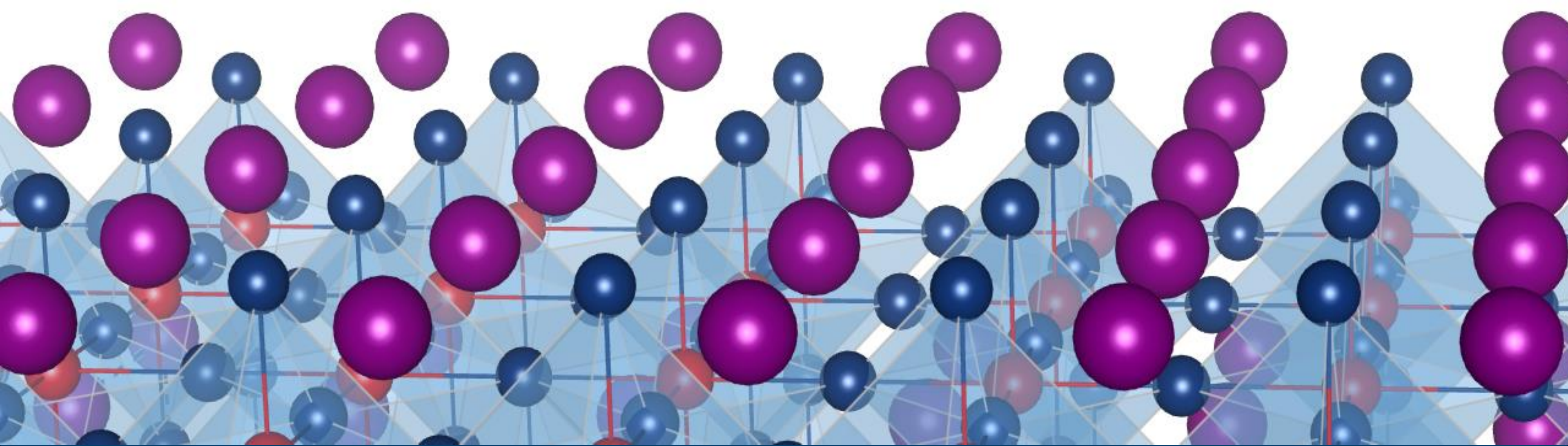
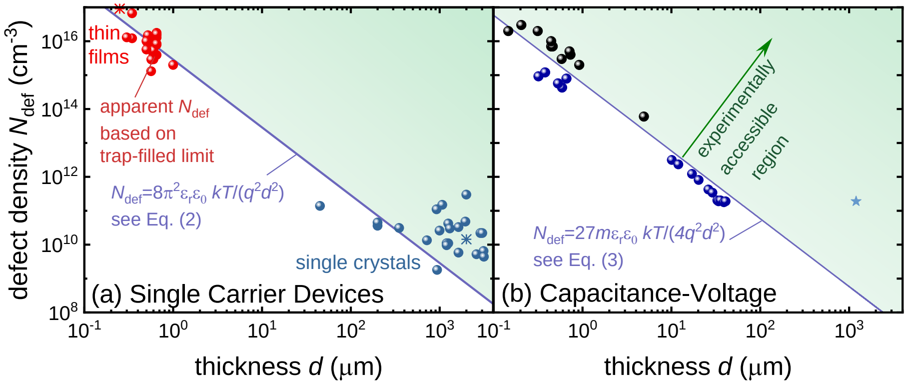


DEFECT DENSITIES AND CHARGE CARRIER LIFETIMES IN LEAD-HALIDE PEROVSKITES

Thomas Kirchartz

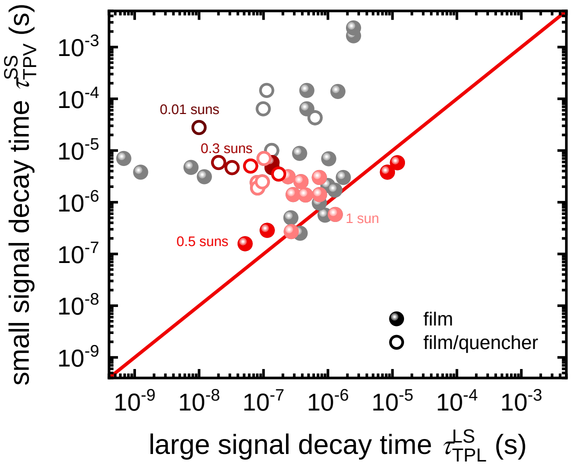
IEK-5 Photovoltaik, Forschungszentrum Jülich

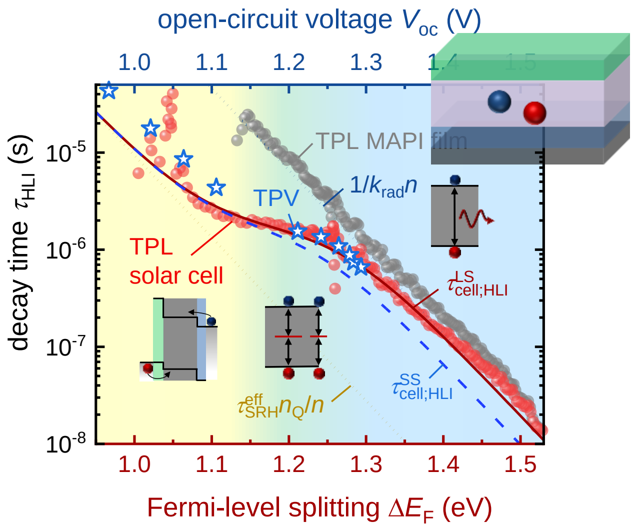
Outline



1) Defect densities (and how they change with thickness)

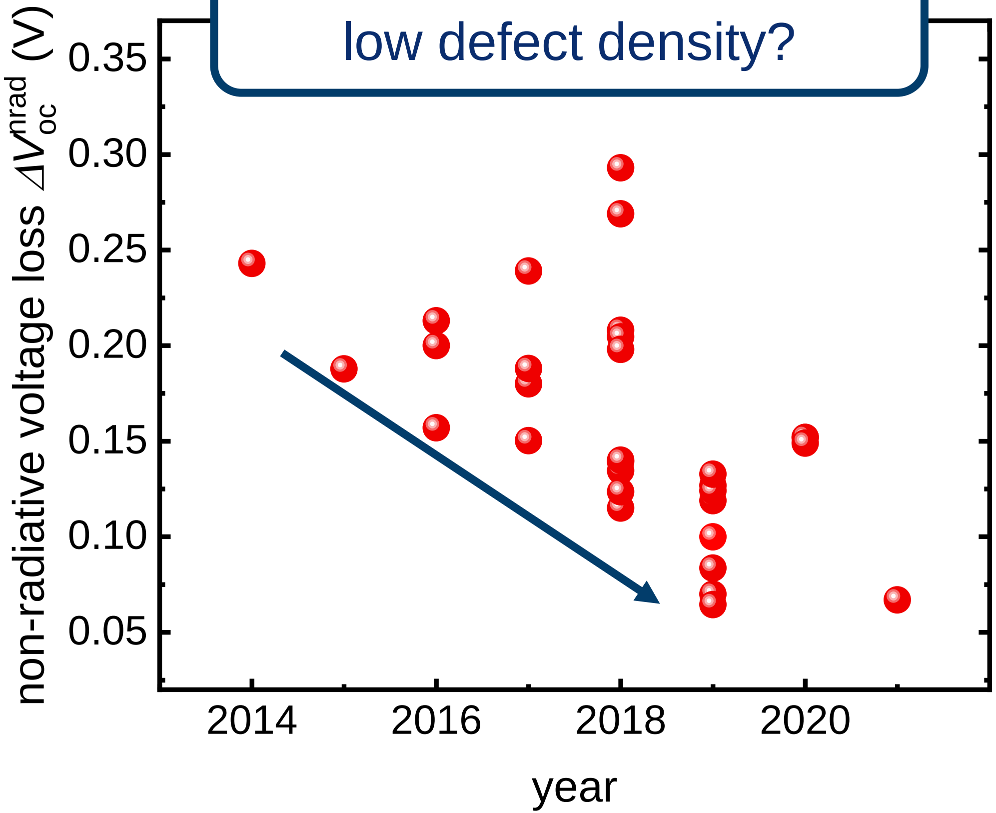
2) “Lifetimes” and how they change with carrier density

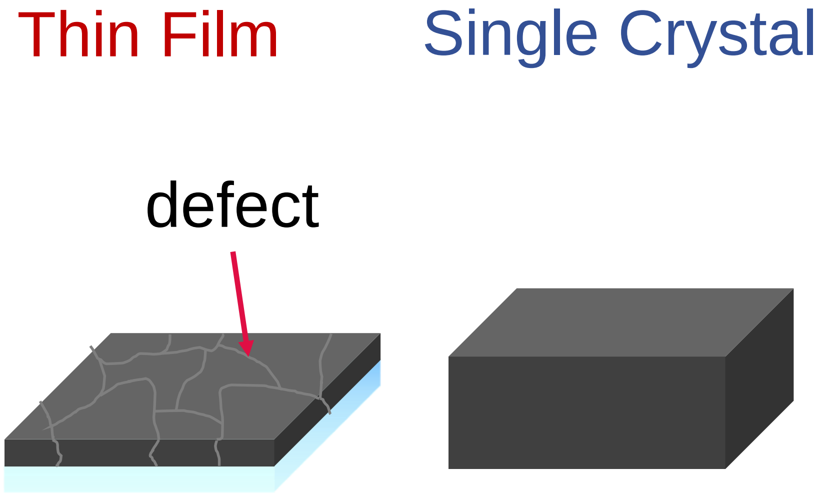




Reduced Non-Radiative Recombination

small non-radiative loss → low defect density?

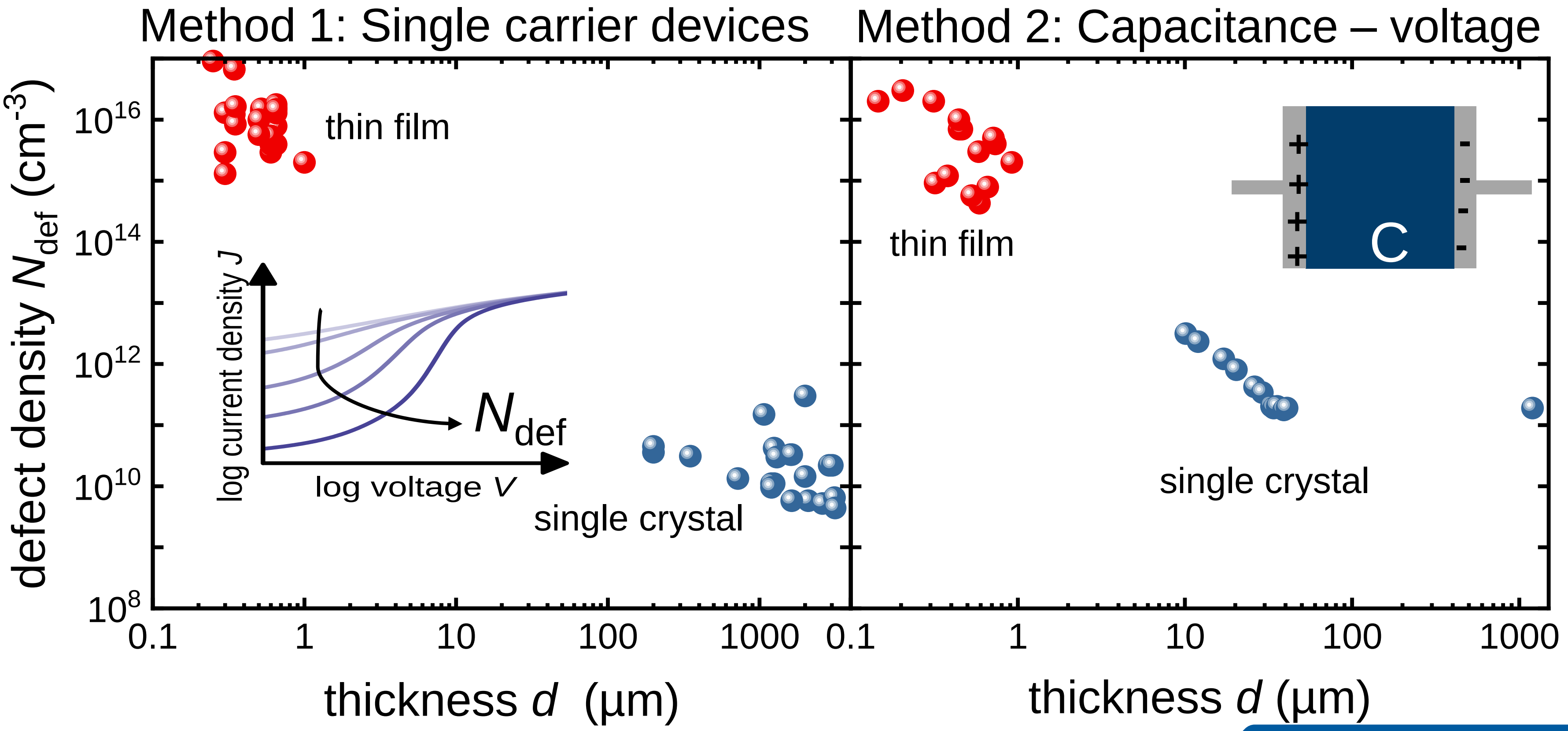




single crystals could have an even lower defect-density

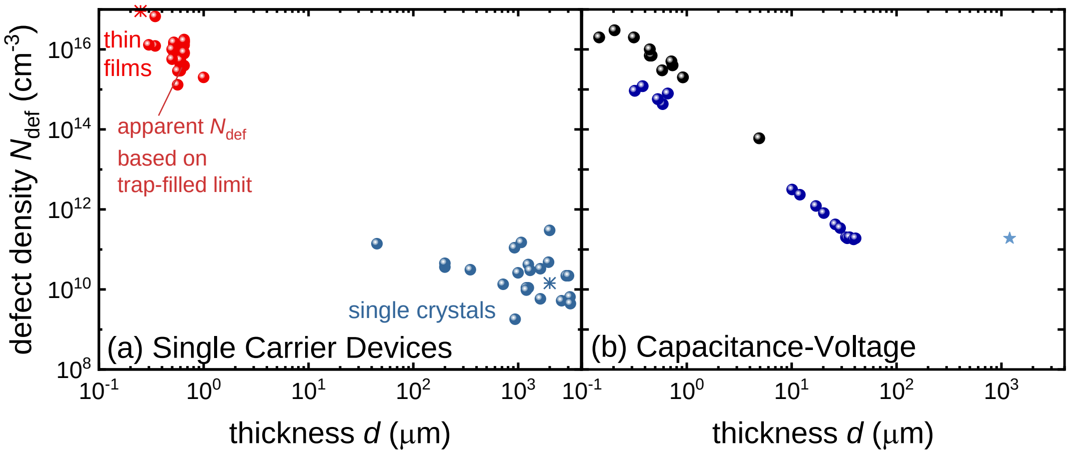
Apparent Defect Density

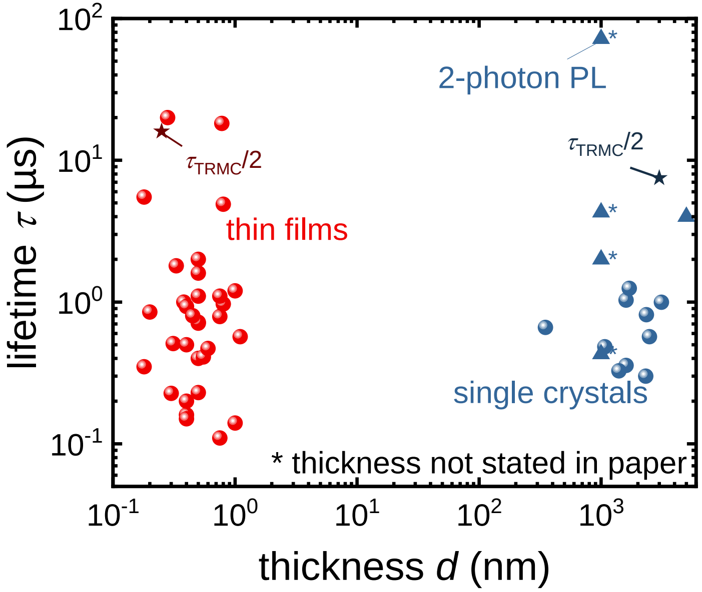
Charge sensitive measurements:



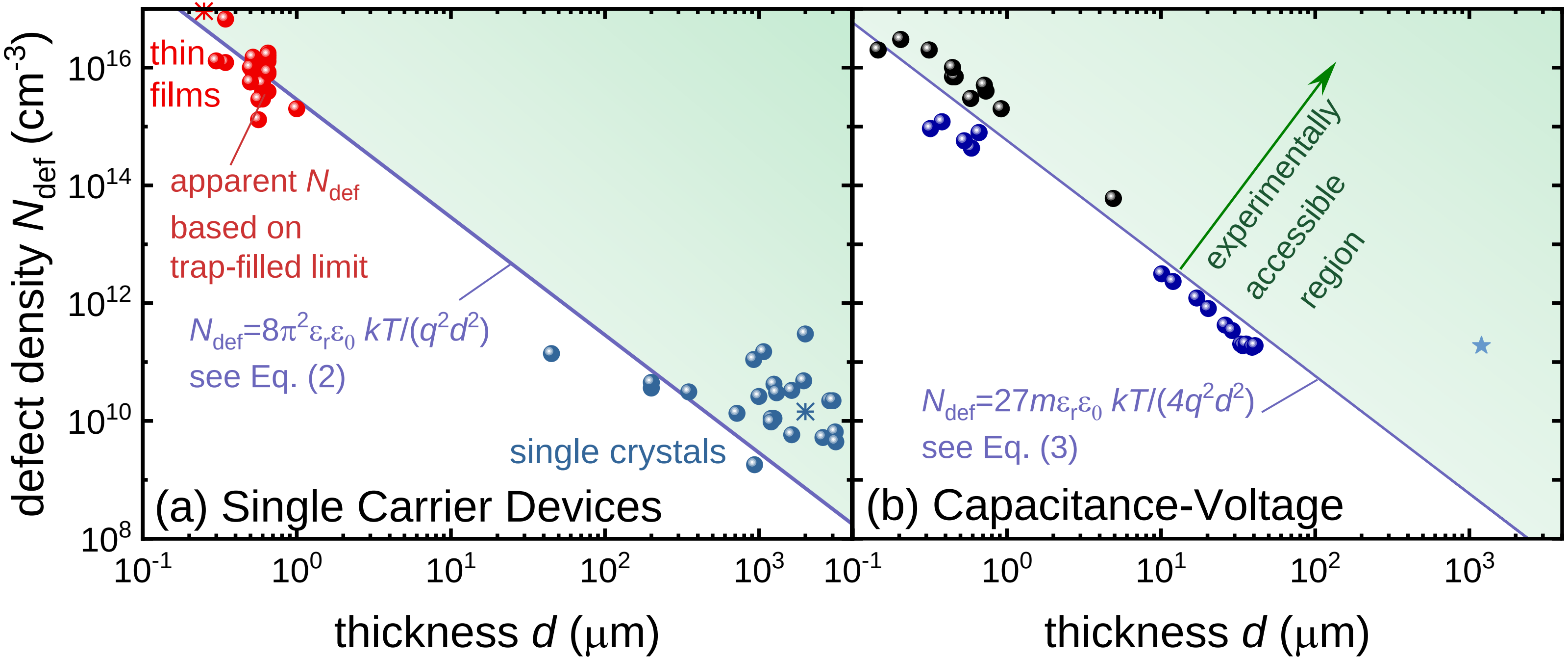
Single crystals much better than thin films

Metaanalysis of trap densities





Sensitivity thresholds for measuring charge densities



Ravishankar et al. Science 371, 6532 (2021), Siekmann et al. ACS Energy Lett. 6, 3244 (2021)

Surface charges compete with the trap charges

The thickness trend explained.

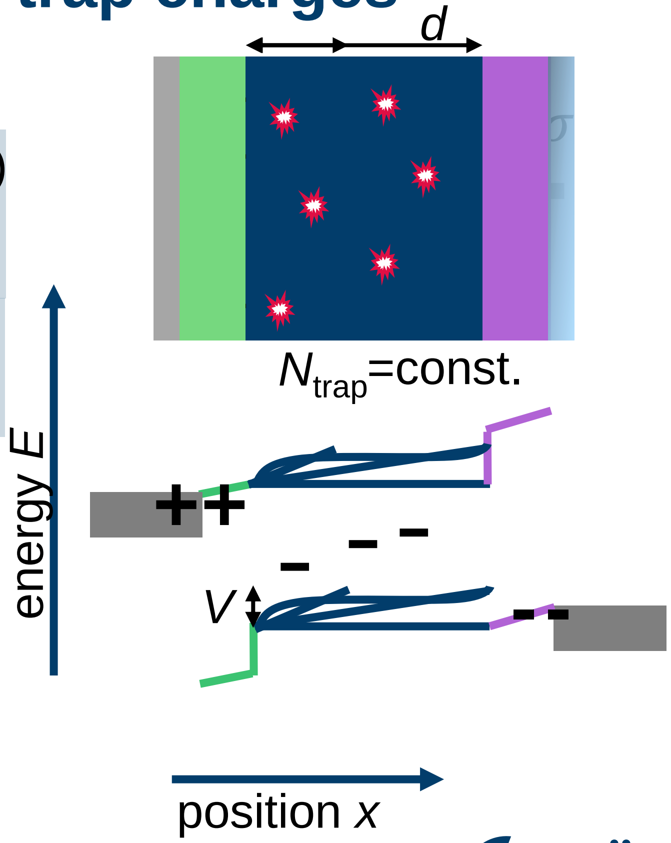
surface charge (C/cm²) | volume charge (C/cm³)

$$\sigma \ll d q N_{\text{trap}}$$

$$\sigma = \frac{\varepsilon V}{d} \ll d q N_{\text{trap}}$$

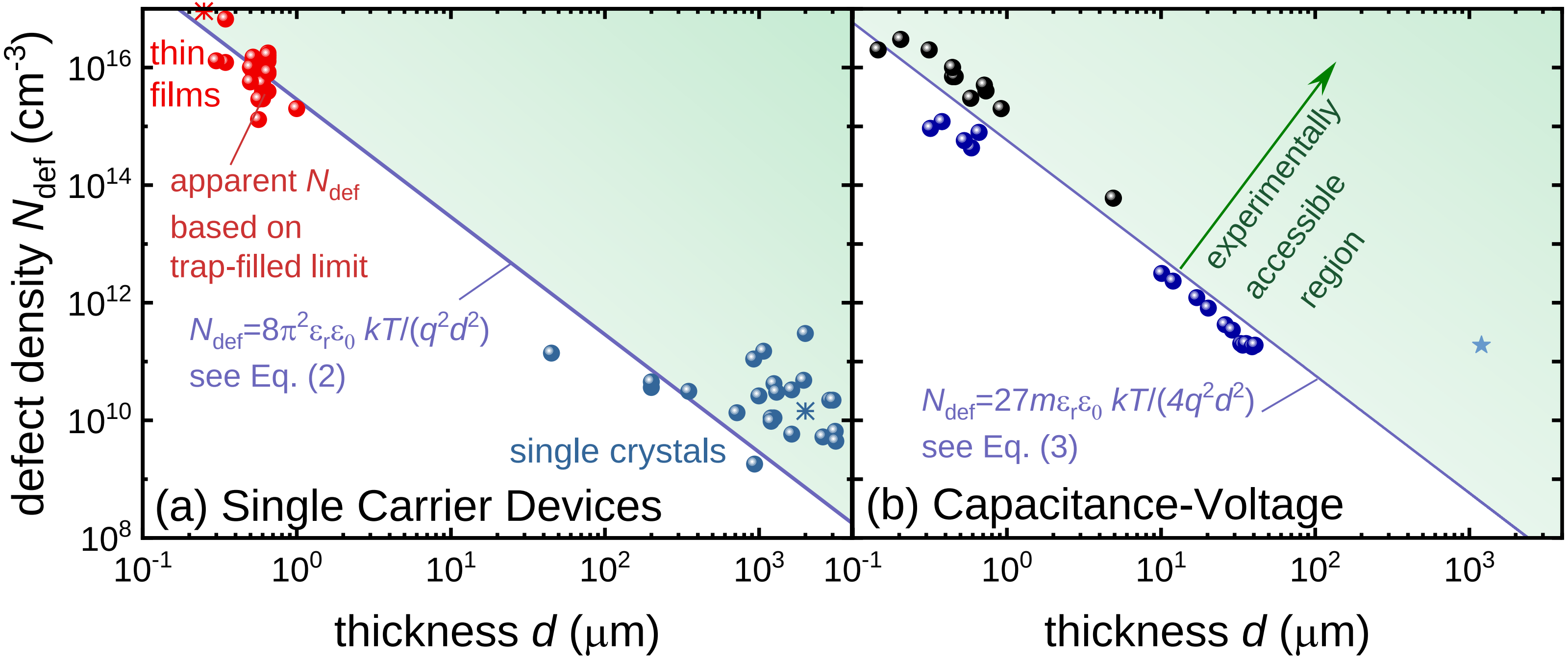
$$\rightarrow \frac{\varepsilon V}{q d^2} \ll N_{\text{trap}}$$

→ higher N_{trap} needed for thinner films



σ: surface charge density
ε: permitivity
q: elementary charge
d: thickness

Sensitivity thresholds for measuring charge densities



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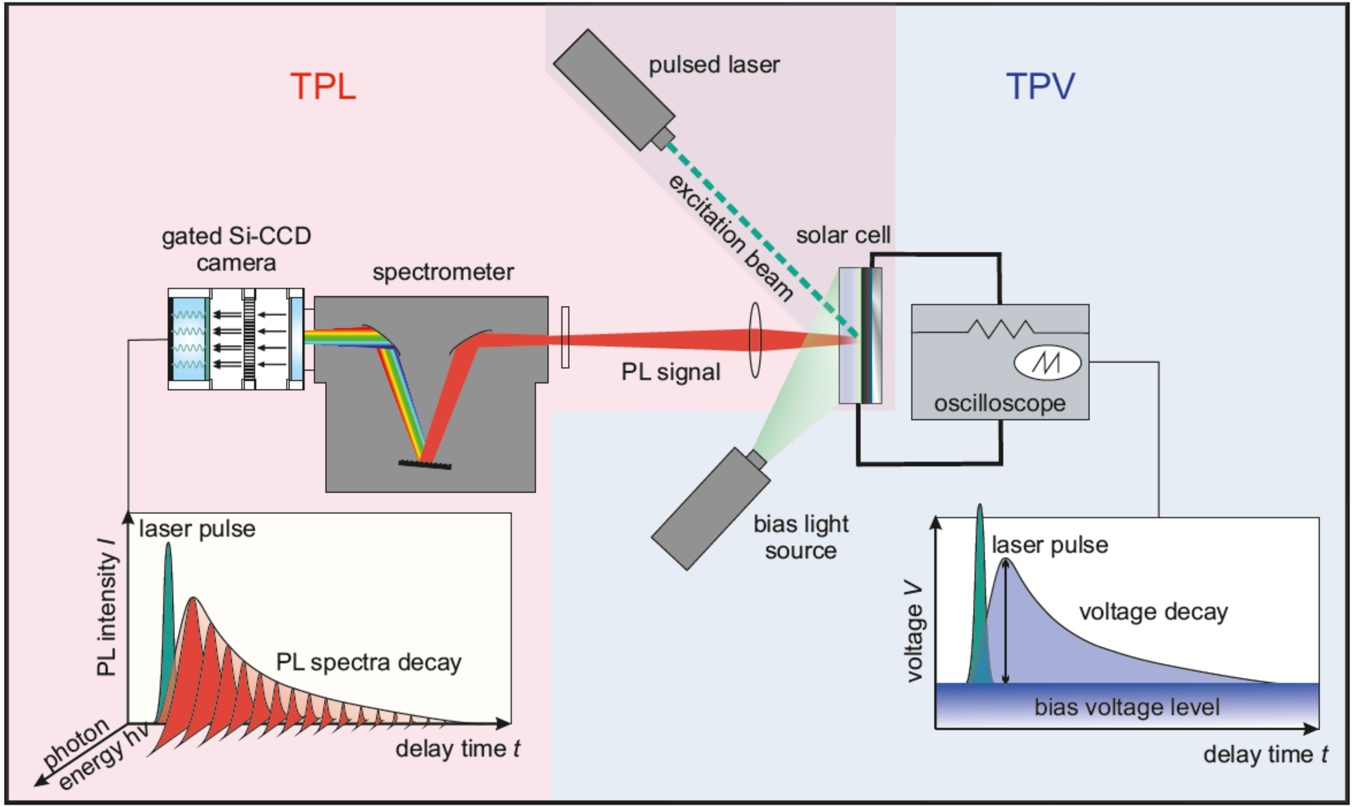
Transient Photoluminescence (TPL)

Transient Photovoltage (TPV)

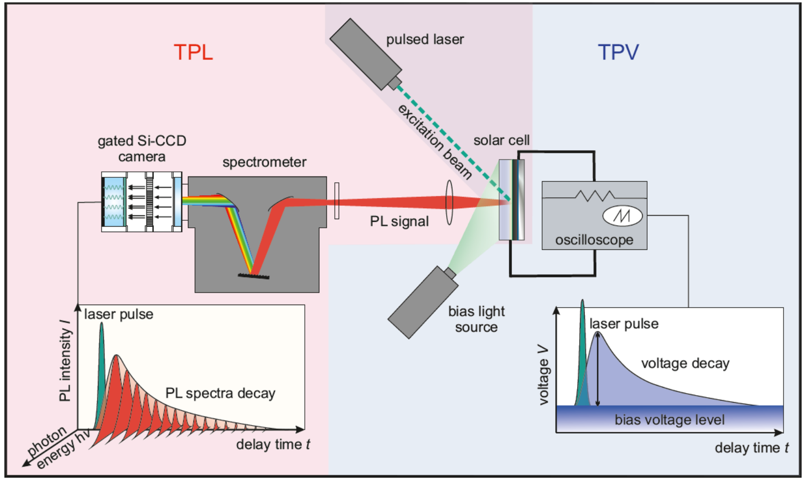
measures the luminescence decay

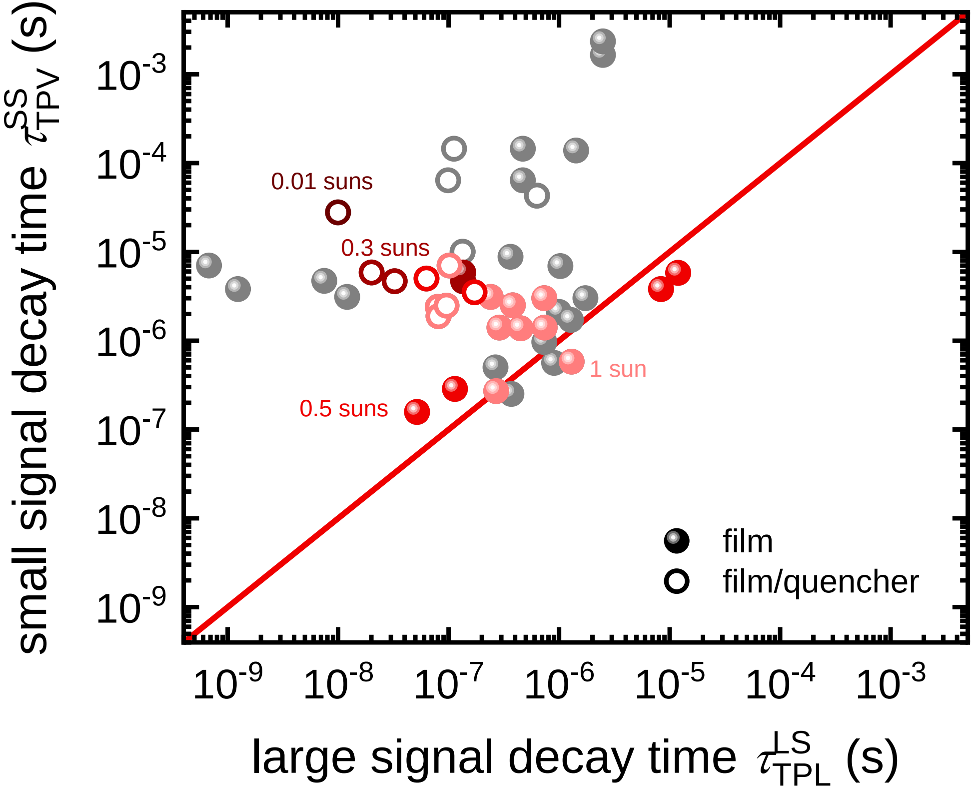
measures the decay of an external voltage



Metaanalysis: TPV vs. TPL

Fairly poor correlation between the two decay times. Why?





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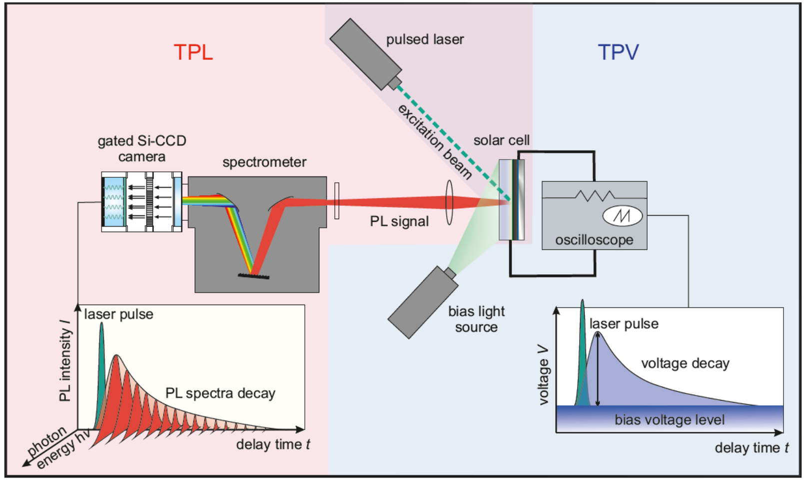
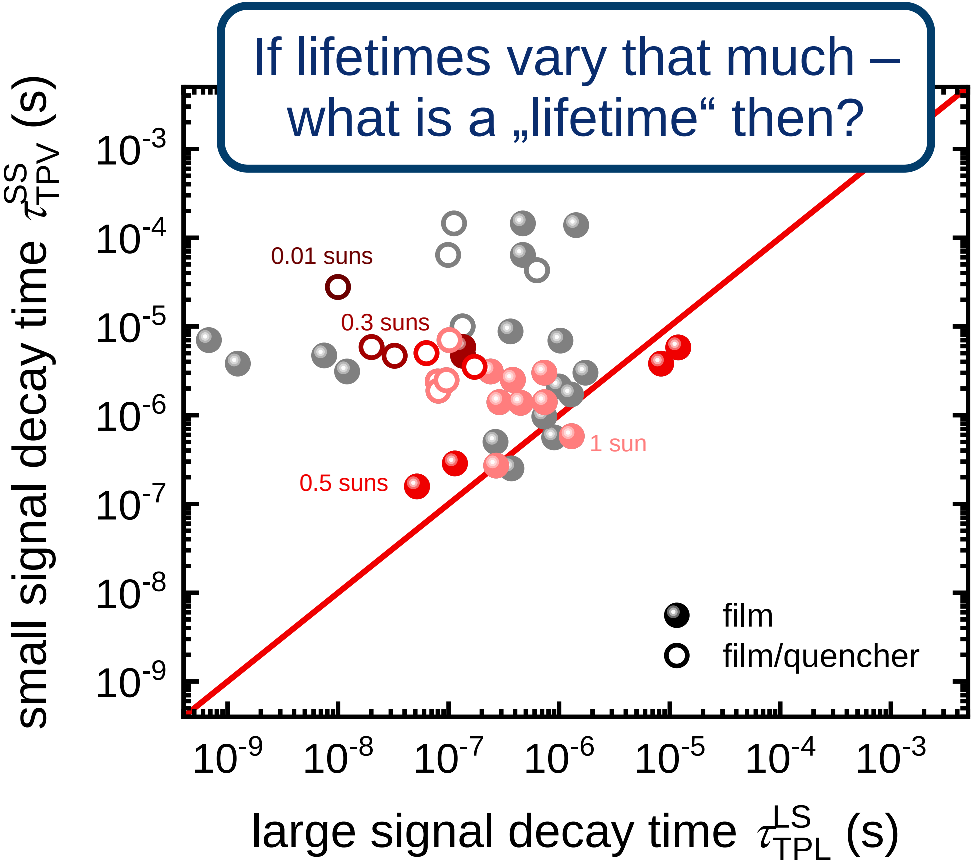
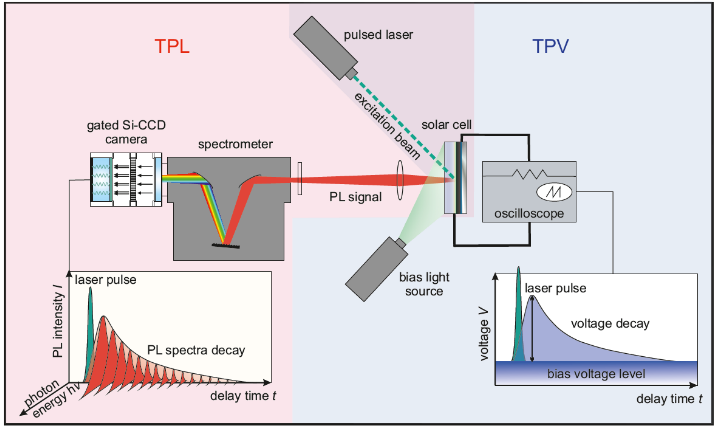
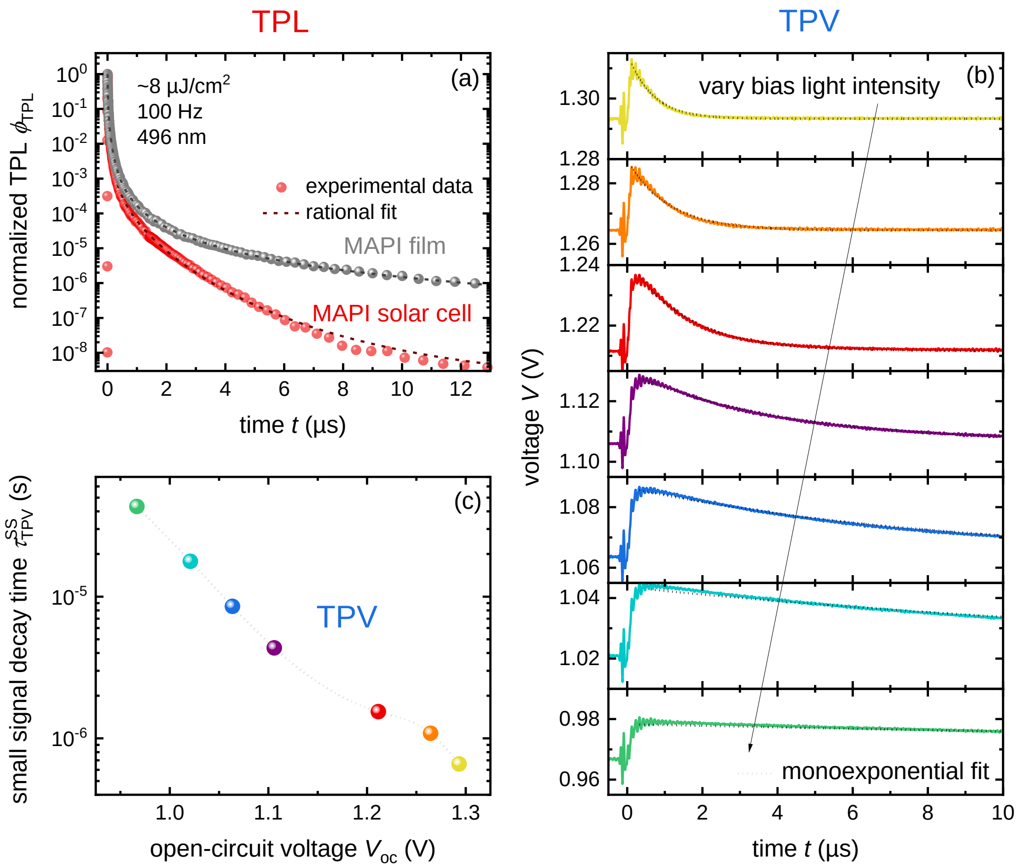


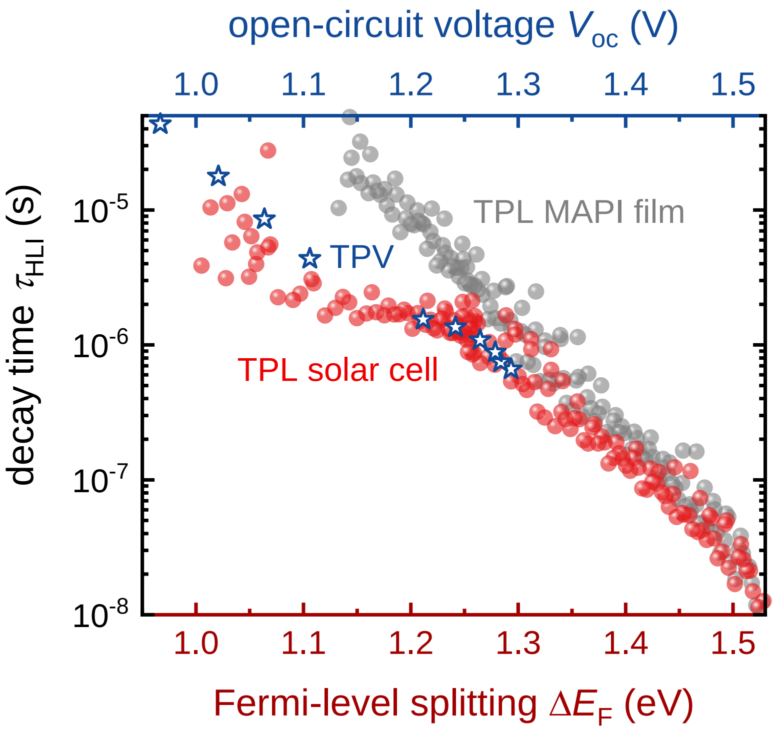


What is typically measured?





How to make sense out of it

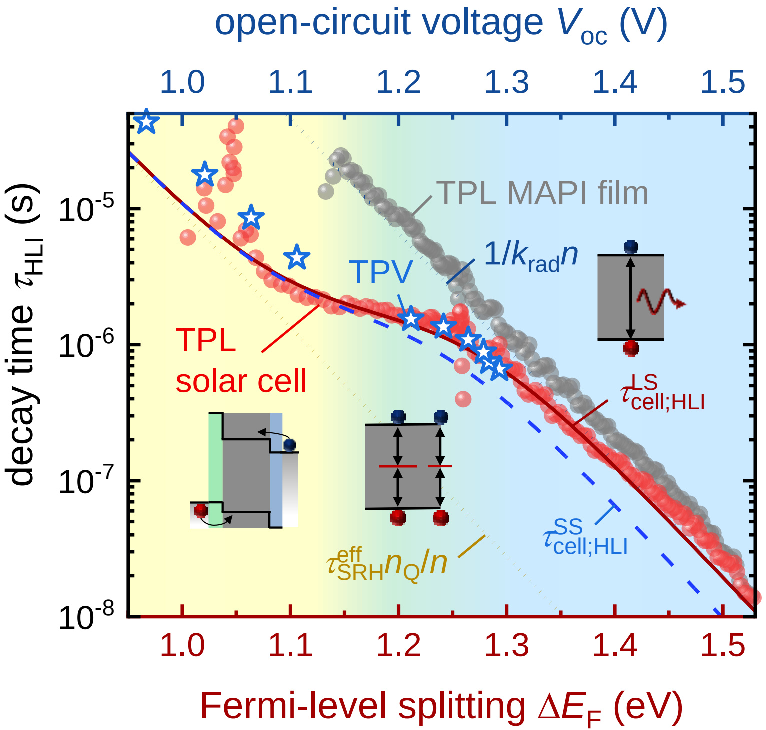


$$\frac{dn(t)}{dt} = -k_{rad} n(t)^2 - \frac{n(t)}{\tau_{SRH}^{bulk}} - \frac{C_{area}}{q d_{pero}} \frac{dV_{ext}(t)}{dt} + G$$

$$\tau_{cell,\ HLI}^{LS} = -\frac{n(t)}{dn(t)/dt} = \frac{n_Q/n(t)+1}{k_{rad} n(t) + 1/\tau_{SRH}^{eff}}$$

$$n_Q = 2 C_{area} k_B T / \left(q^2 d_{pero}\right)$$

How to make sense out of it



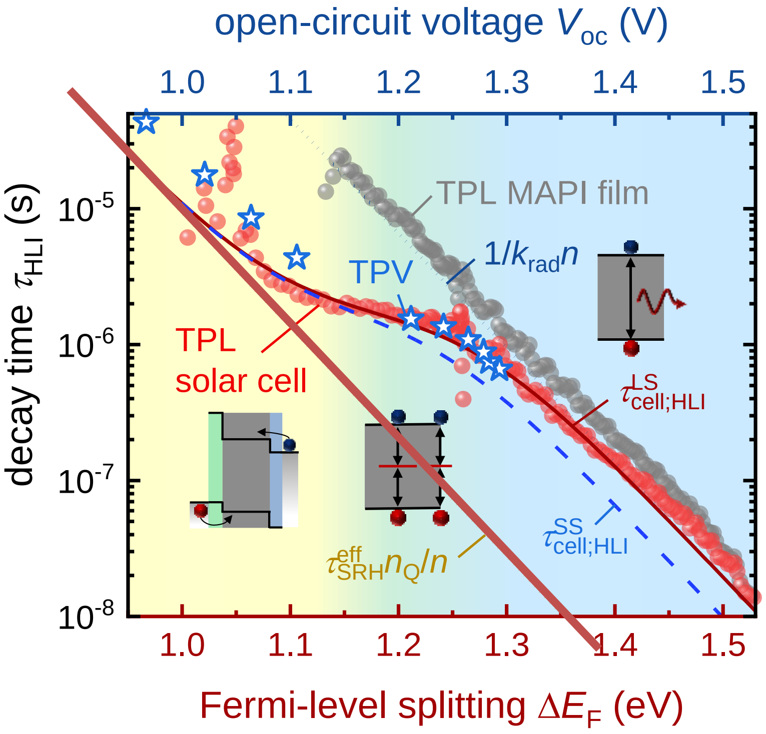
Capacitive charging of the electrodes

$$\frac{dn(t)}{dt} = -k_{rad} n(t)^2 - \frac{n(t)}{\tau_{SRH}^{bulk}} - \frac{C_{area}}{qd_{pero}} \frac{dV_{ext}(t)}{dt} + G$$

$$\tau_{cell,\,HLI}^{LS} = -\frac{n(t)}{dn(t)/dt} = \frac{n_Q/n(t)+1}{k_{rad}n(t)+1/\tau_{SRH}^{eff}}$$

$$n_Q = 2C_{area}k_BT/\left(q^2 d_{pero}\right)$$

How to make sense out of it



$$\frac{dn(t)}{dt} = -k_{rad} n(t)^2 - \frac{n(t)}{\tau_{SRH}^{bulk}} - \frac{C_{area}}{q d_{pero}} \frac{dV_{ext}(t)}{dt} + G$$

$$\tau_{cell,\,HLI}^{LS} = -\frac{n(t)}{dn(t)/dt} = \frac{n_Q / n(t) + 1}{k_{rad} n(t) + 1/\tau_{SRH}^{eff}}$$

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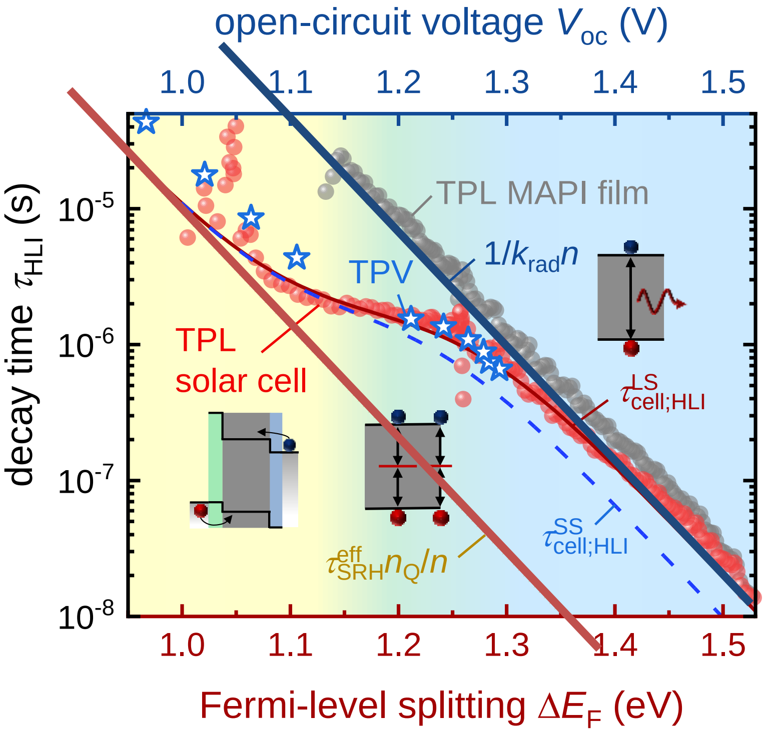

$$\frac{dn(t)}{dt} = -k_{rad}n(t)^2 - \frac{n(t)}{\tau^{bulk}_{SRH}} - \frac{C_{area}}{qd_{pero}}\frac{dV_{ext}(t)}{dt} + G$$

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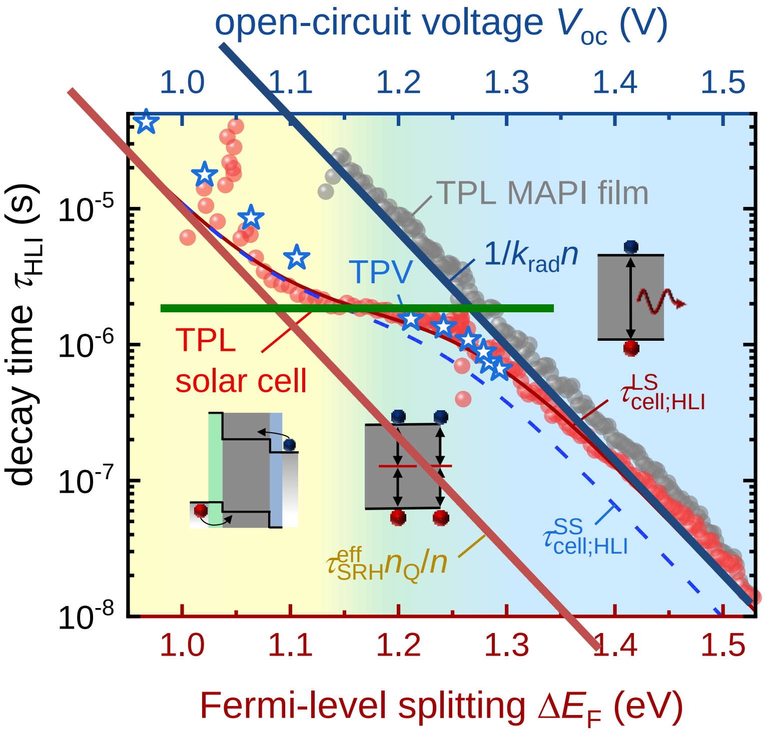
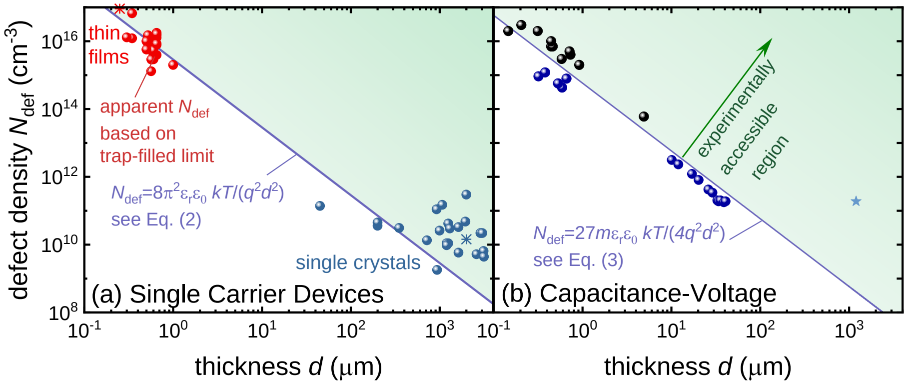


$$\frac{dn(t)}{dt} = -k_{rad}n(t)^2 - \frac{n(t)}{\tau_{SRH}^{bulk}} - \frac{C_{area}}{qd_{pero}}\frac{dV_{ext}(t)}{dt} + G$$

$$\tau^{LS}_{cell, HLI} = -\frac{n(t)}{dn(t)/dt} = \frac{n_Q/n(t)+1}{k_{rad}n(t)+1/\tau^{eff}_{SRH}}$$

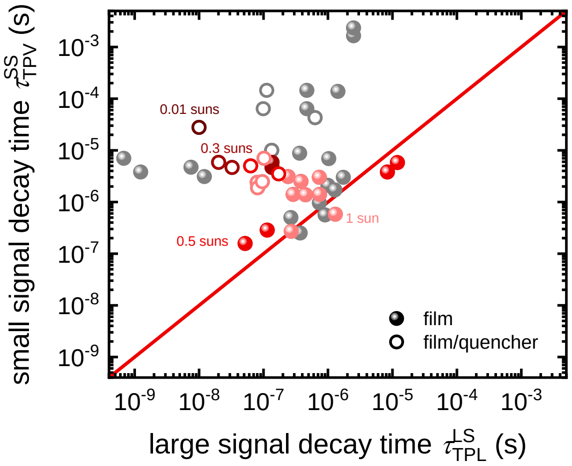
$$n_Q = 2C_{area}k_BT/\left(q^2d_{pero}\right)$$

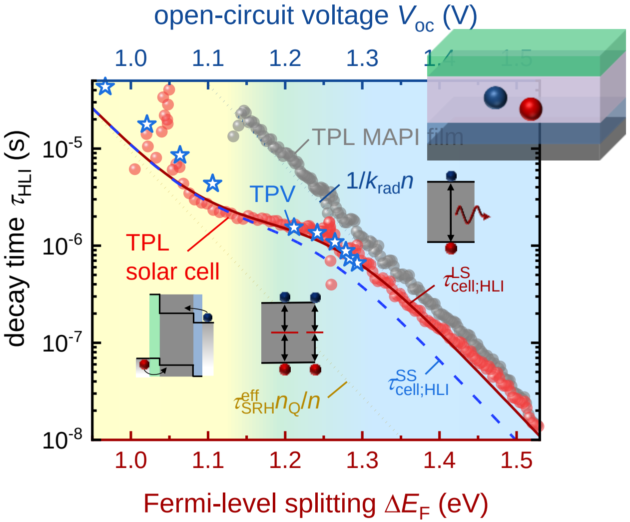
Summary



1) **Beware of detection thresholds, when measuring defect densities**

2) **Huge method to method variation in "lifetimes" results from voltage dependence of decay times**





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Thank you for your attention

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