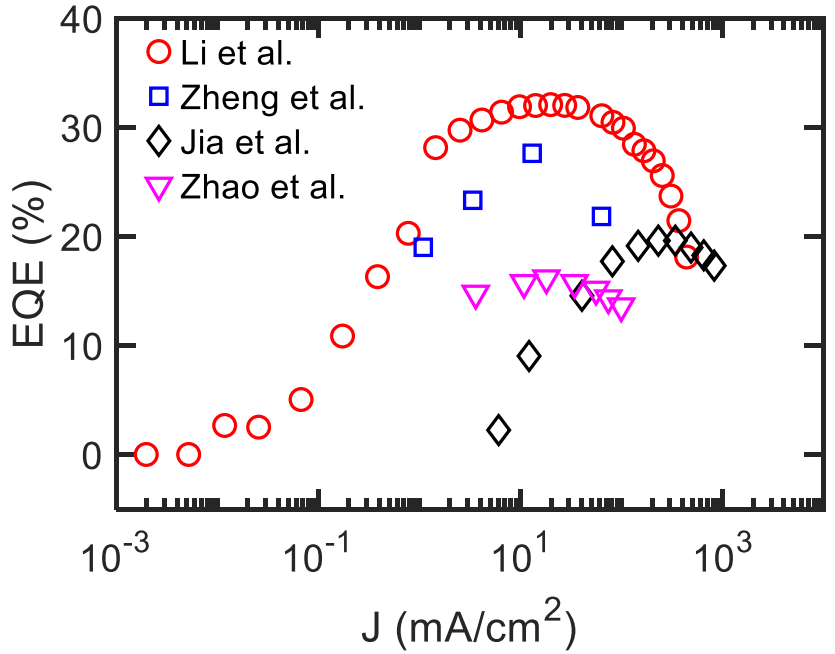
$$EQE = \frac{k_2 n^2 W \times \eta_{OC}}{J/q}$$

EQE roll-off and additional voltage drop are unexplained..

Puzzles galore!



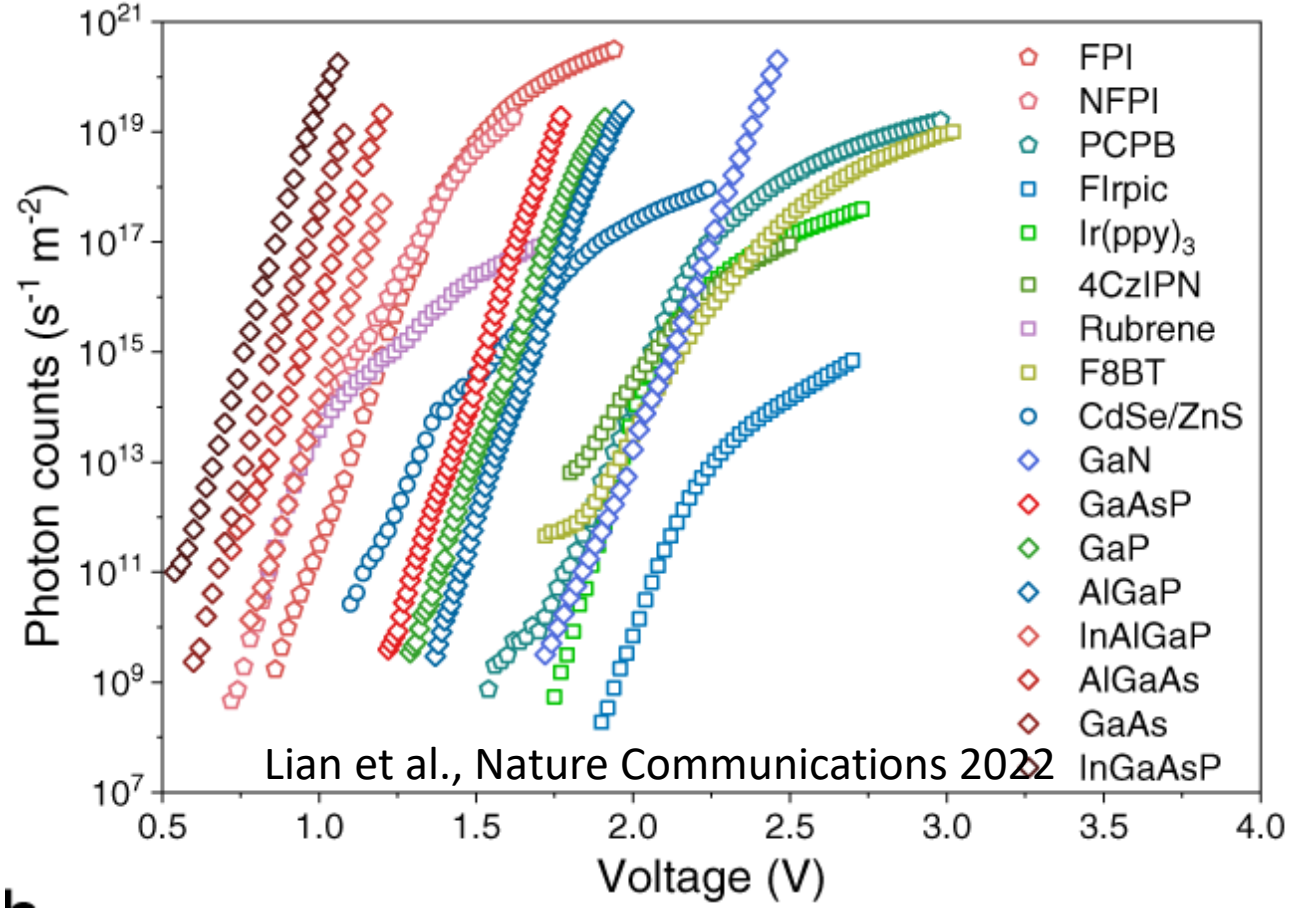


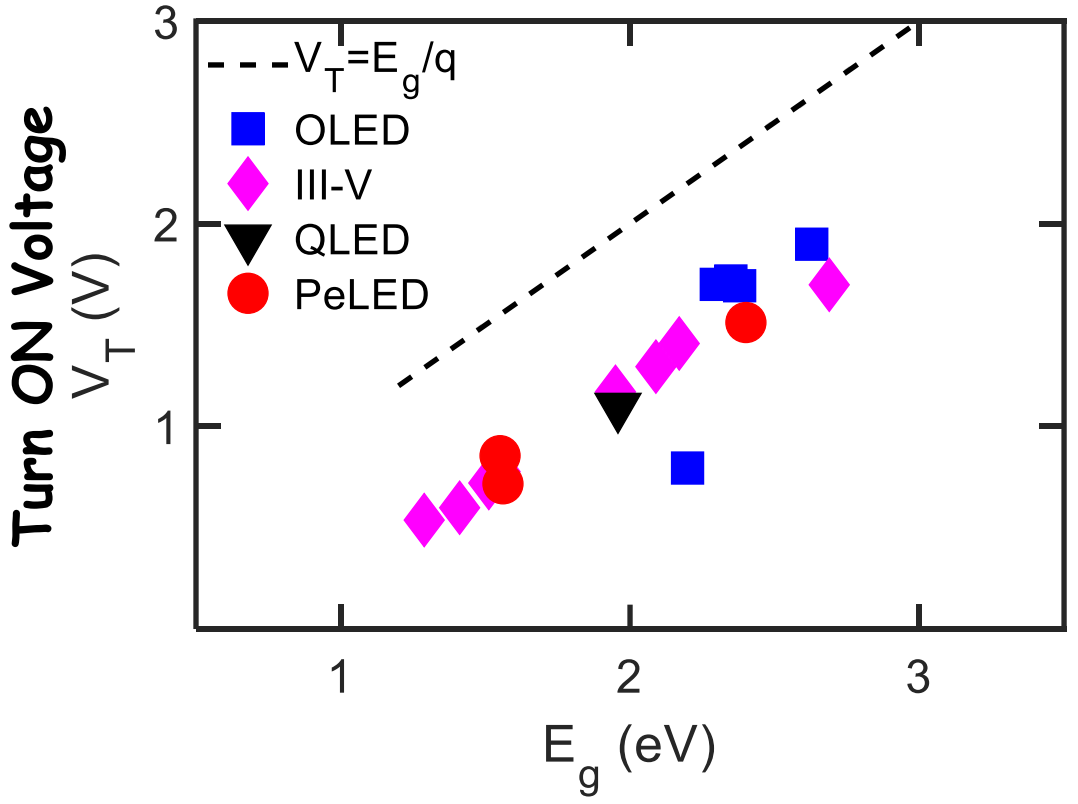


Voltage at maximum EQE (V_{EQE}) seems independent of Eg

Is sub-Eg high efficiency operation possible?

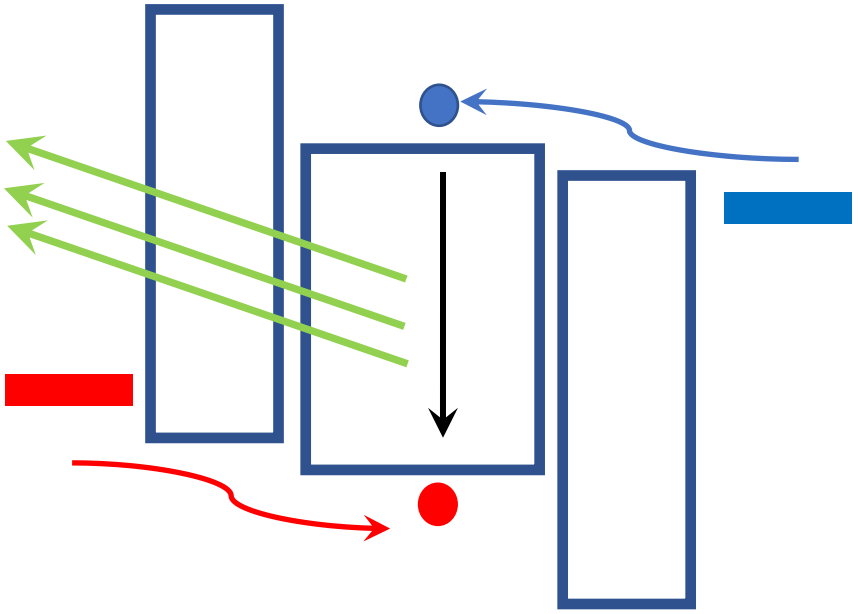
Ultra low voltage operation of LEDs



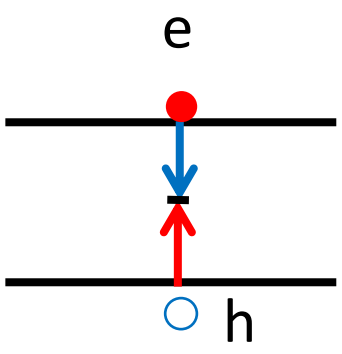


Turn on voltage (V_T) scales with Eg with an offset of about 0.8V

Simple model is not good enough..



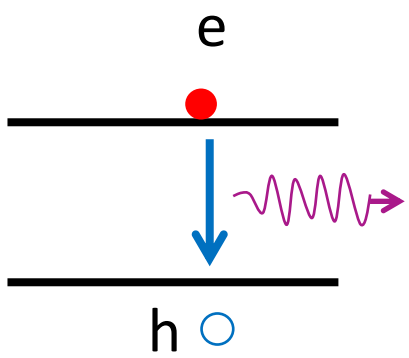
Trap assisted SRH



$k_1 n$

monomolecular

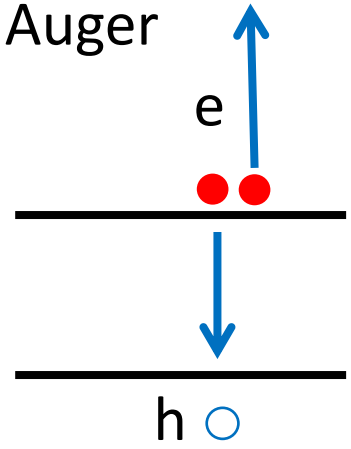
Radiative



$k_2 n^2$

bimolecular

Auger



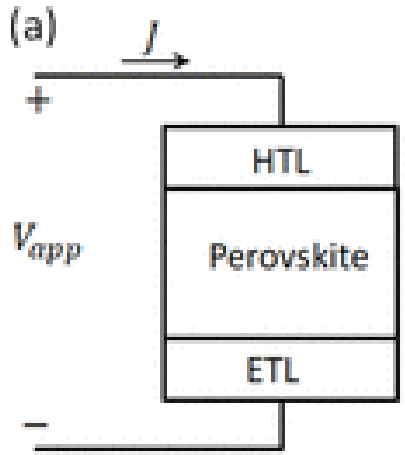
$k_3 n^3$

Auger

$$J = qW(k_1 n + k_2 n^2 + k_3 n^3)$$

$$EQE = \frac{k_2 n^2 W \times \eta_{OC}}{J/q}$$

Multi-Physics model for PeLEDs



- Carrier recombination
- Space charge effect
- Series and shunt resistances
- Joule heating and increase in temperature
- Temperature dependent carrier recombination

$$J_{\mathrm{rec}} = q(k_1 n + k_2 n^2 + k_3 n^3) W_{\mathrm{p}}$$

$$V_{\mathrm{D}} = \frac{2k_{\mathrm{B}}T}{q} \ln\left(\frac{n}{n_{\mathrm{i}}}\right)$$

$$V_{\mathrm{SC}} = K_{\mathrm{SC}} J_{\mathrm{rec}}^{\alpha}$$

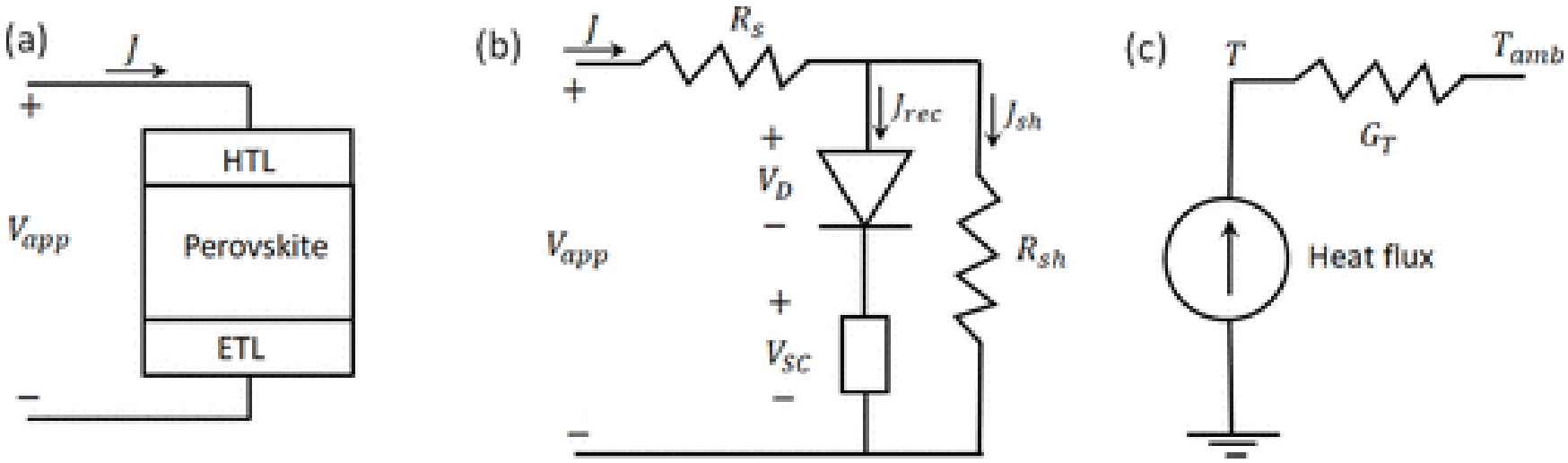
$$J = J_{\mathrm{rec}} + (V_{\mathrm{D}} + V_{\mathrm{SC}})/R_{\mathrm{sh}}$$

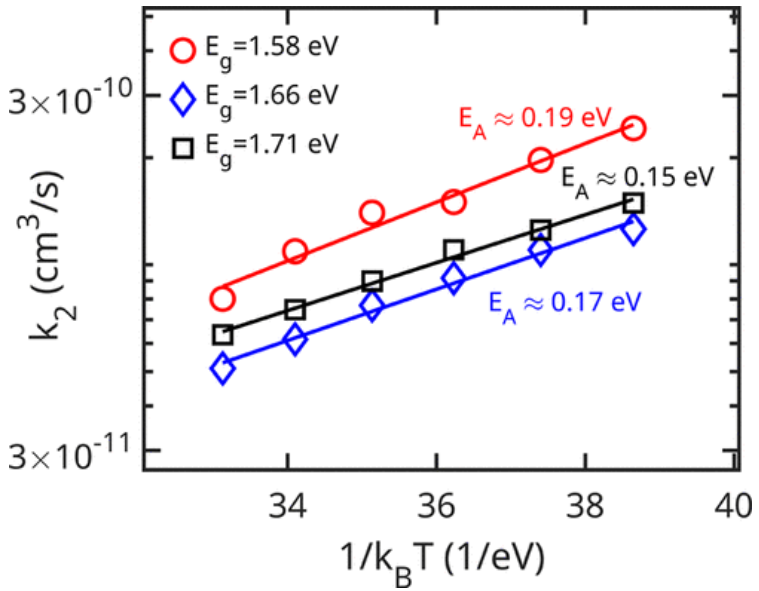
$$V_{\mathrm{app}} = V_{\mathrm{D}} + V_{\mathrm{SC}} + JR_{\mathrm{s}}$$

$$G_{\mathrm{T}}(T - T_{\mathrm{amb}}) = JV_{\mathrm{app}} - qk_2 n^2 W_{\mathrm{p}} E_{\mathrm{g}} \times \eta_{\mathrm{OC}}$$

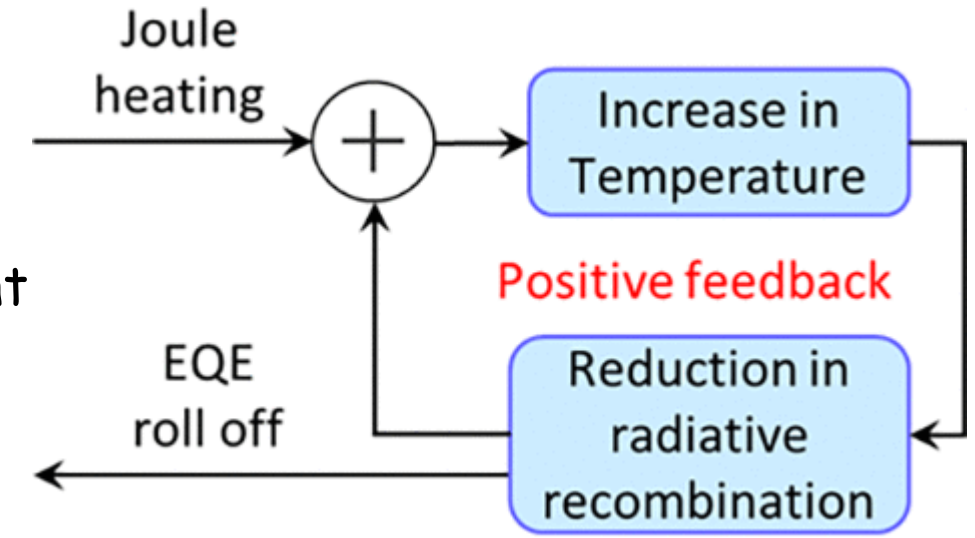
$$k_2 \propto T^{-3} \mathrm{e}^{E_{\mathrm{g}}/k_{\mathrm{B}}T} \int_0^{\infty} \kappa E^2 \mathrm{e}^{-E/k_{\mathrm{B}}T} \mathrm{d}E$$

Multi-Physics model for PeLEDs

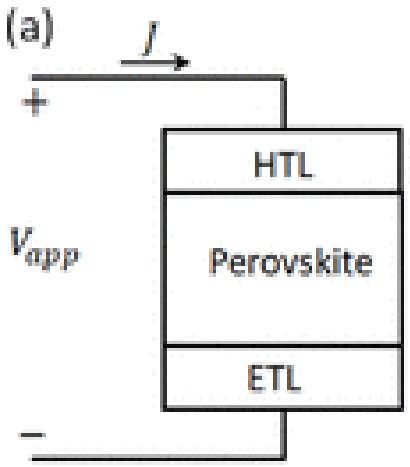


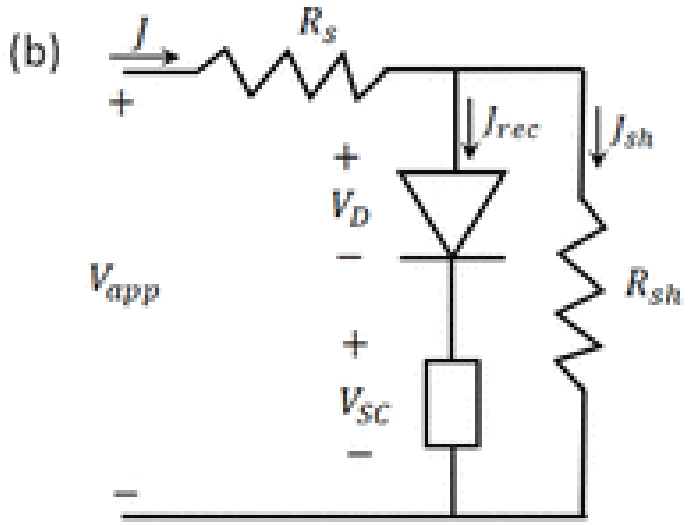


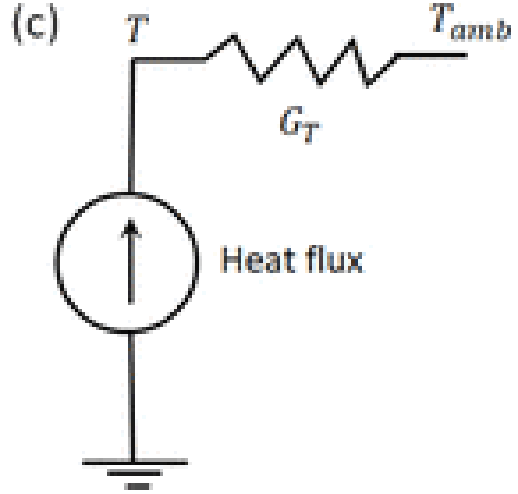
- Joule heating
- Space charge effect
- Thermal transport
- Temperature dependent carrier recombination

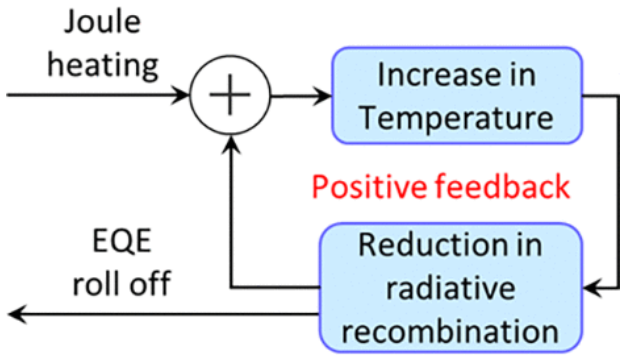


Validation of model

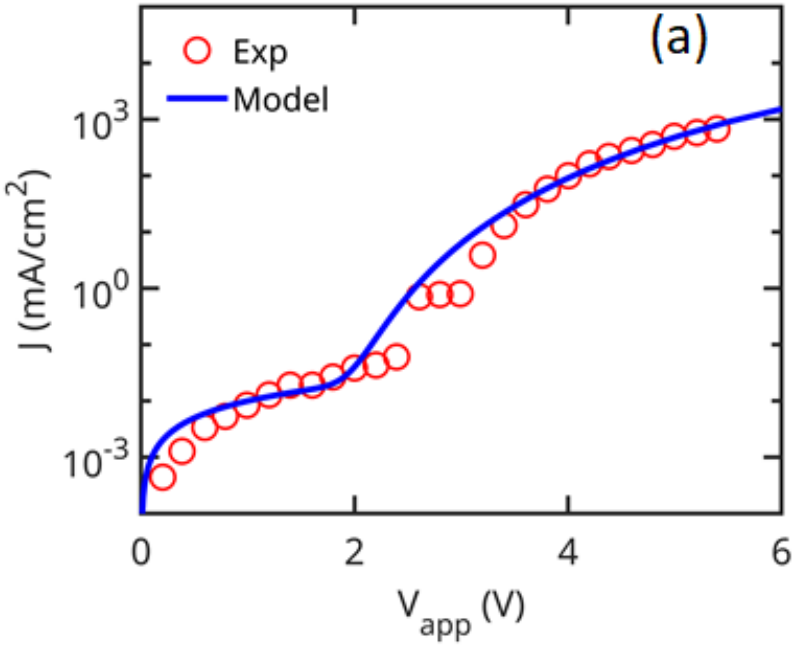


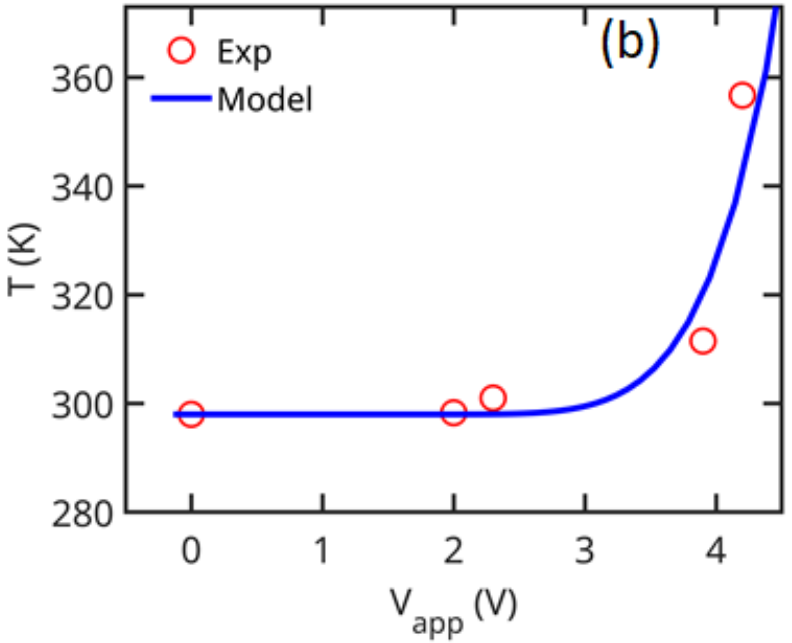


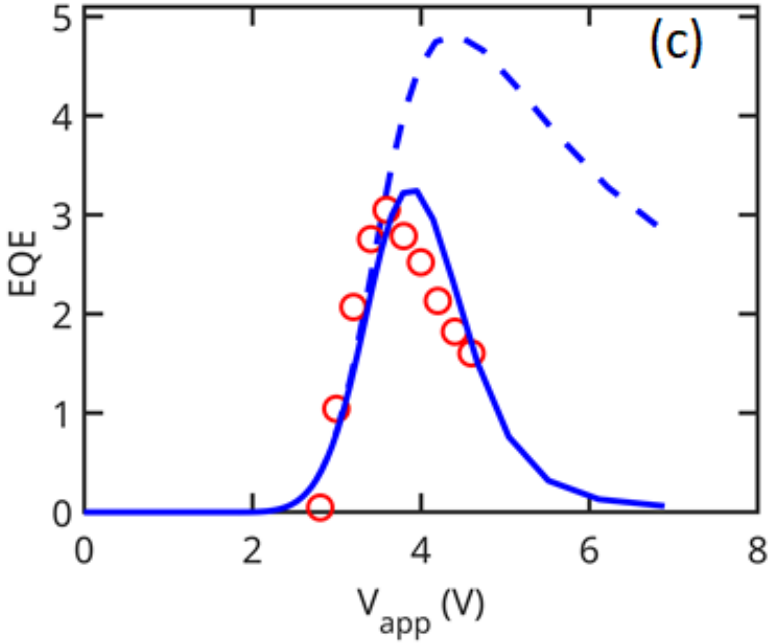




ACS AEM, 2025

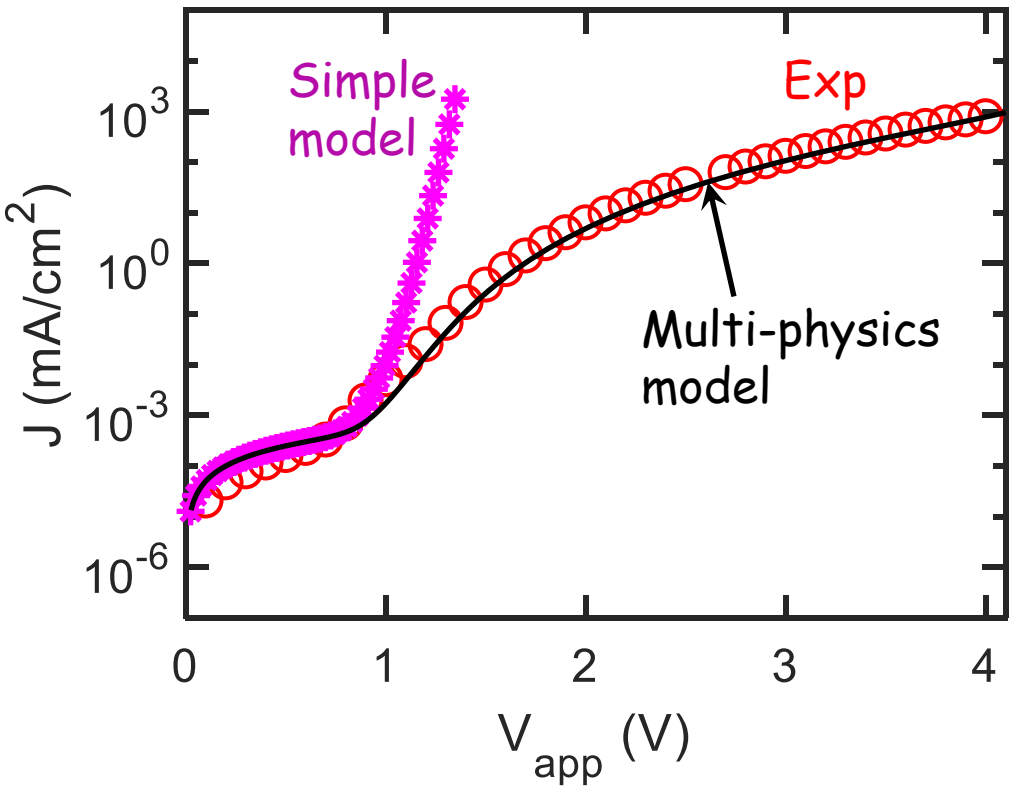


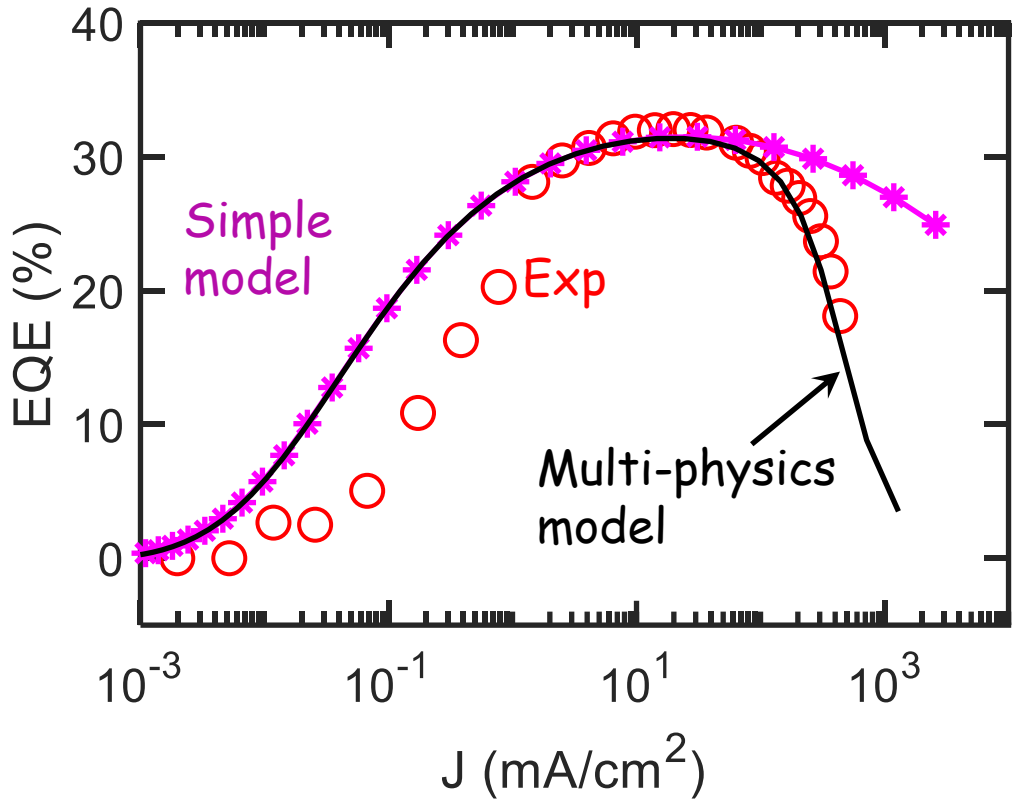




Interpretation of experiments (1)

Exp results from Li et al., Nature 2024

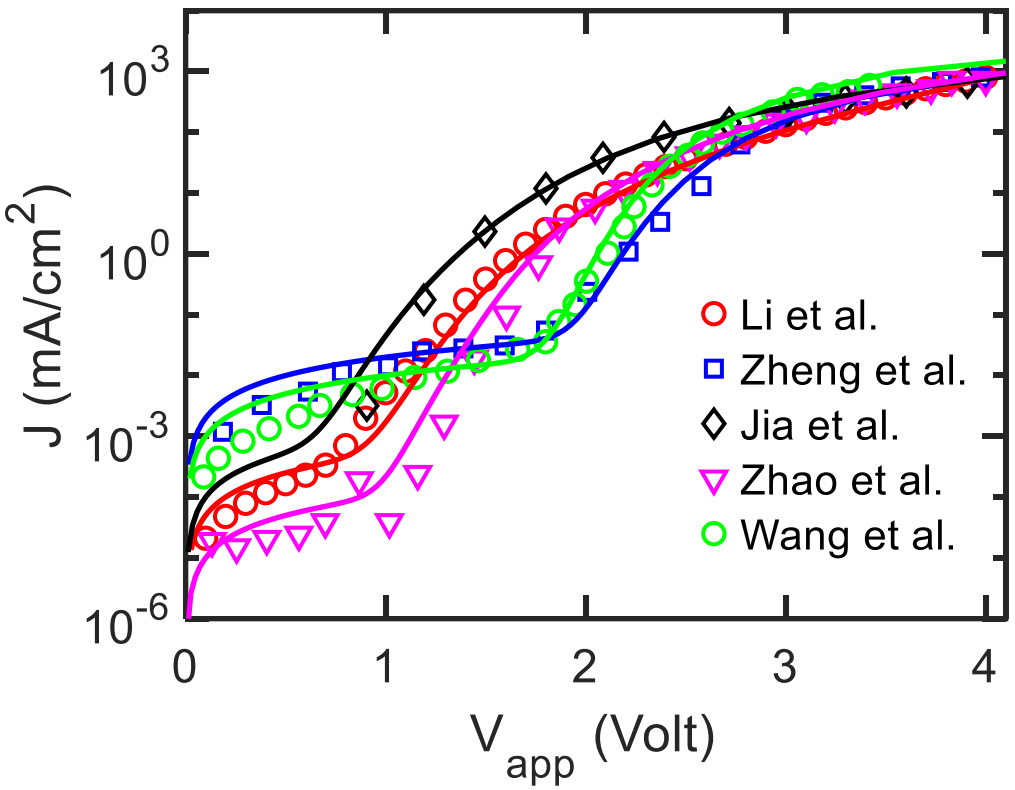


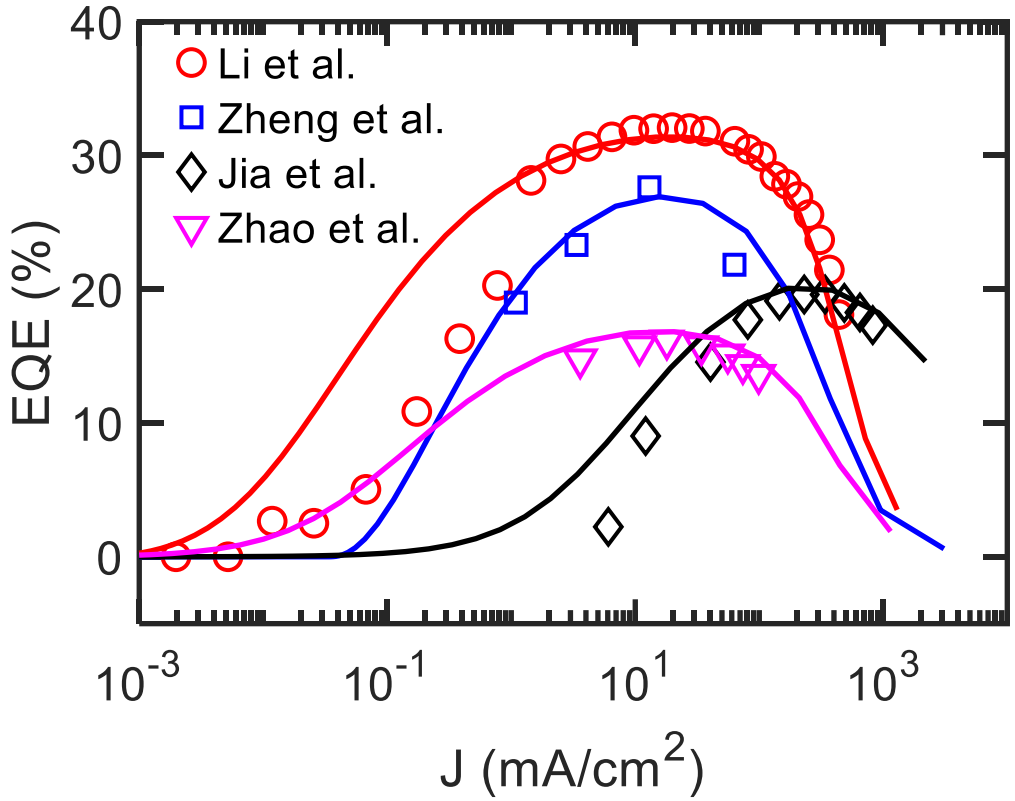


Space charge limited transport ➔ additional voltage

Positive feedback mechanism ➔ EQE droop

Interpretation of experiments (2)



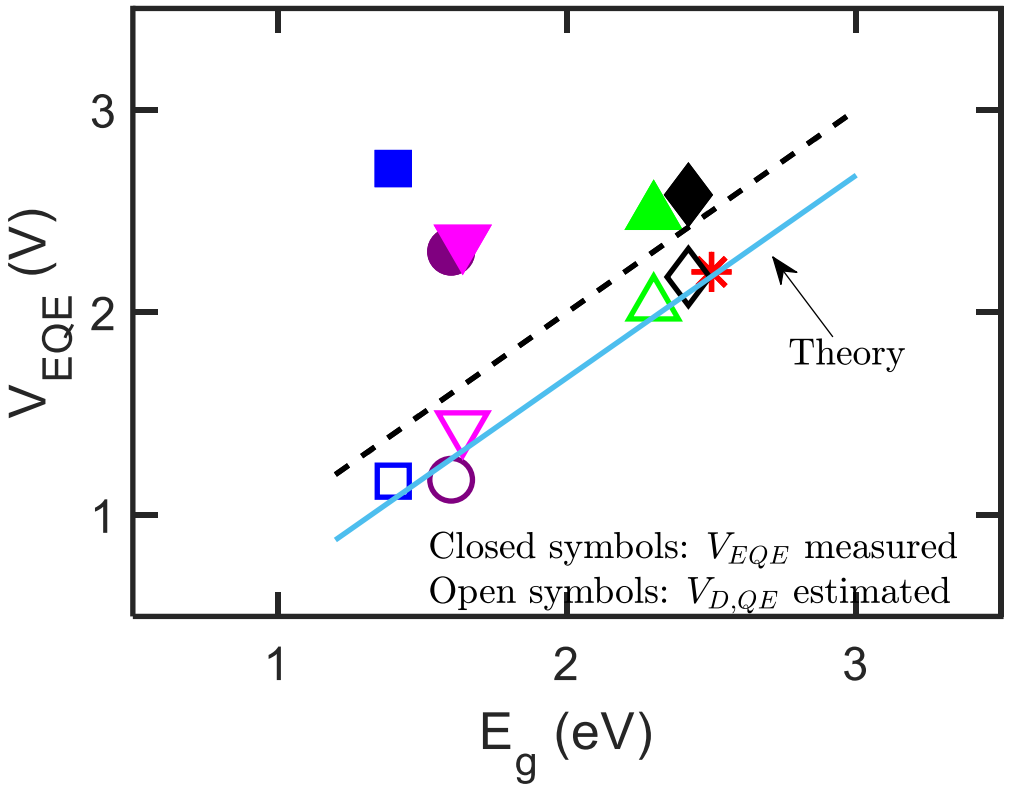


ACS AEM, 2025

Space charge limited transport ➔ additional voltage
Positive feedback mechanism ➔ EQE droop

Interpretation of experiments (3)

Voltage at maximum EQE

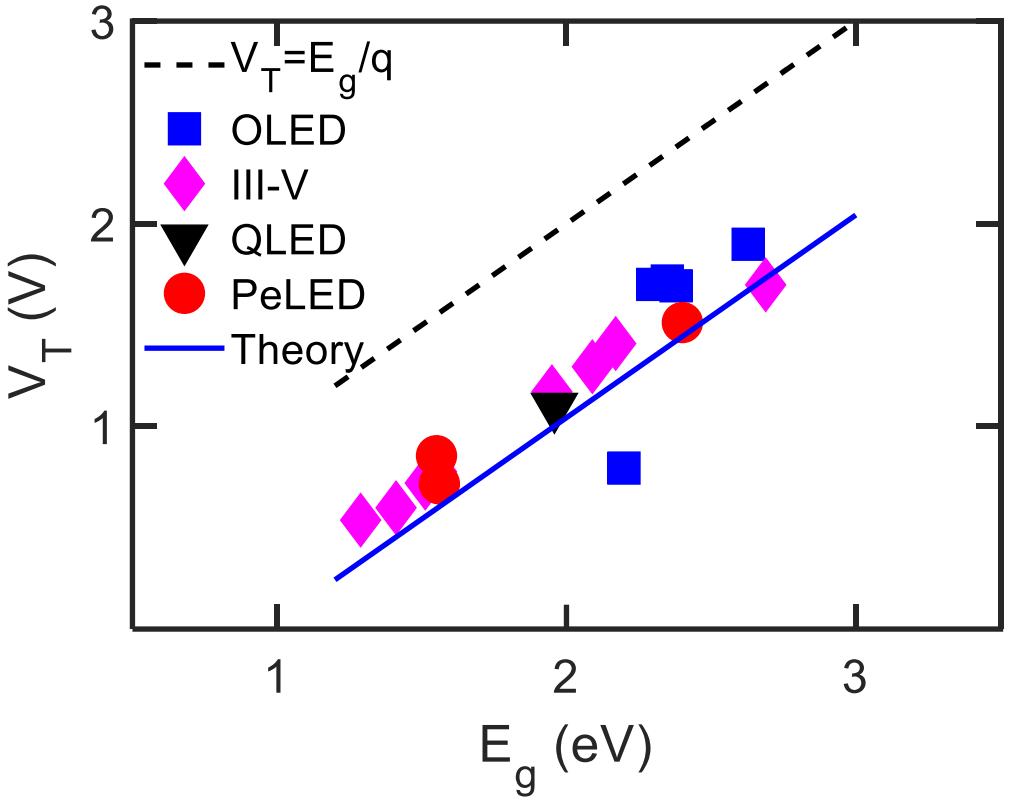


$$V_{D,QE} = \frac{E_g}{q} + \frac{k_B T}{q}\ln\left(\frac{k_1}{k_3}\right) - \frac{k_B T}{q}\ln(N_C N_V)$$

$$J_{QE} = q\frac{k_1}{k_3} \times (k_2 + 2\sqrt{k_1 k_3})W_P$$

$$V_{EQE} = V_{D,QE} + K_{SC} \times J_{QE}^{\alpha}$$

Turn ON Voltage



$$V_T = \frac{E_g}{q} + \frac{k_B T}{q}\ln\left(\frac{N_T}{k_2 N_C N_V W_P \times \eta_{OC}}\right)$$

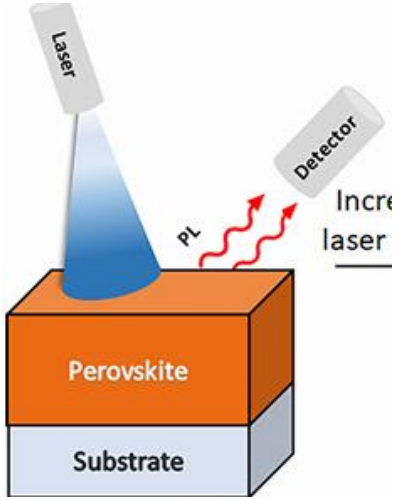
ACS Energy Letters, 2026

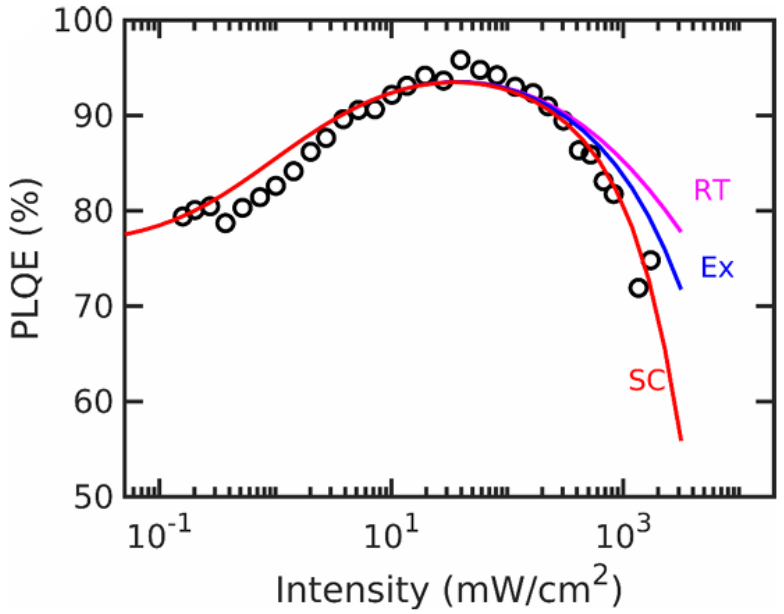
Conclusions

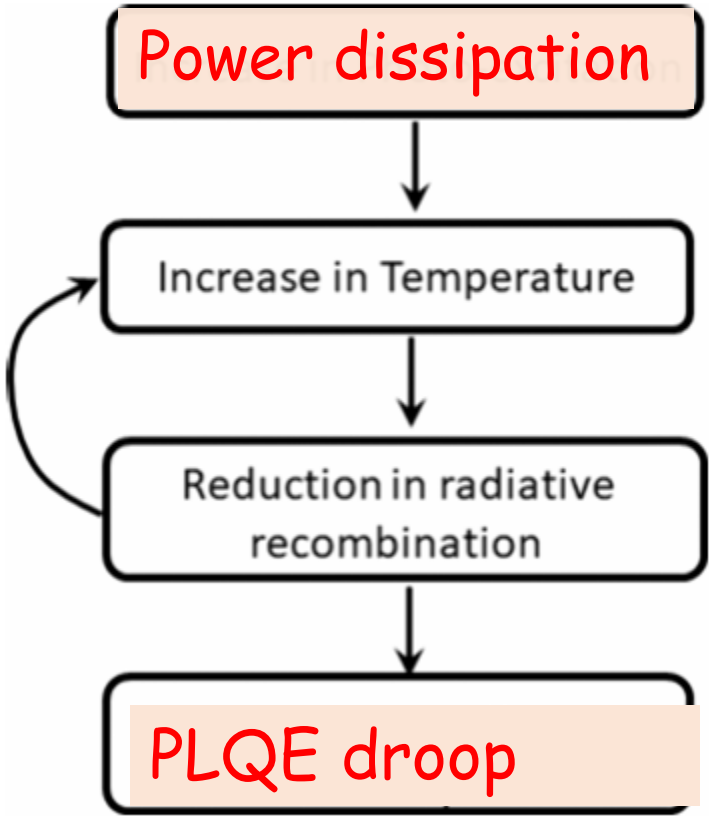
- Multi-probe analysis (PL, JV, EQE, etc.) leads to quantitative insights
- Space charge effects cause undesired power dissipation
- Better electrical and thermal design is essential
- Sub-Eg, high efficiency operation of PeLEDs.
- Model could be extended to understand stability and lifetime

Thank You

PLQE Roll-off : Positive feedback mechanism







- Joule heating
- Space charge effect
- Thermal transport
- Temperature dependent carrier recombination

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