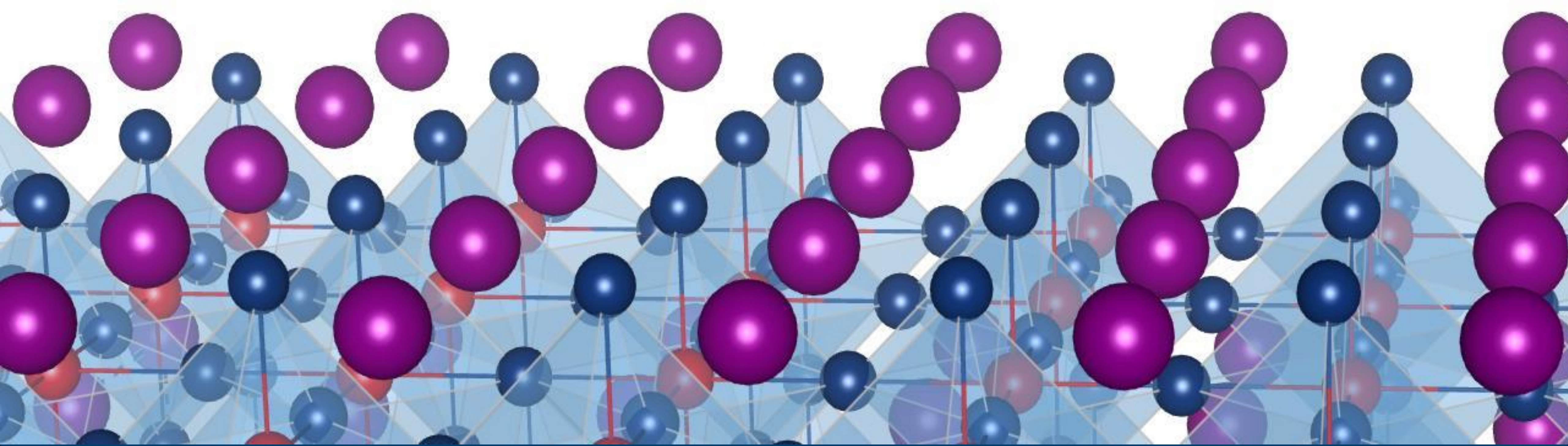


Disorder and Recombination in Organic Solar Cells

Thomas Kirchartz[1], Paula Hartnagel[1], Basita Das[2]

[1]IEK-5 Photovoltaik, Forschungszentrum Jülich

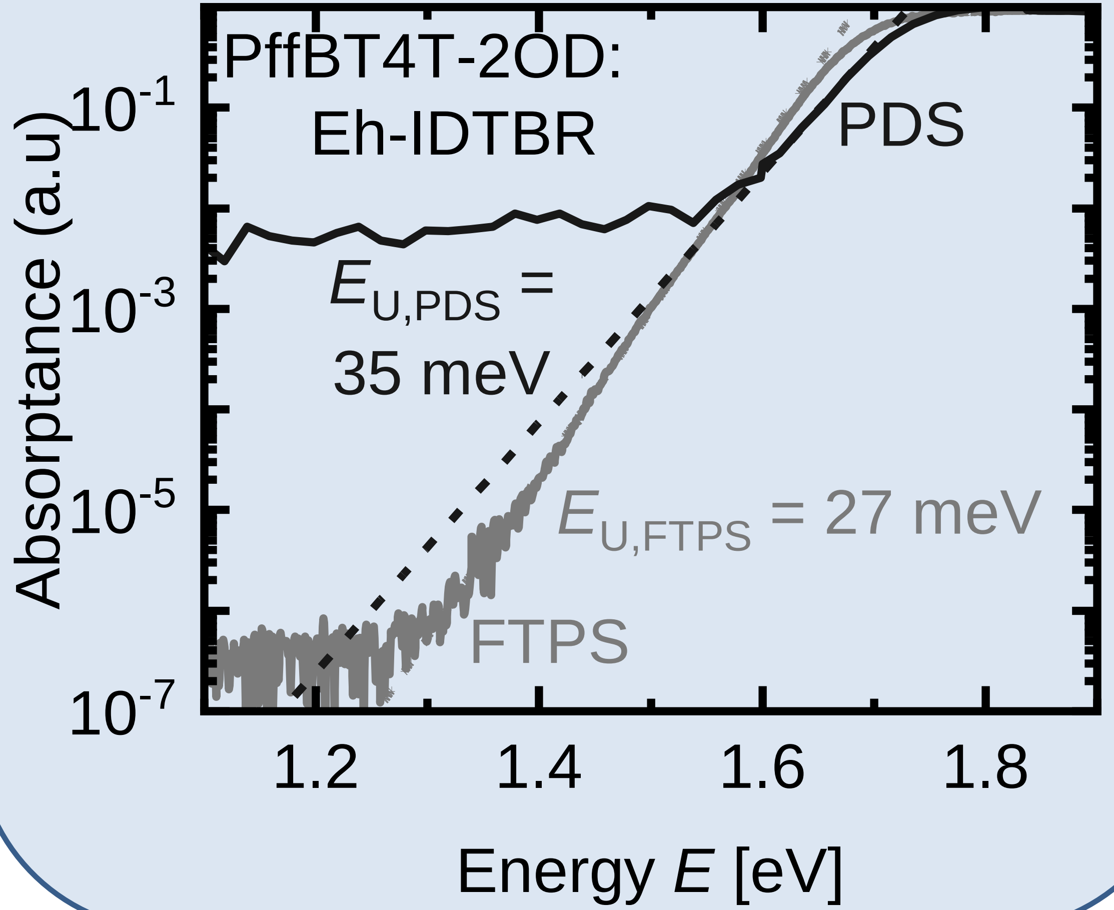
[2]MIT

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

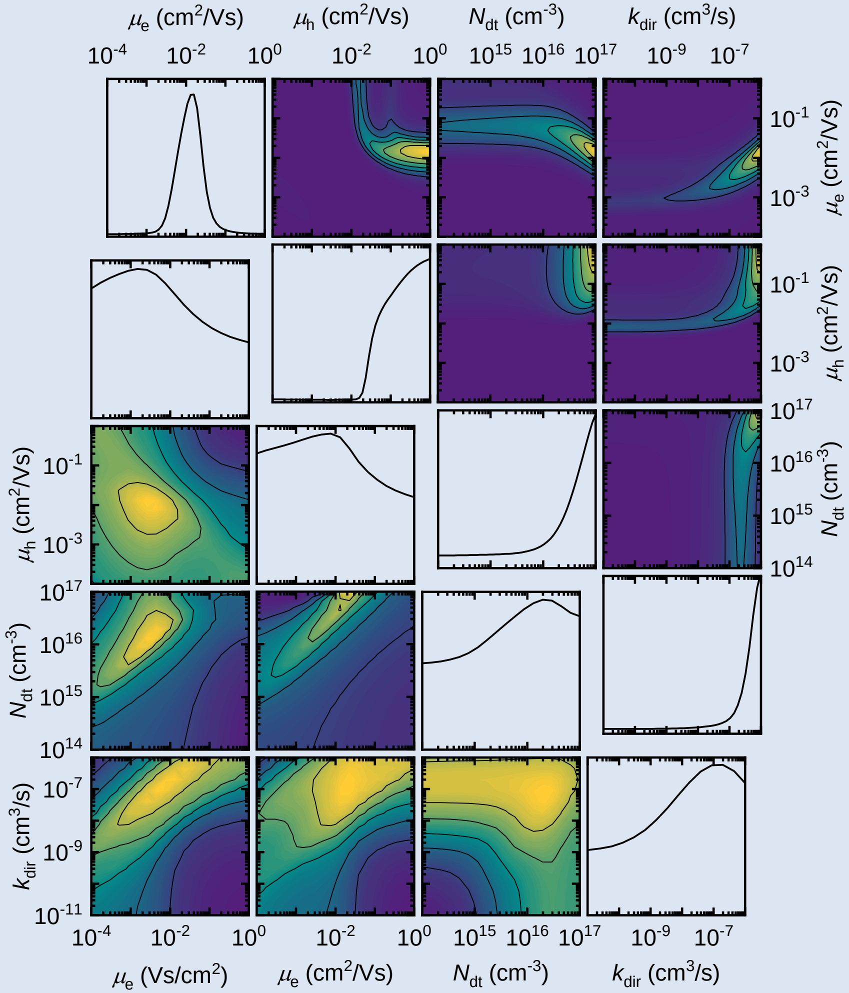
JÜLICH Forschungszentrum

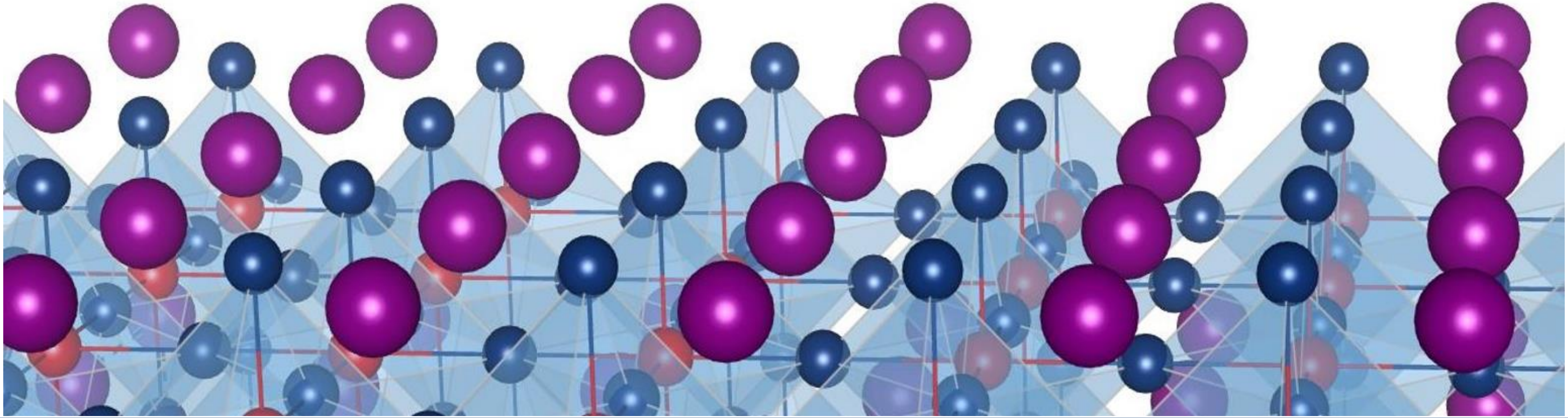
Outline

1) Measuring Disorder



2) Inferring Parameters



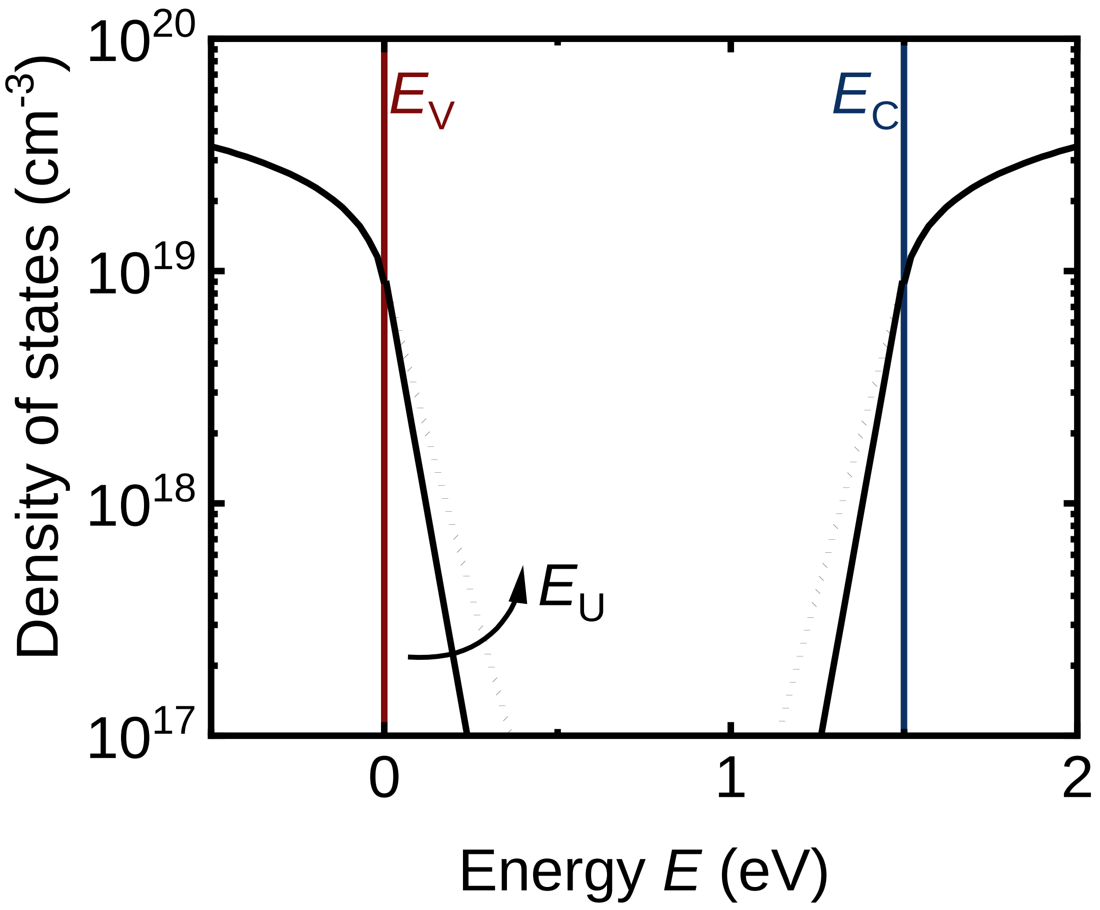


Part 1: Measuring Disorder

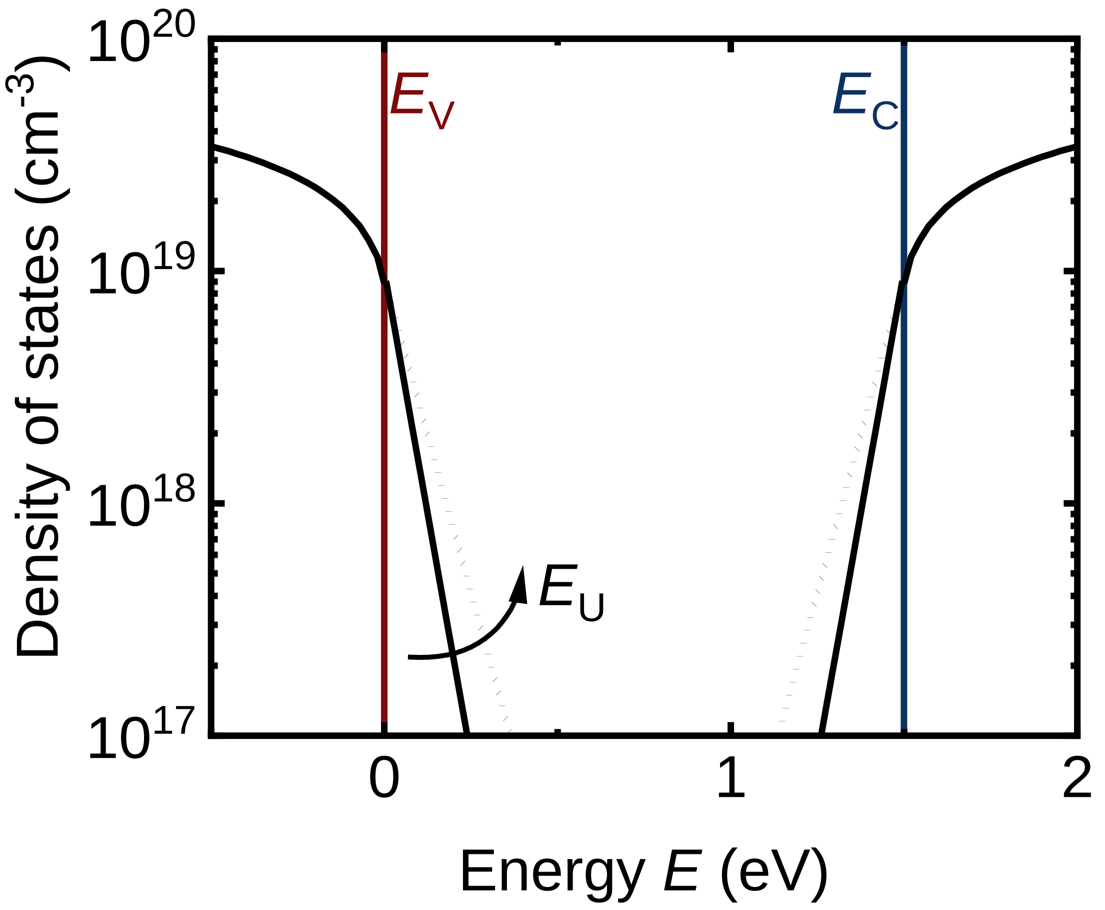
Optical vs. Electrical Methods

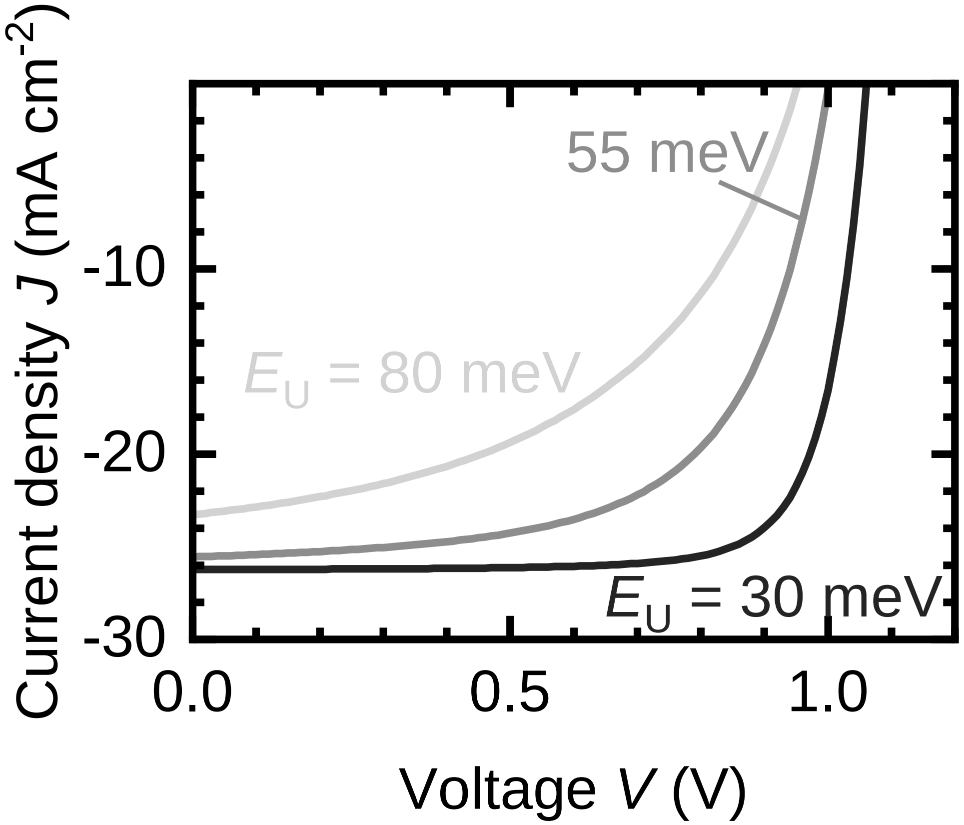
Motivation

The Urbach Energy



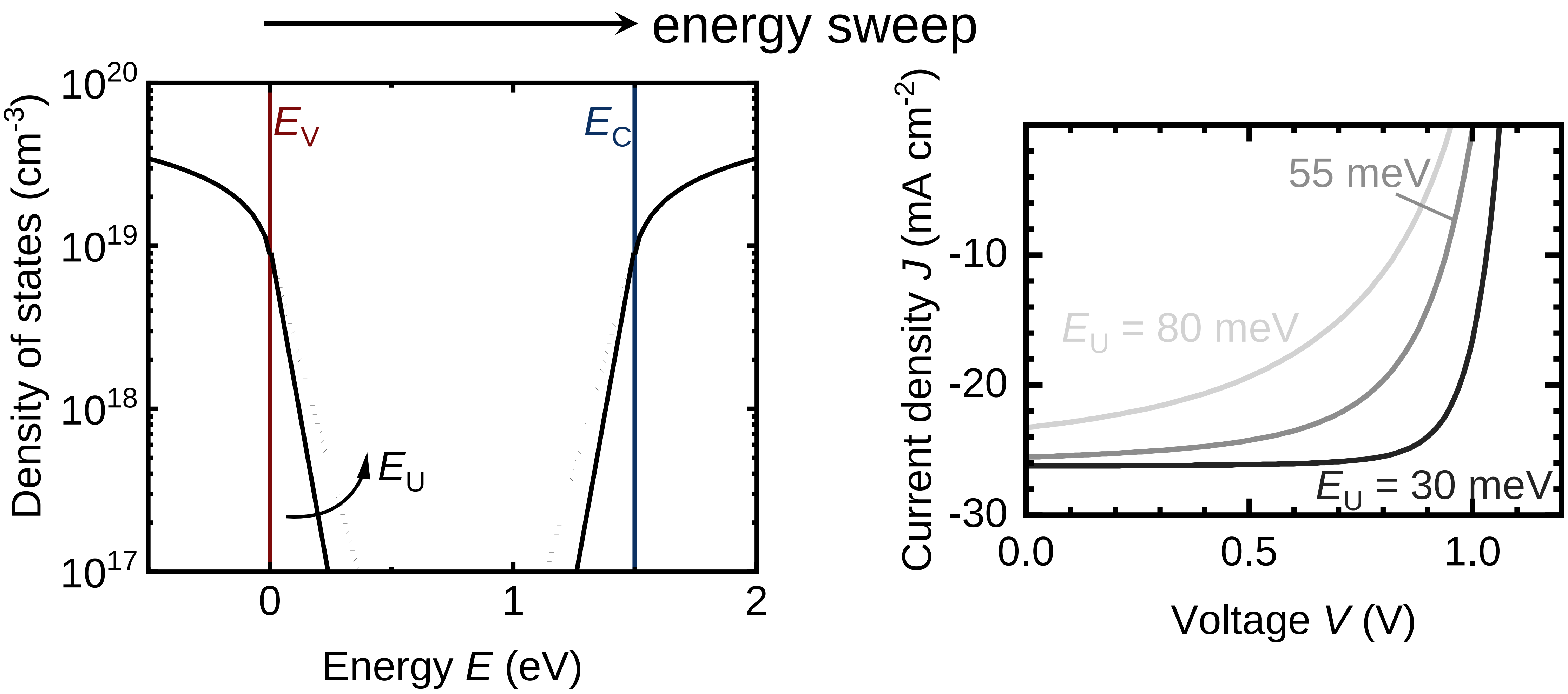
Motivation
The Urbach Energy





Motivation

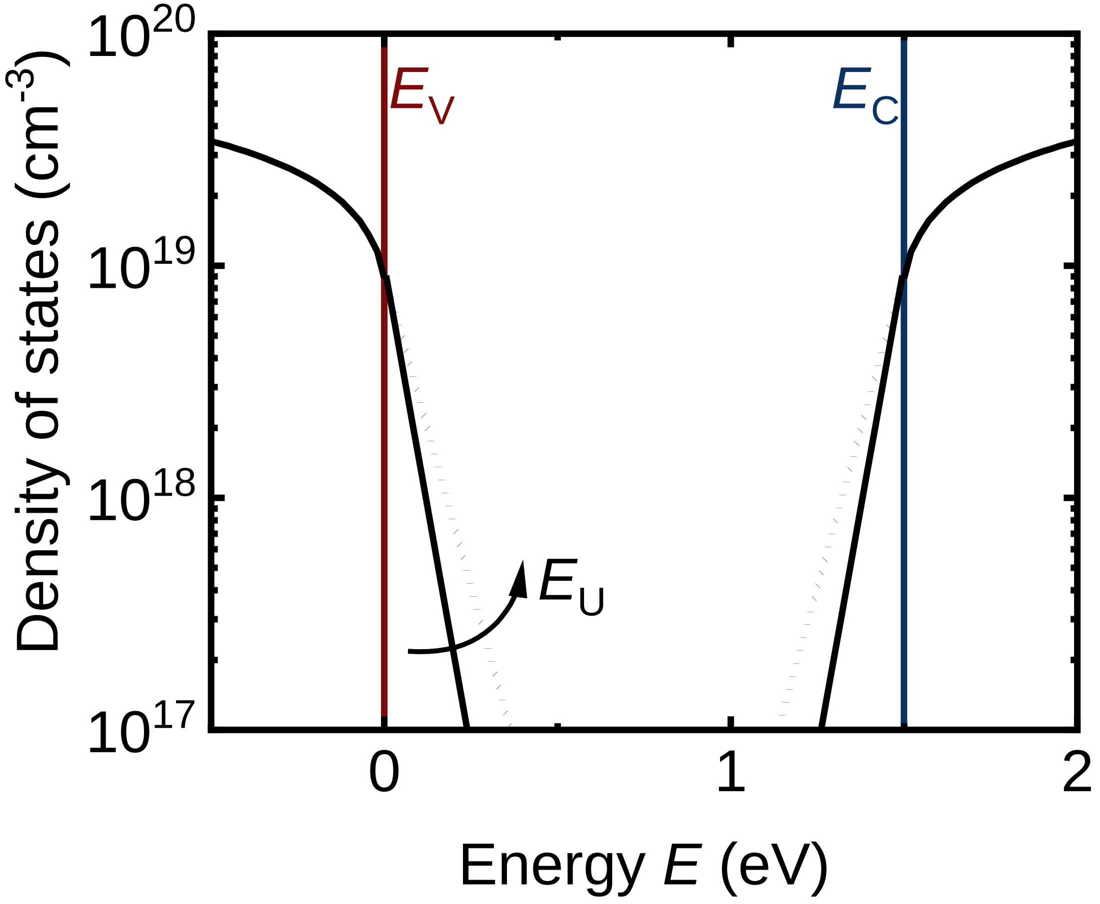
Measuring the Urbach Energy



Motivation

Measuring the Urbach Energy

→ energy sweep → photoexcitation

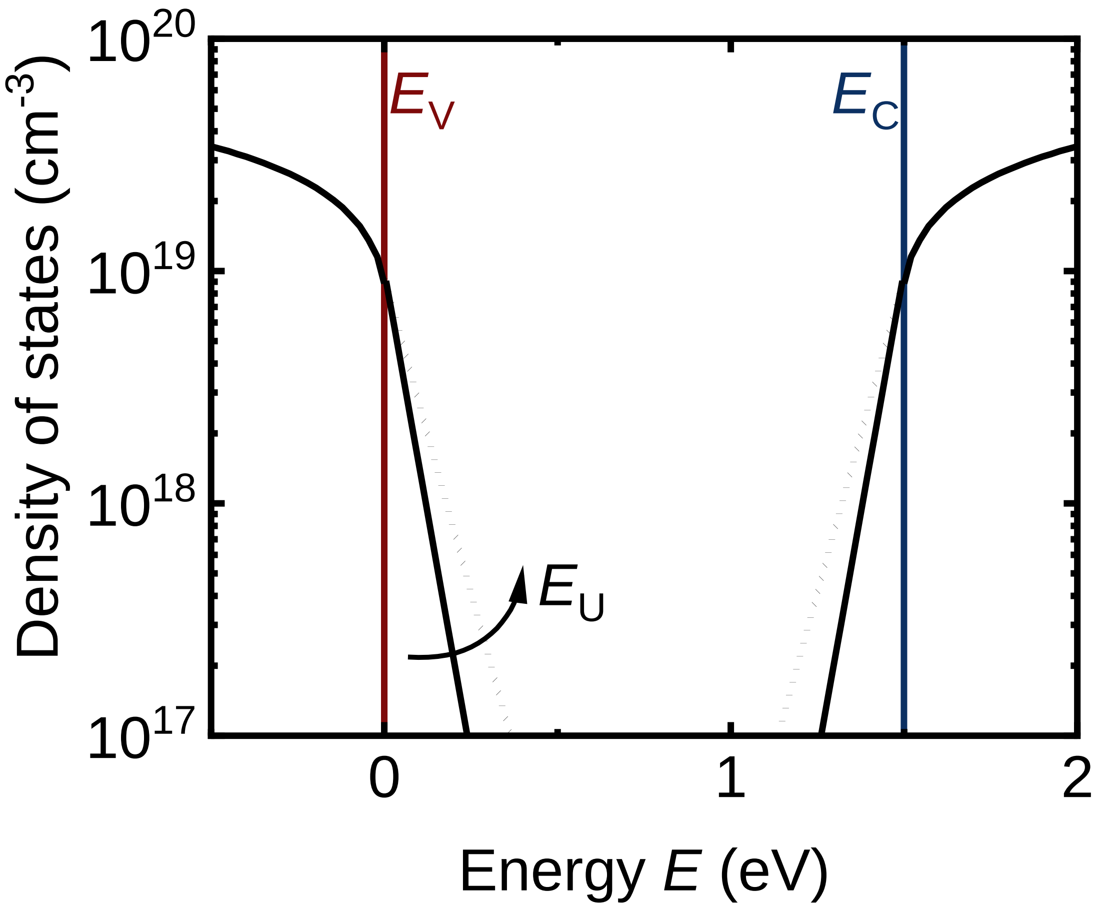


Motivation

Measuring the Urbach Energy

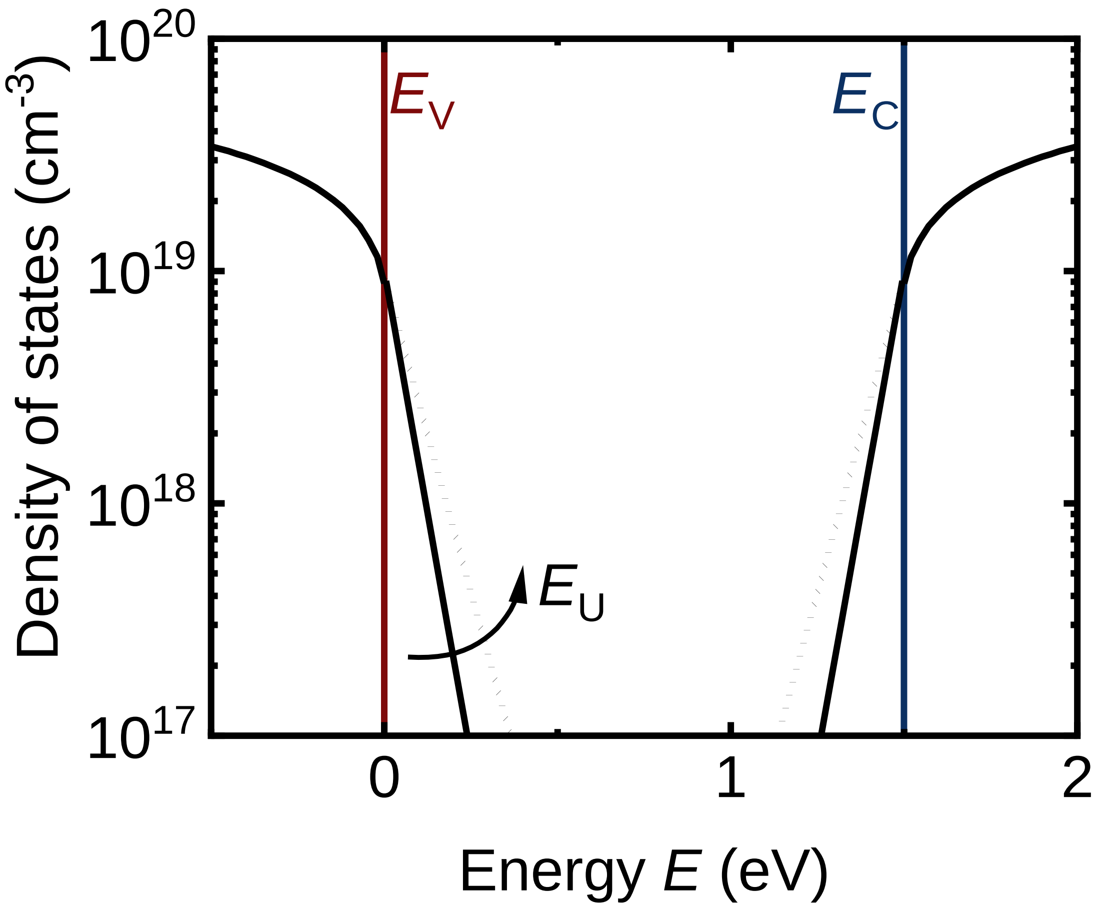
→ energy sweep → photoexcitation

→ voltage



Motivation

Measuring the Urbach Energy

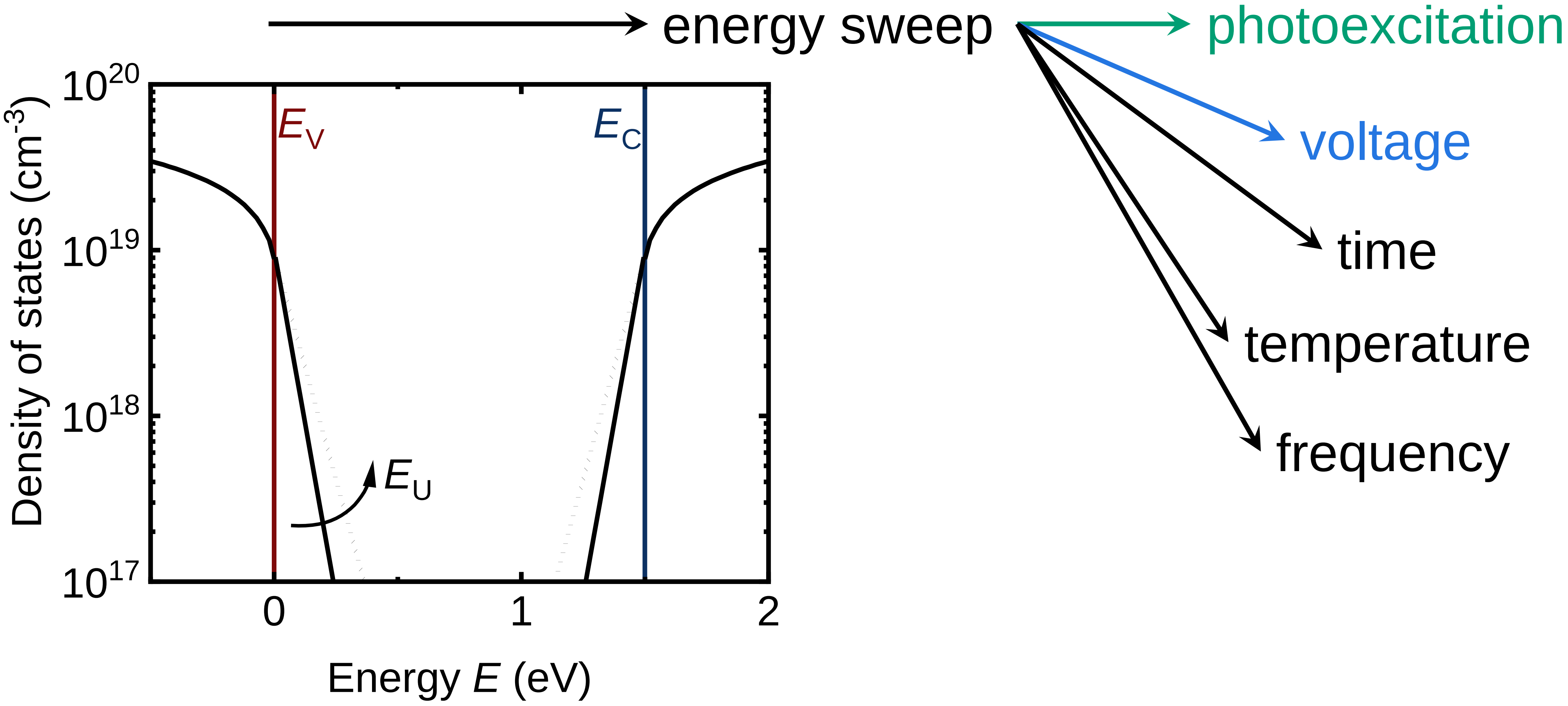


→ energy sweep → photoexcitation

- voltage
- time
- temperature
- frequency

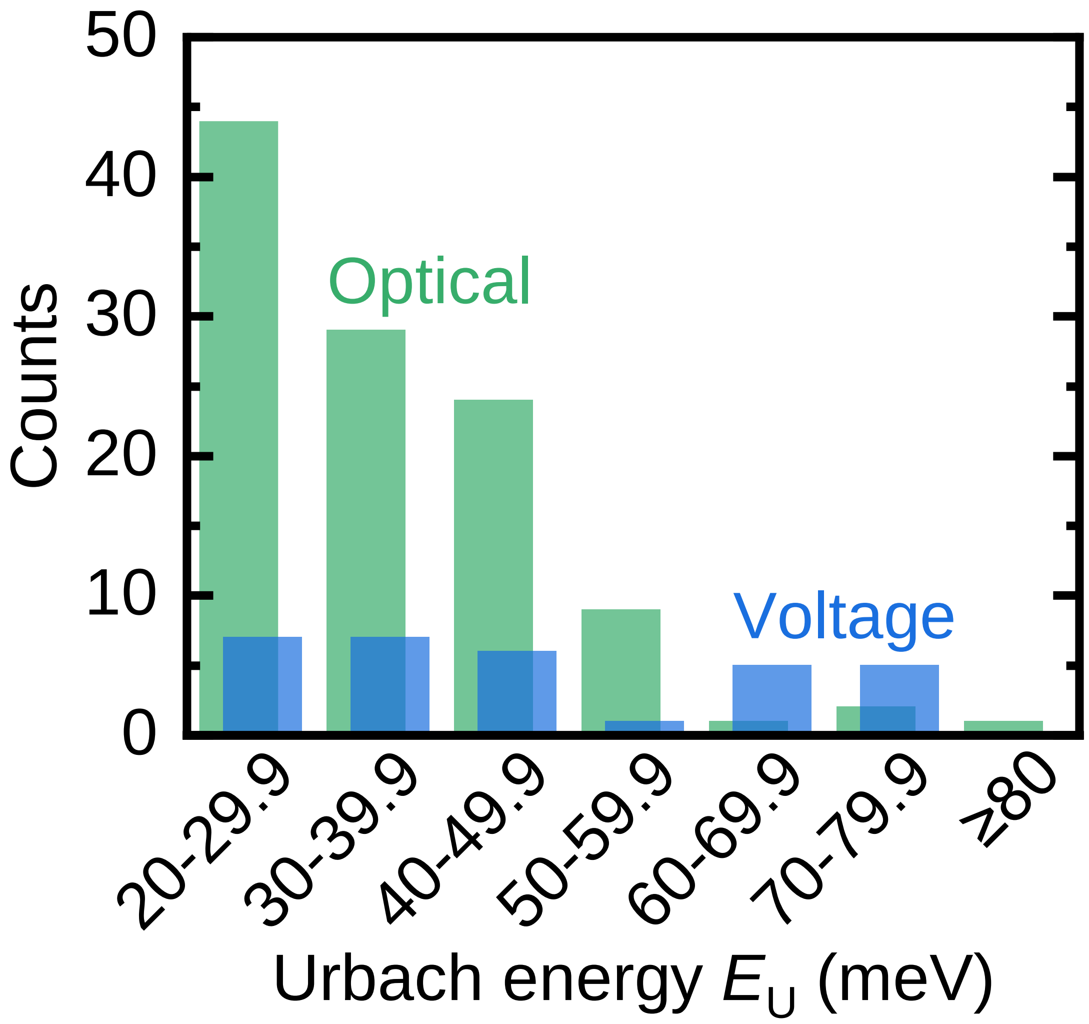
Motivation

Measuring the Urbach Energy



Motivation

Measuring the Urbach Energy



photoexcitation

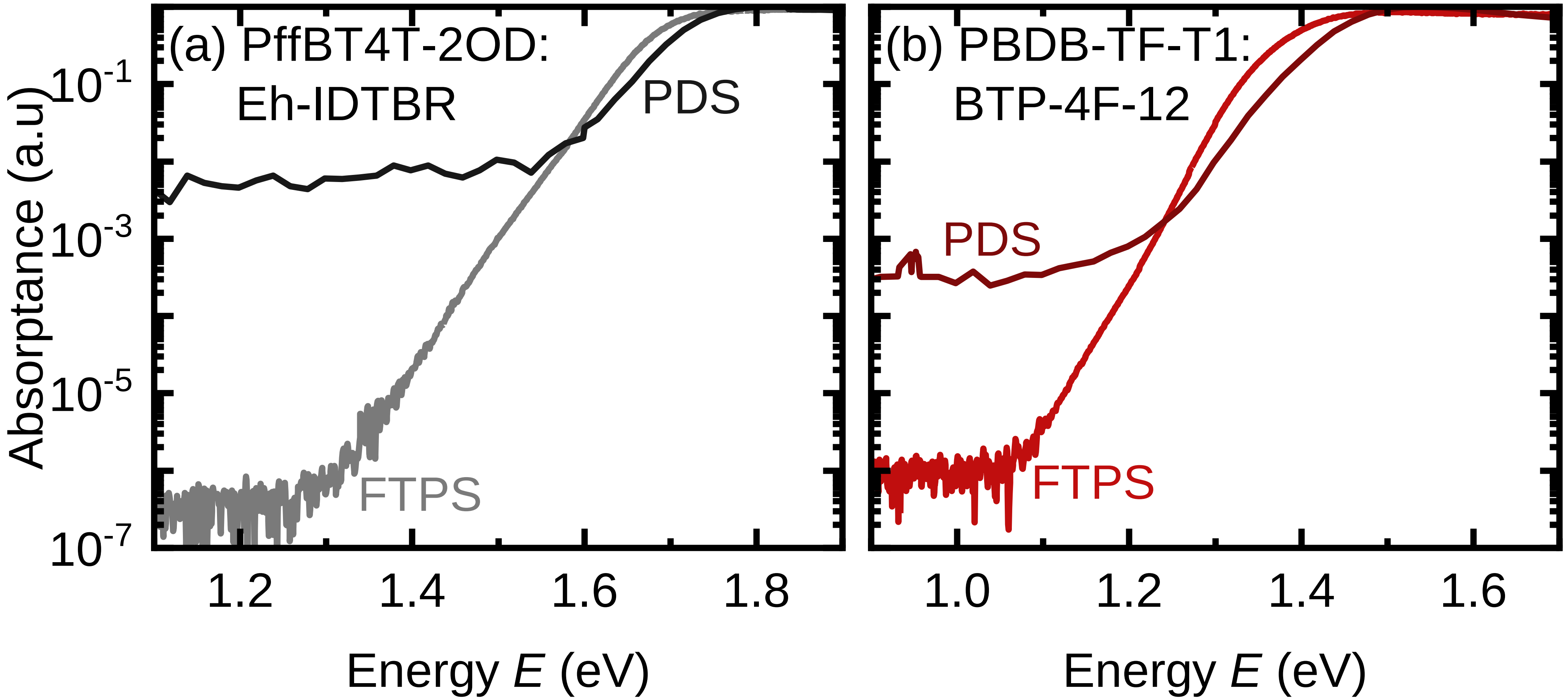
voltage

time

temperature

frequency

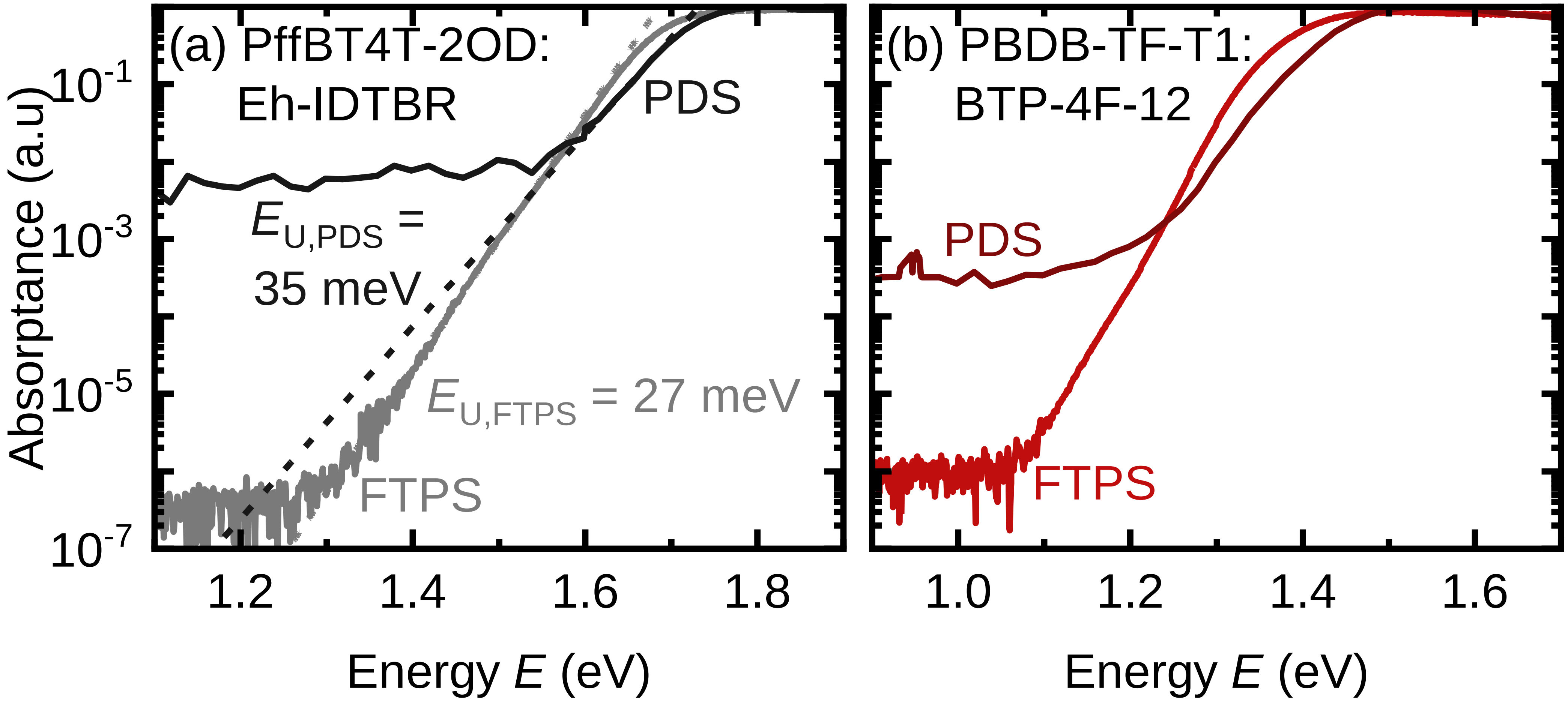
Optical Measurements



high disorder

low disorder

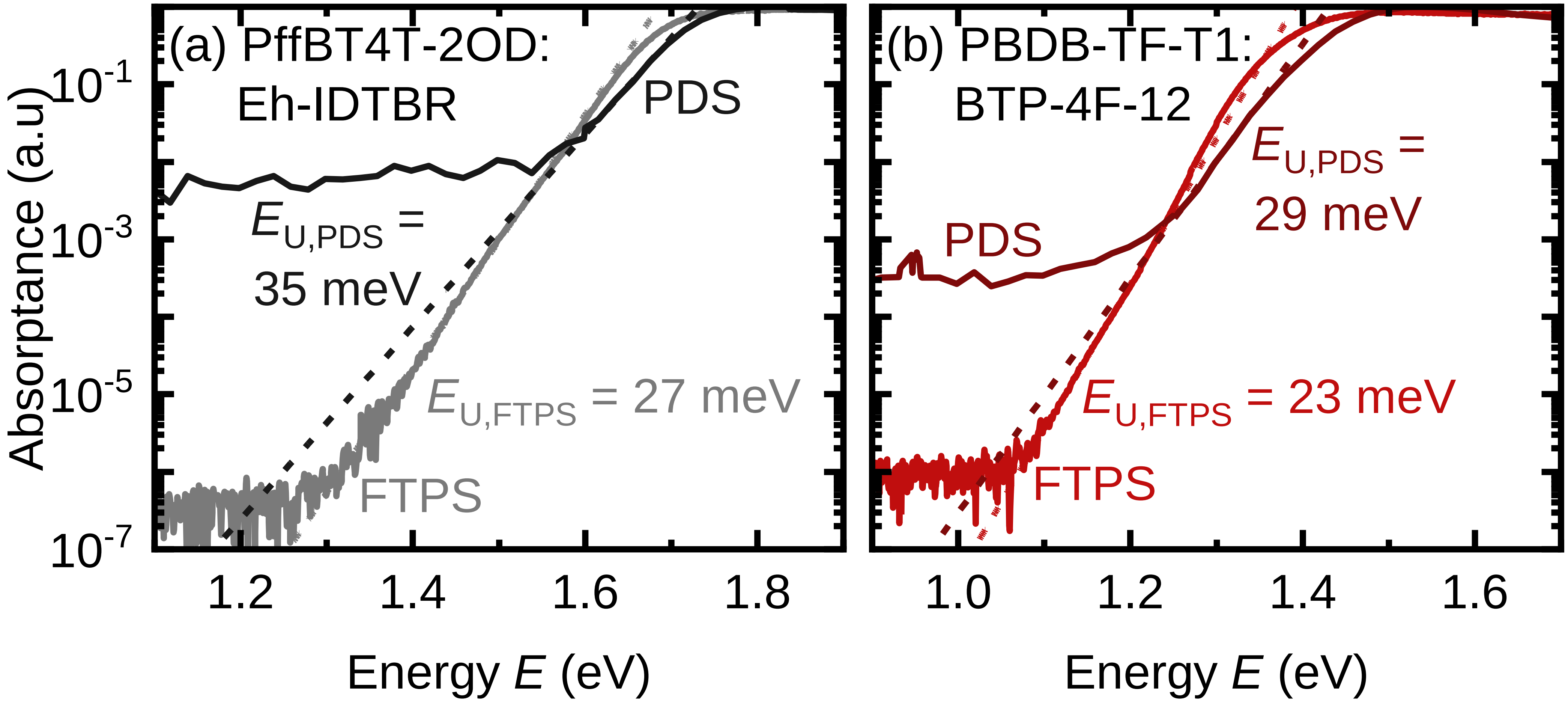
Optical Measurements



“high” disorder

low disorder

Optical Measurements



“high” disorder

low disorder

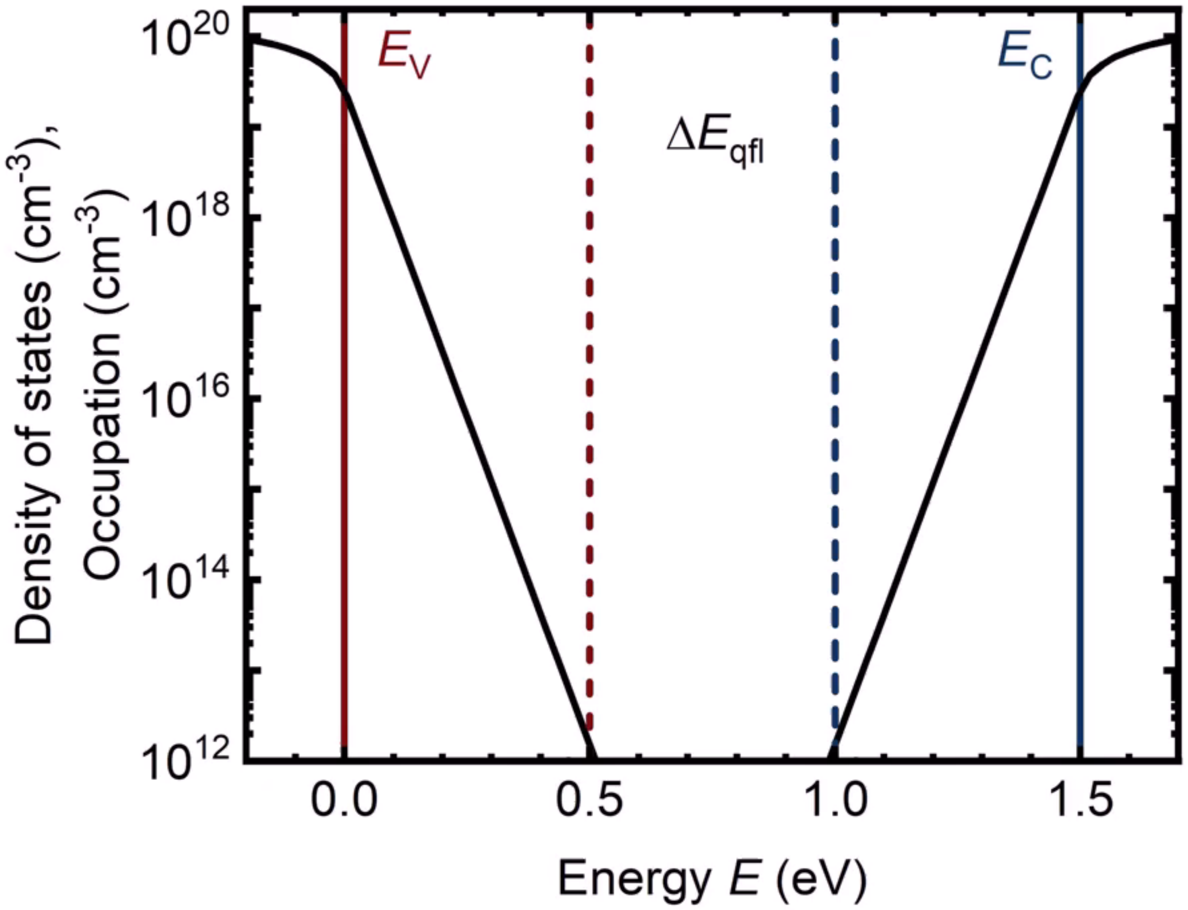
Voltage-dependent measurements

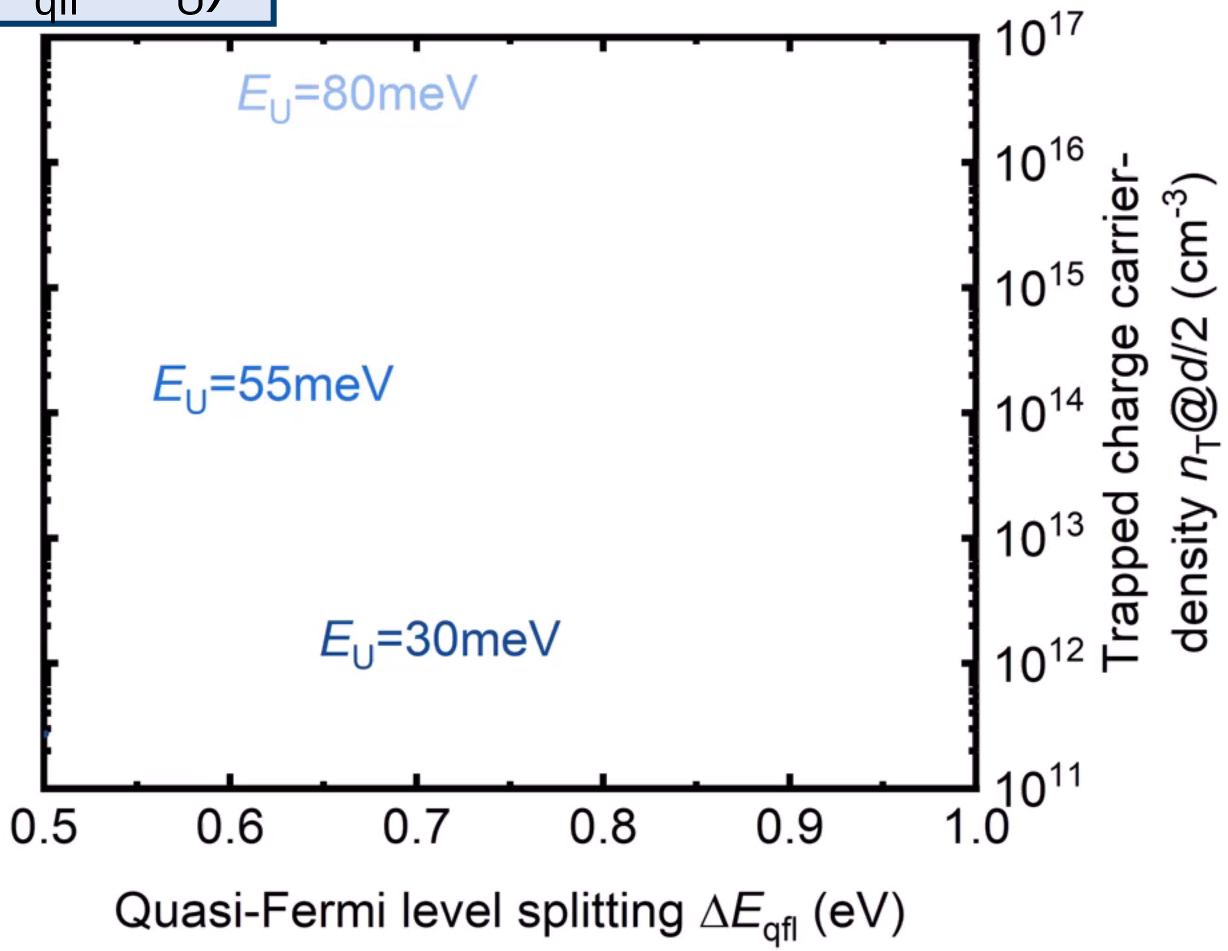
Scanning the density of states

Voltage-dependent measurements

Scanning the density of states

$n_T \sim \exp(\Delta E_{qfl}/2E_U)$

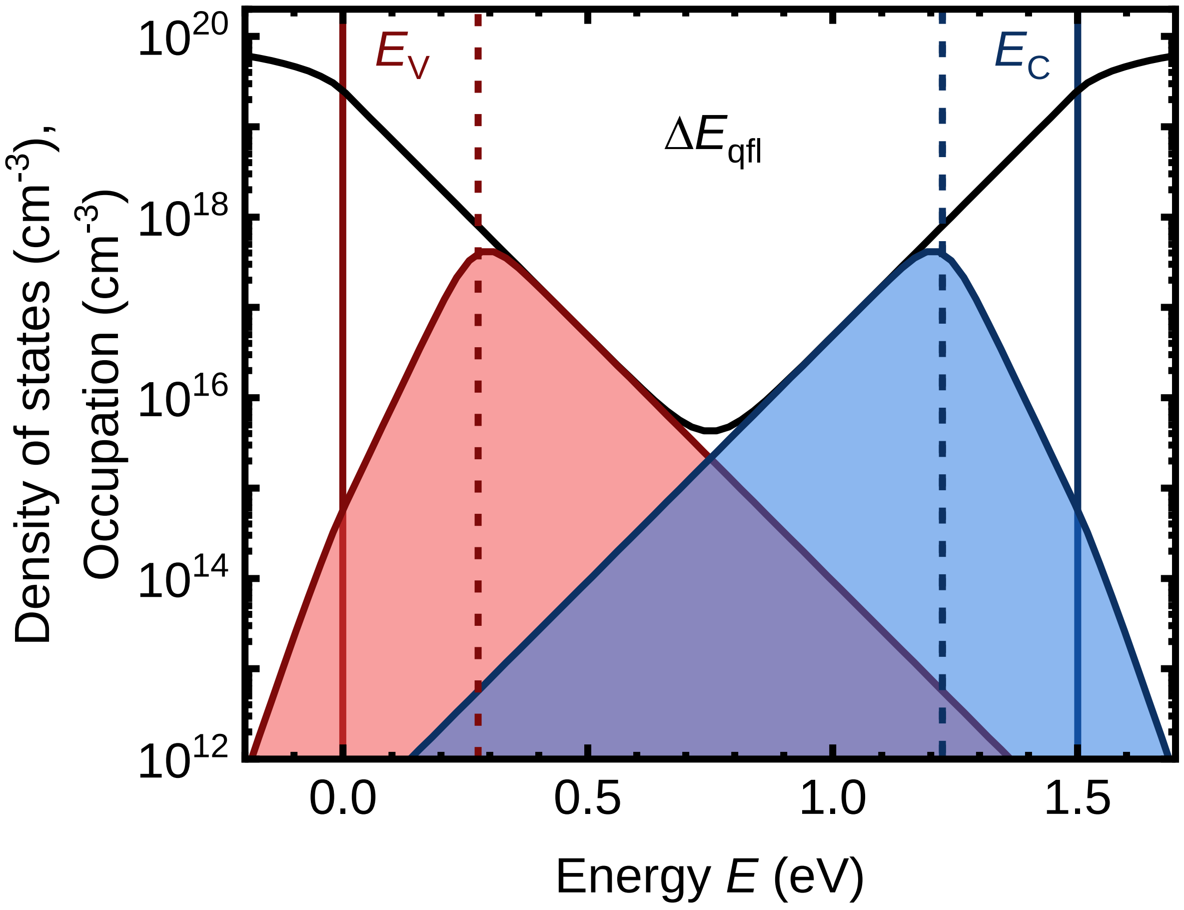


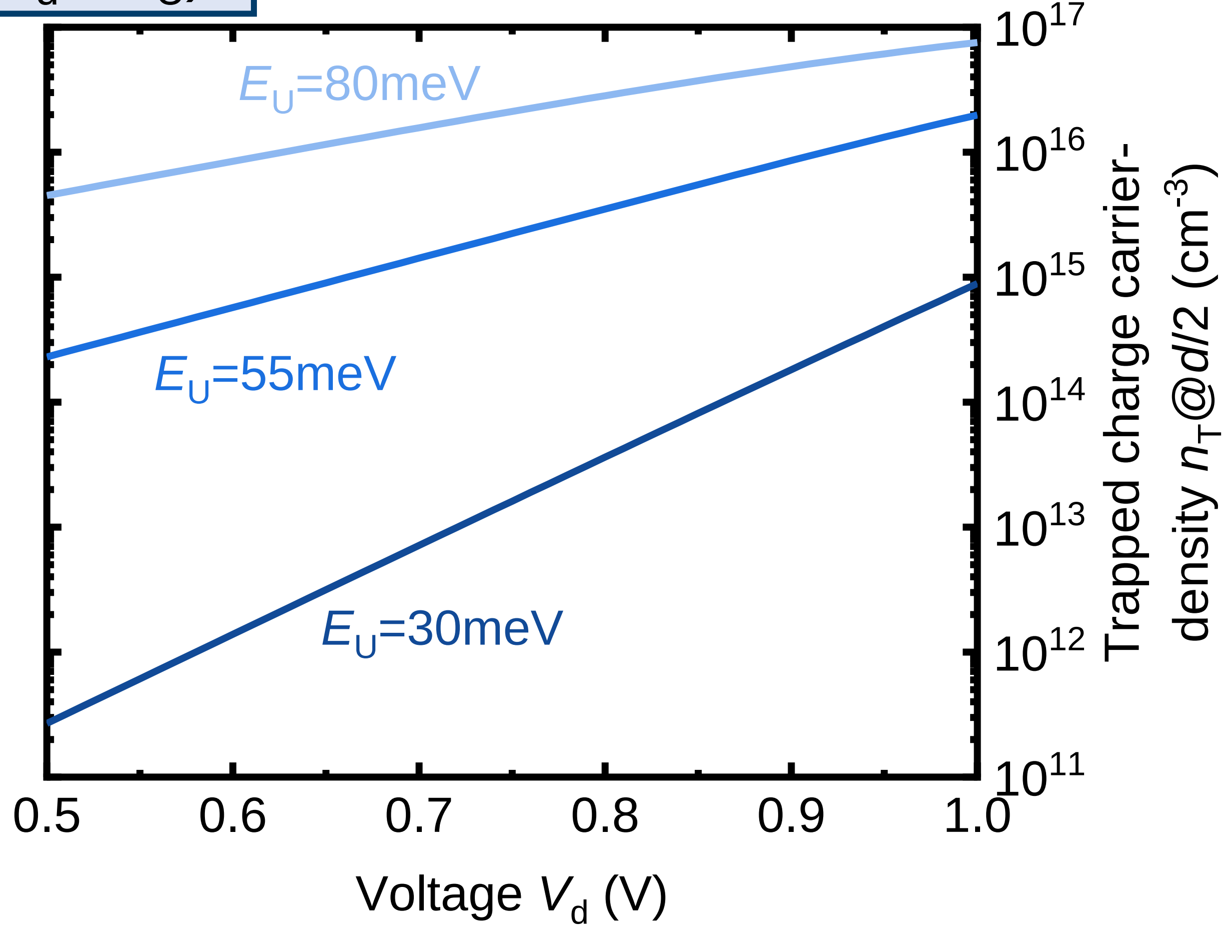


Voltage-dependent measurements

Scanning the density of states

$n_T \sim \exp(qV_d/2E_U)$

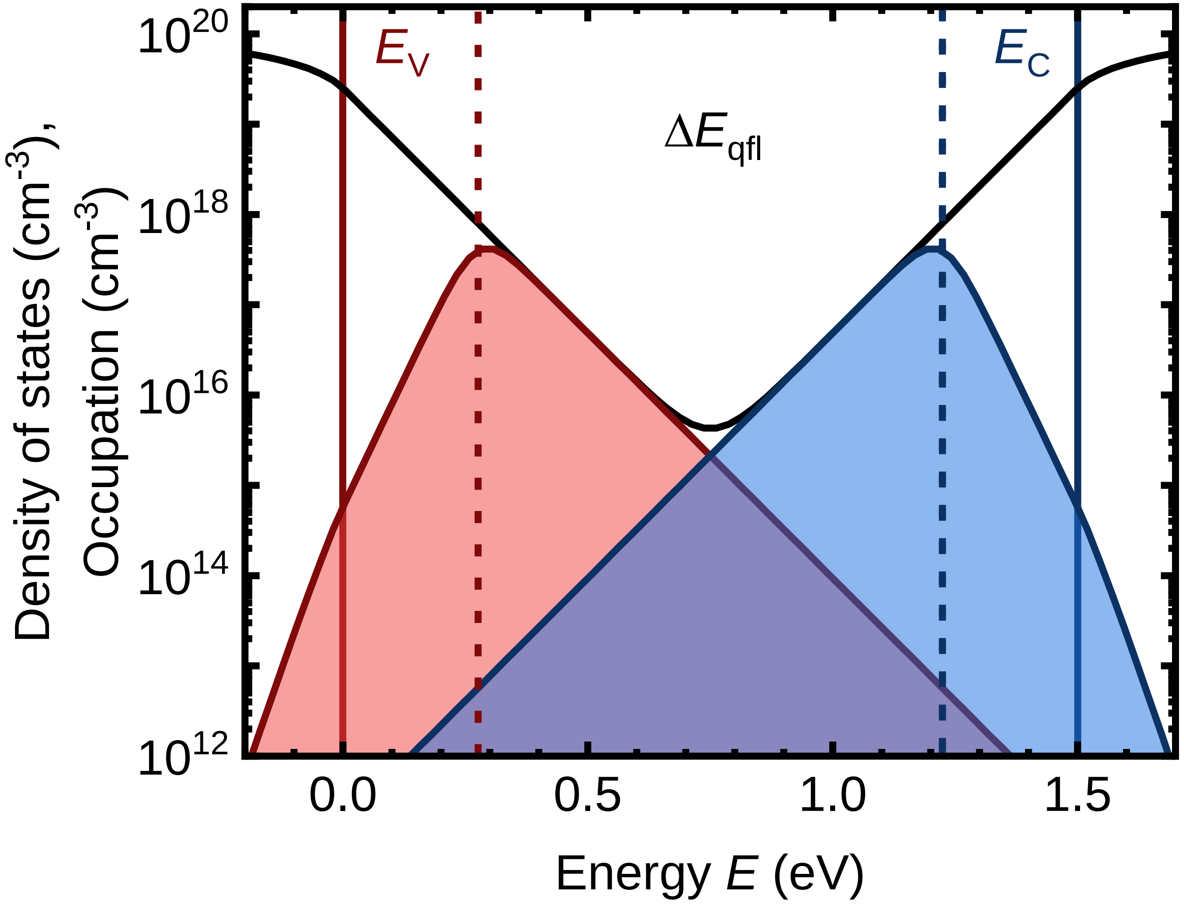


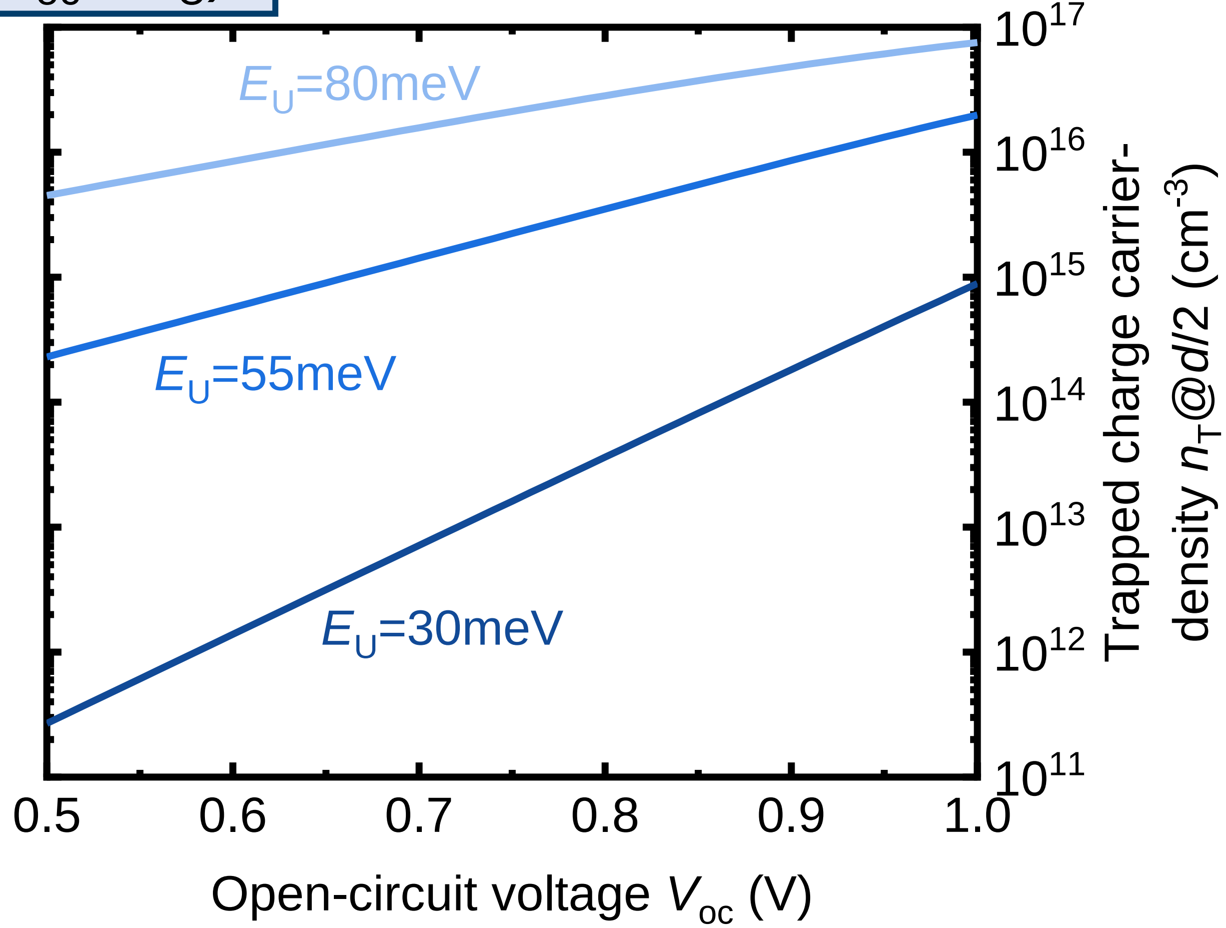


Voltage-dependent measurements

Scanning the density of states

$$n_T \sim \exp(qV_{oc}/2E_U)$$

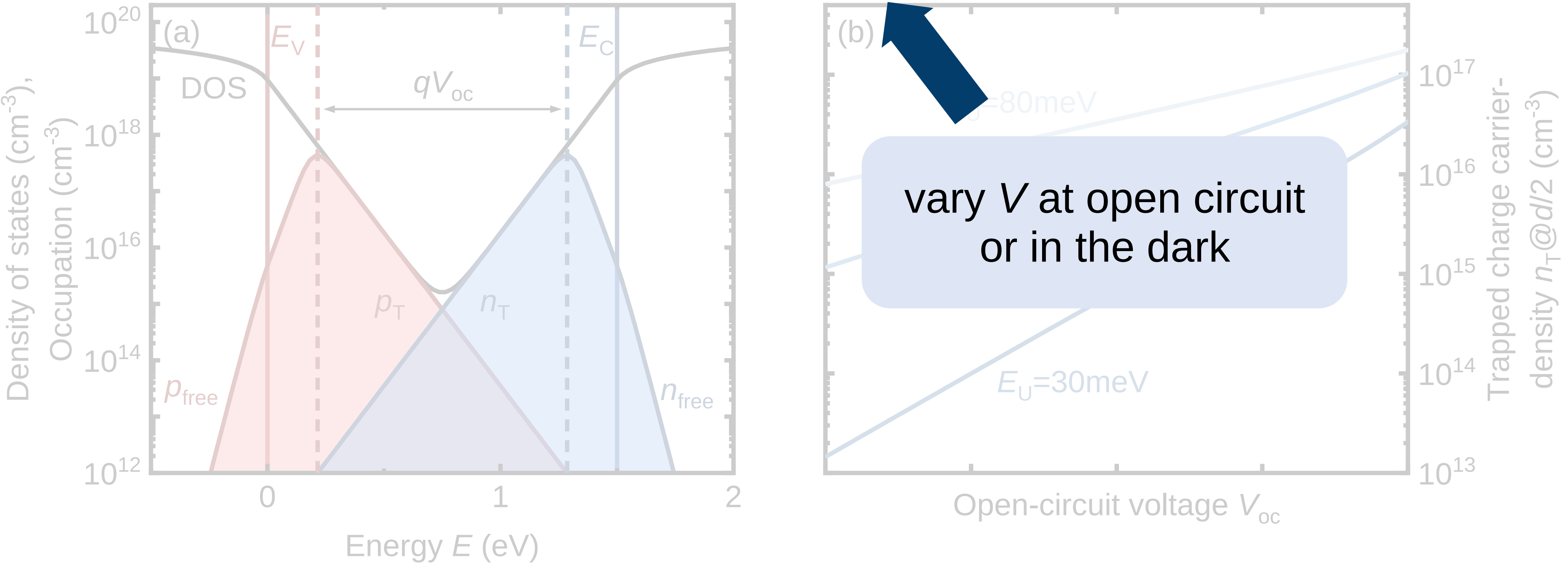




Voltage-dependent measurements

Scanning the density of states

$n_T \sim \exp(qV/2E_U)$

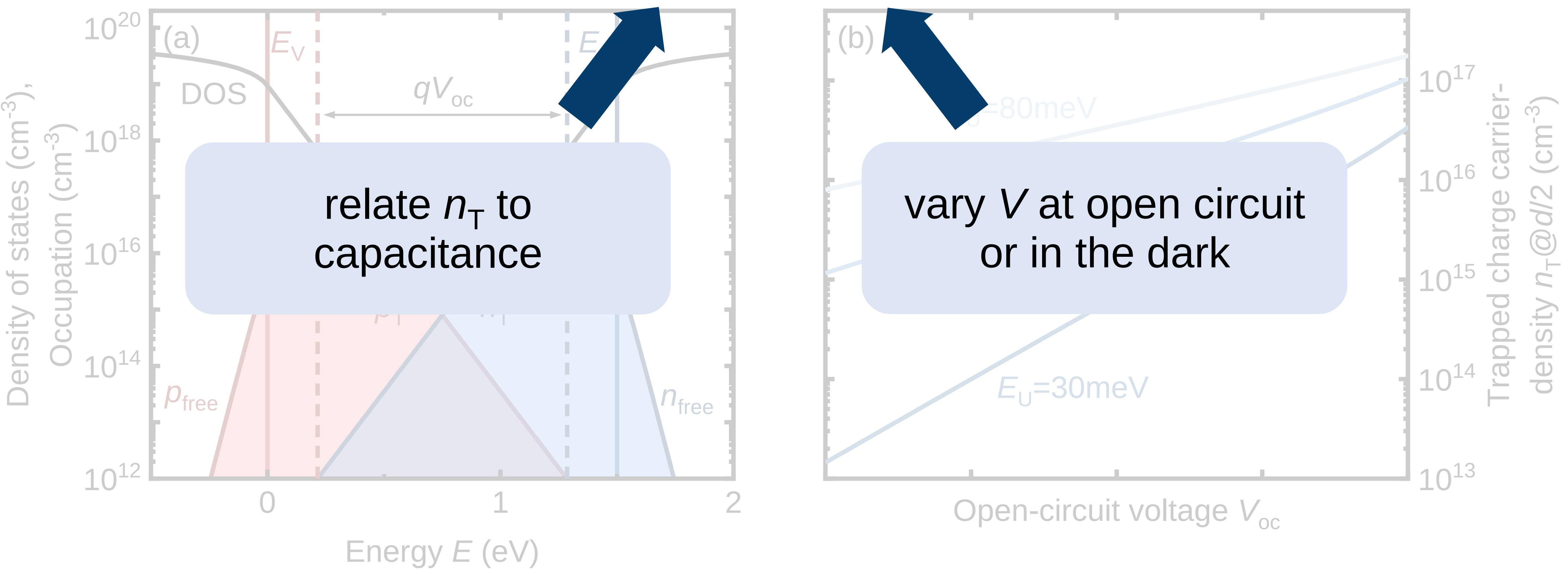


vary V at open circuit
or in the dark

Voltage-dependent measurements

Scanning the density of states

$$n_T \sim \exp(qV/2E_U)$$

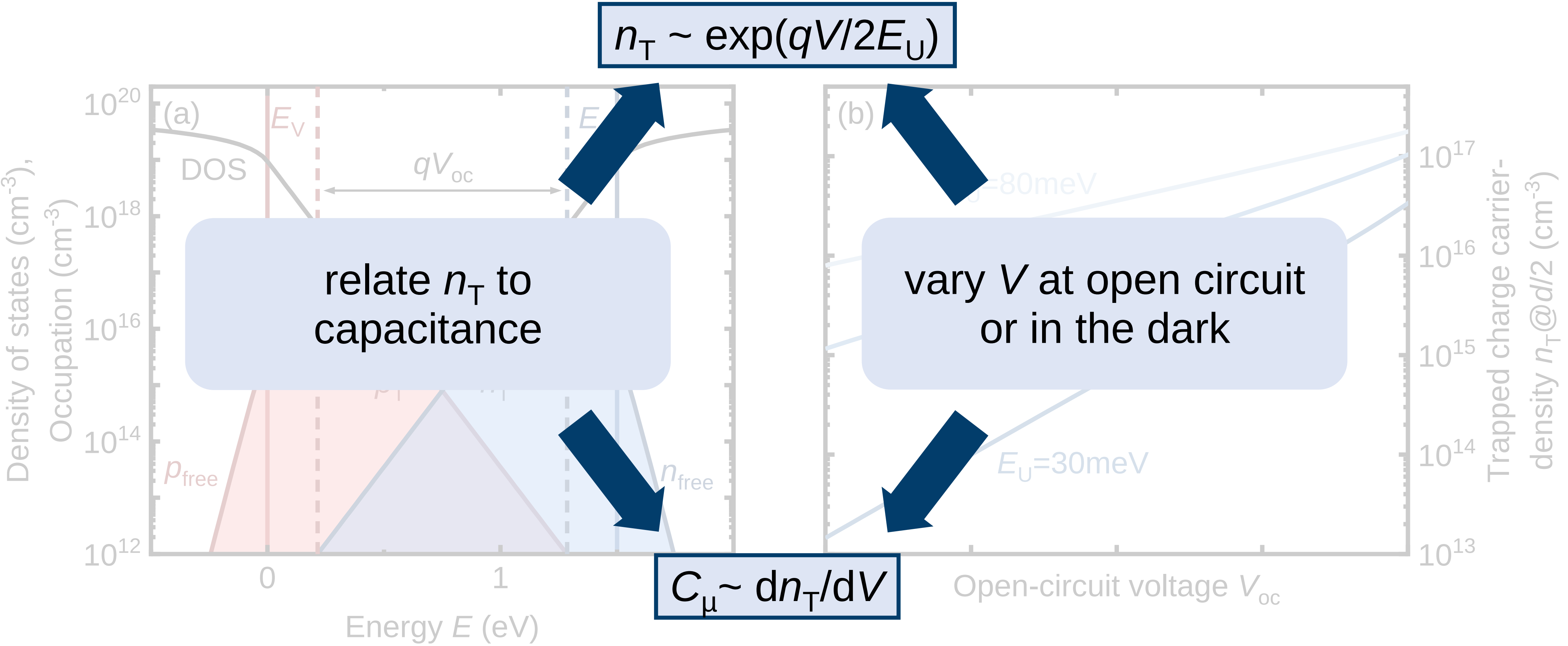


relate n_T to capacitance

vary V at open circuit or in the dark

Voltage-dependent measurements

Scanning the density of states



Voltage-dependent measurements

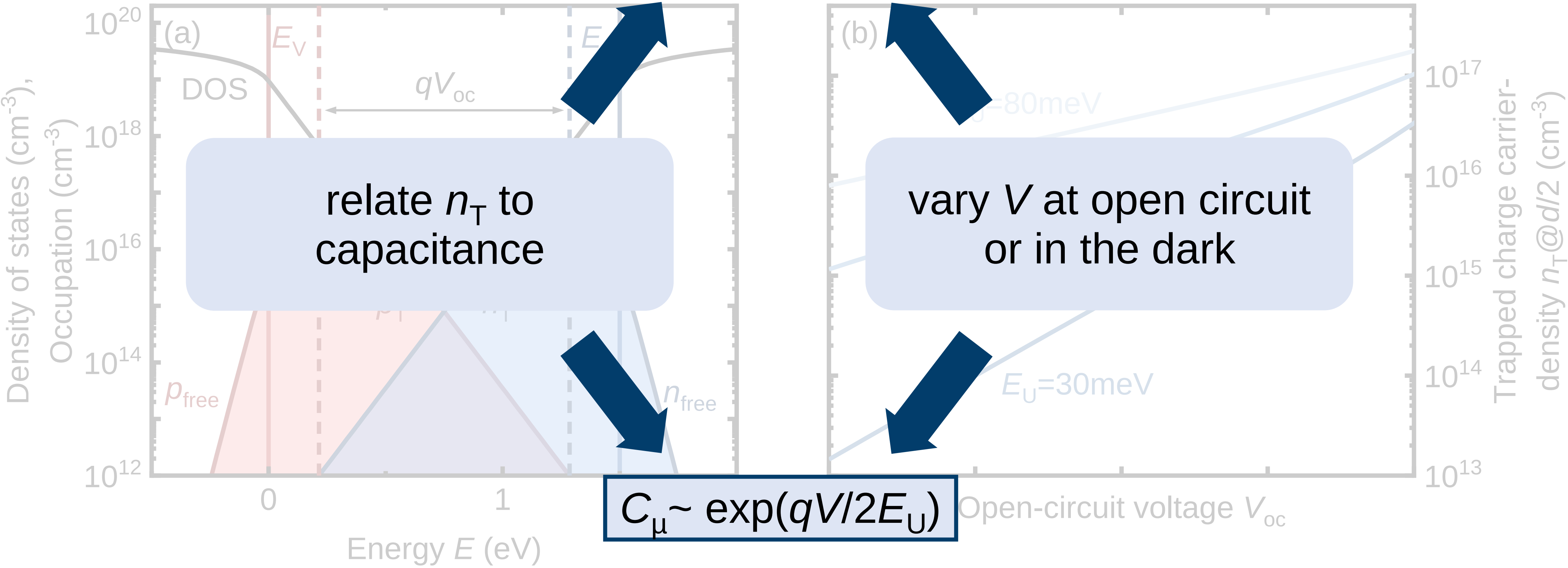
Scanning the density of states

$n_T \sim \exp(qV/2E_U)$

relate n_T to capacitance

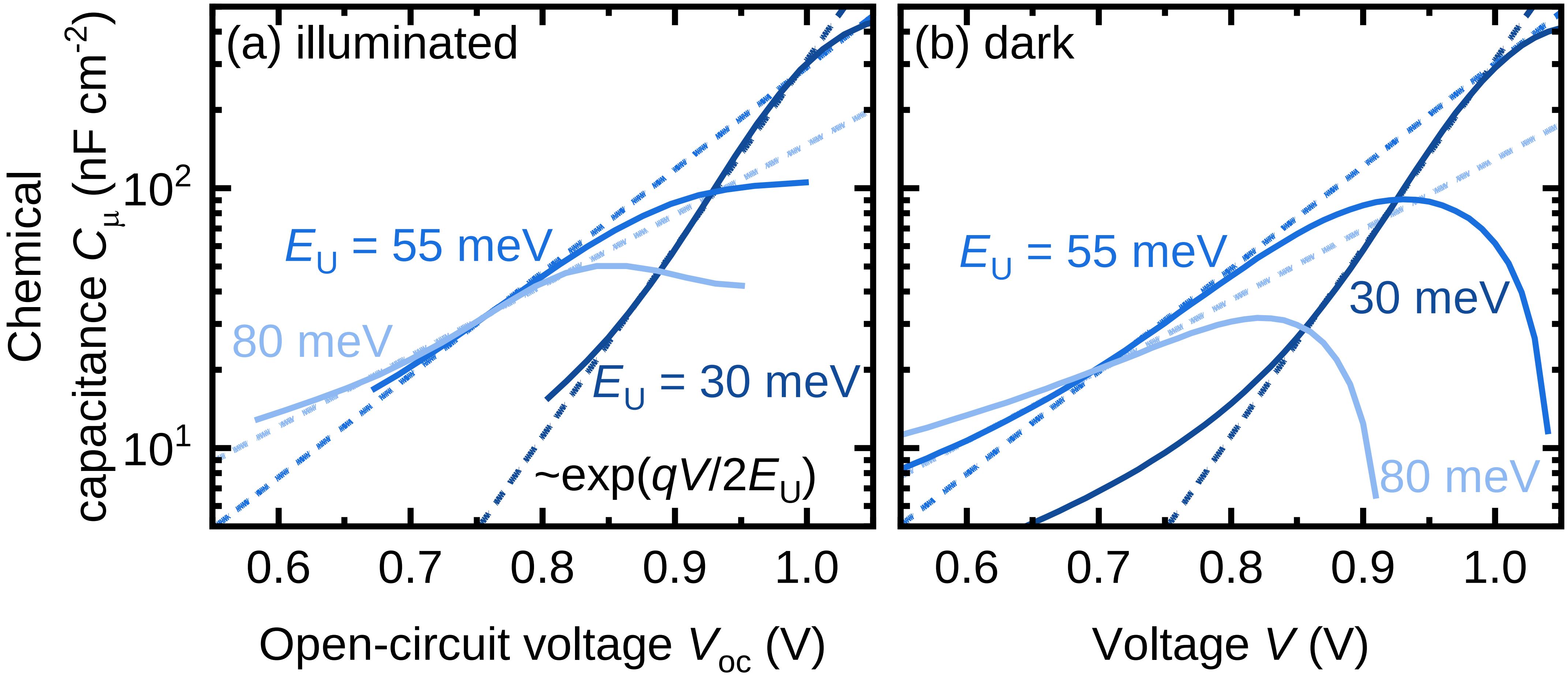
vary V at open circuit or in the dark

$C_\mu \sim \exp(qV/2E_U)$

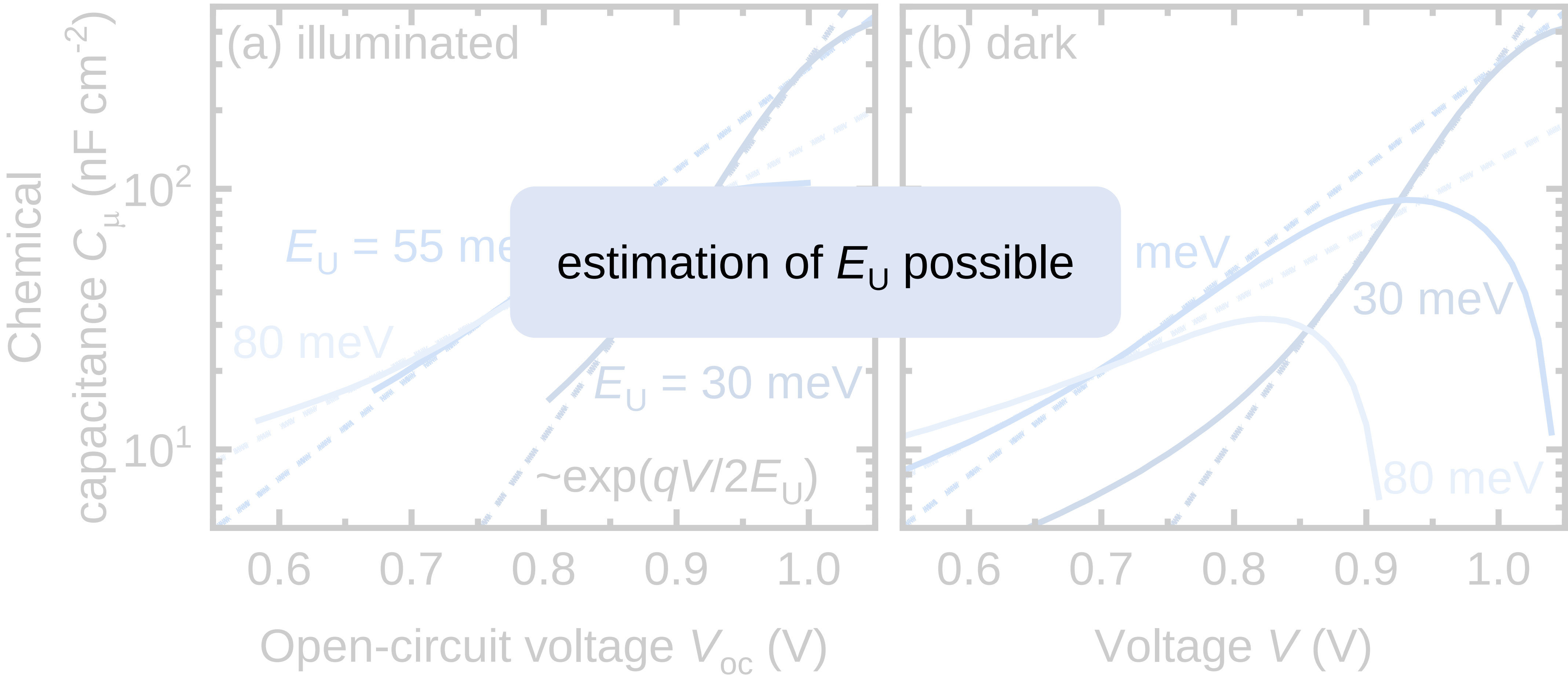


Simulation results

Urbach energy variation



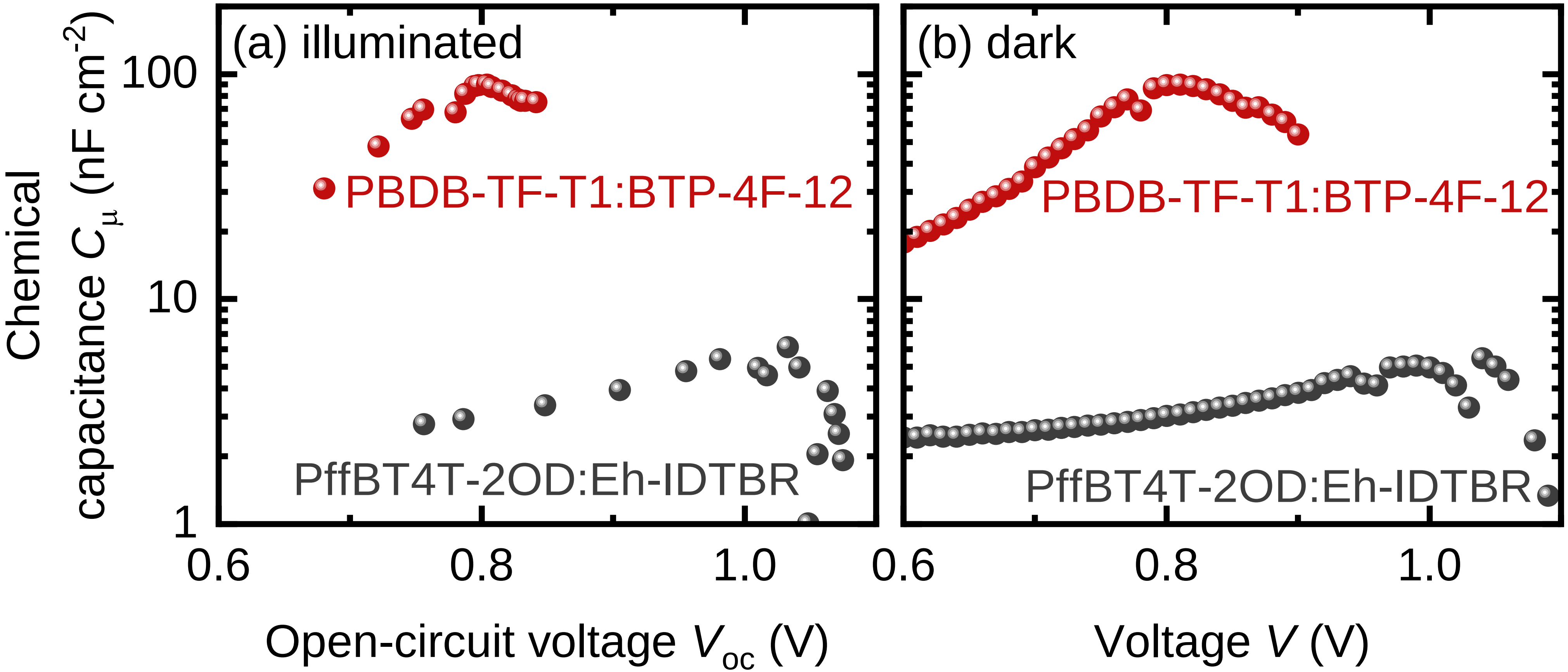
Simulation results
Urbach energy variation



estimation of E_U possible

Voltage-dependent measurements

Experimental results

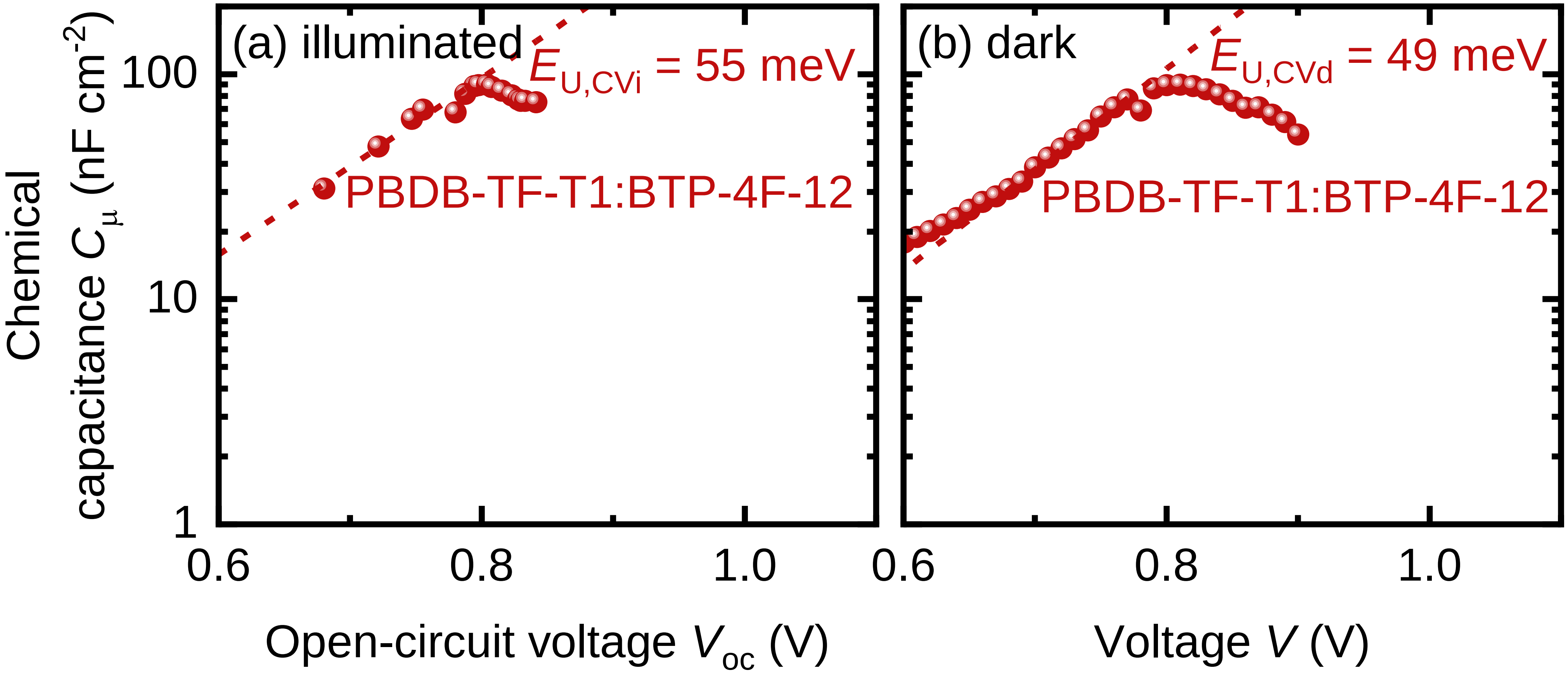


“high” disorder

low disorder

Voltage-dependent measurements

Experimental results

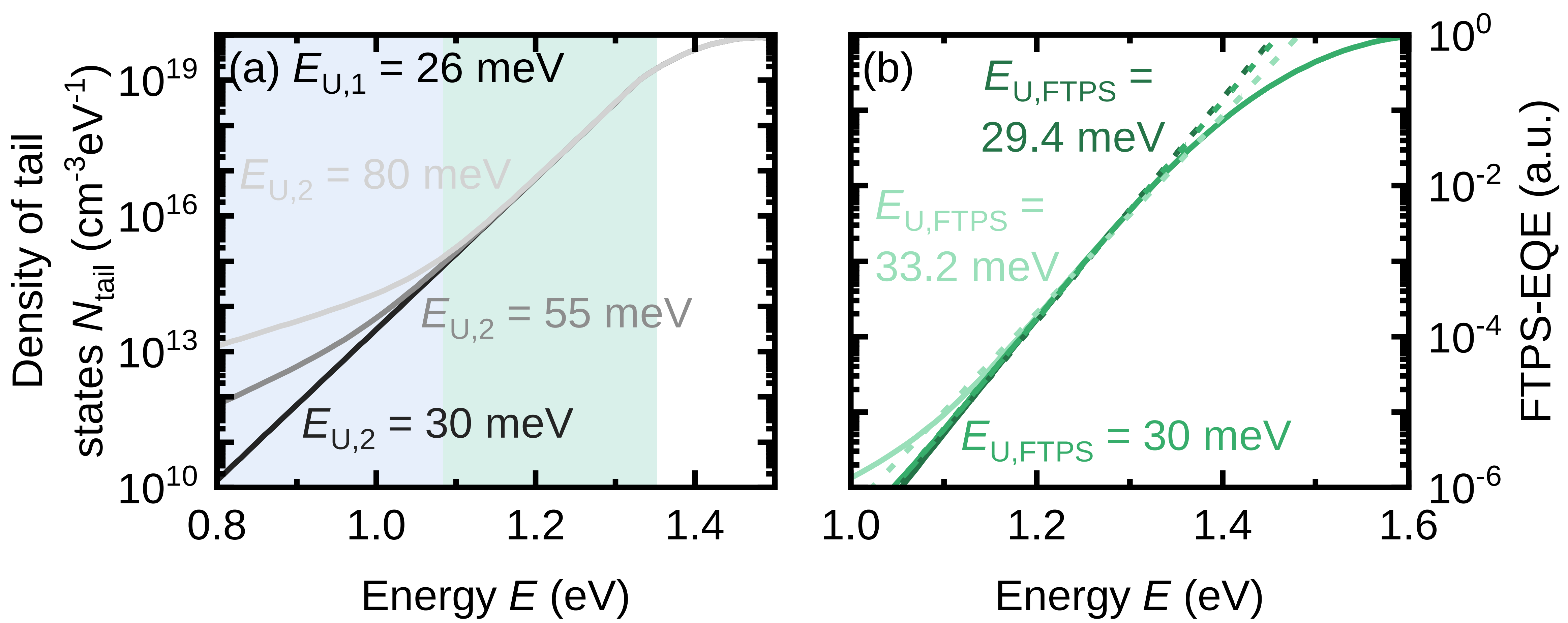


“high” disorder

low disorder

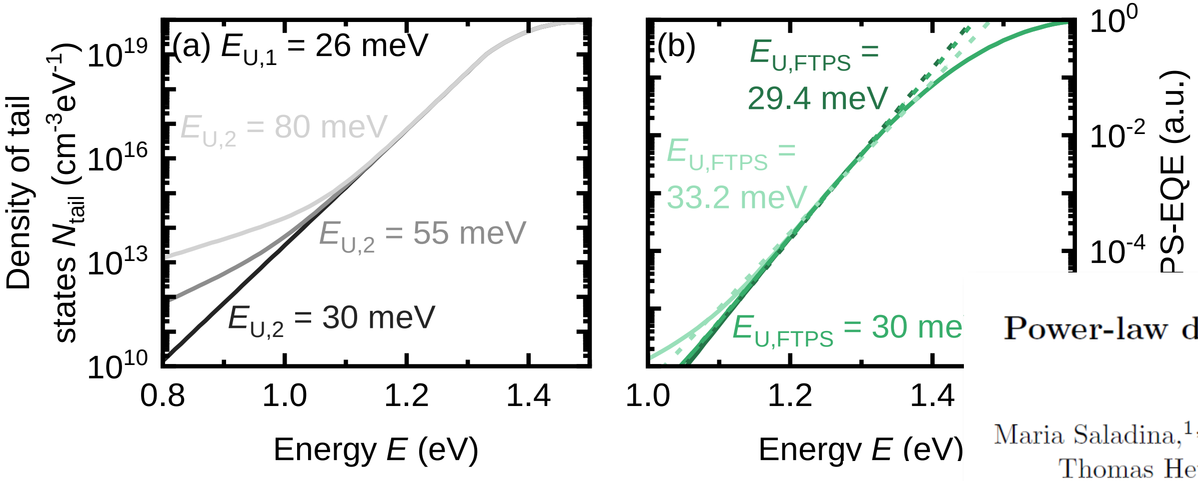
Simulation results

Two exponential band tails



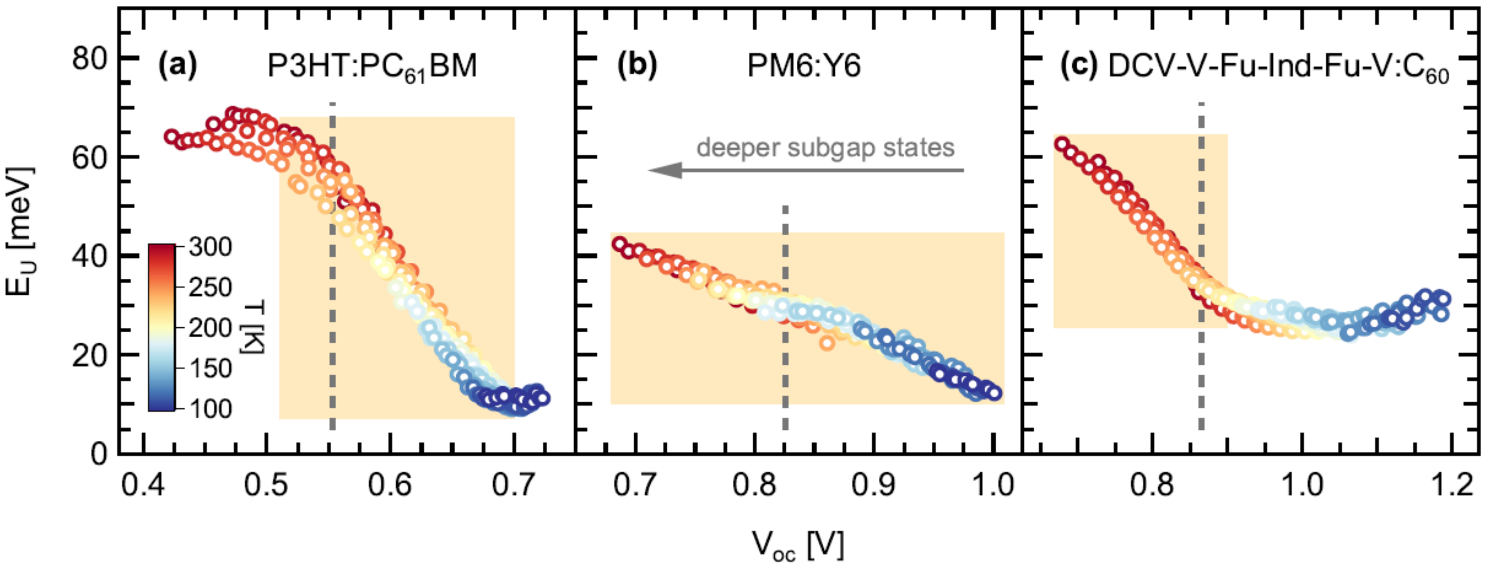
Simulation results

Two exponential band tails



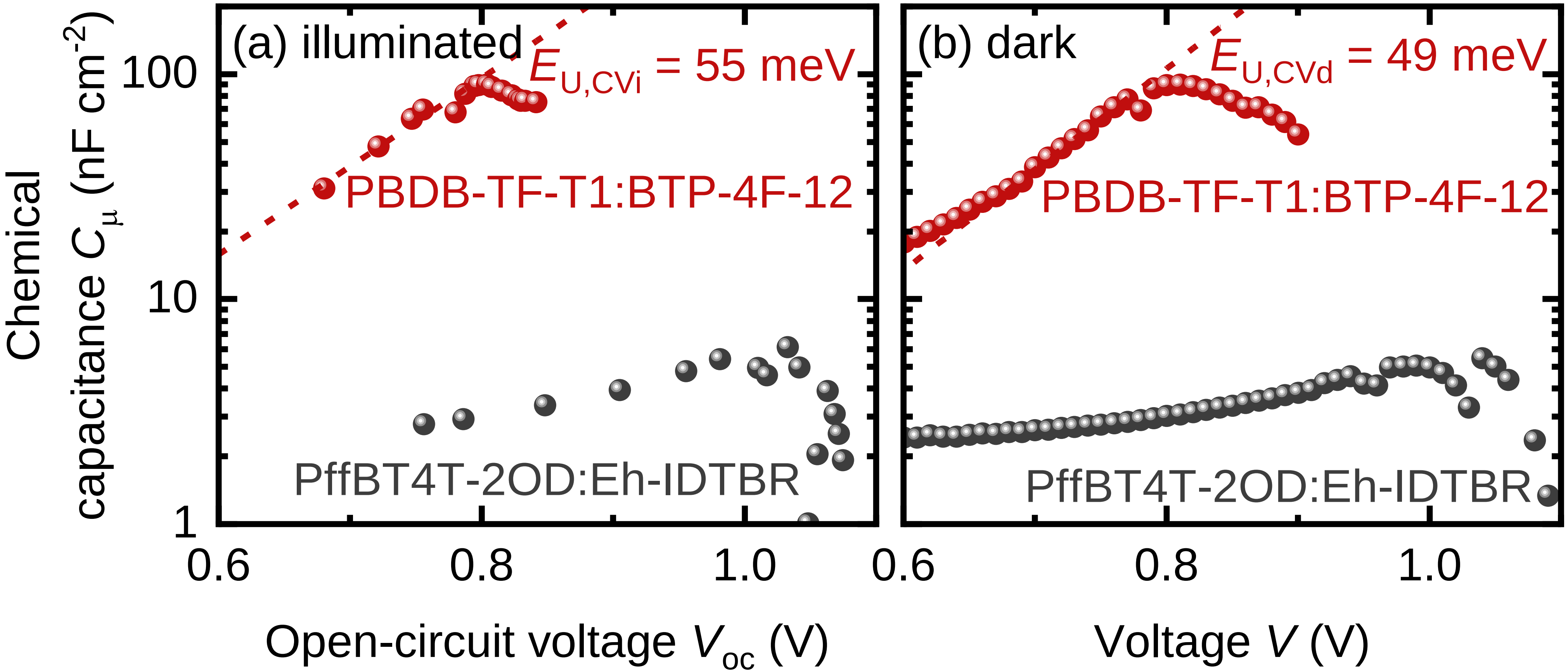
Power-law density of states in organic solar cells revealed by the open-circuit voltage dependence of the ideality factor

Maria Saladina,[1,*] Christopher Wöpke,[1] Clemens Göhler,[1] Ivan Ramirez,[2] Olga Gerdes,[2] Chao Liu,[3,4] Ning Li,[3,4,5] Thomas Heumüller,[3,4] Christoph J. Brabec,[3,4] Karsten Walzer,[2] Martin Pfeiffer,[2] and Carsten Deibel[1,†]



Voltage-dependent measurements

Experimental results

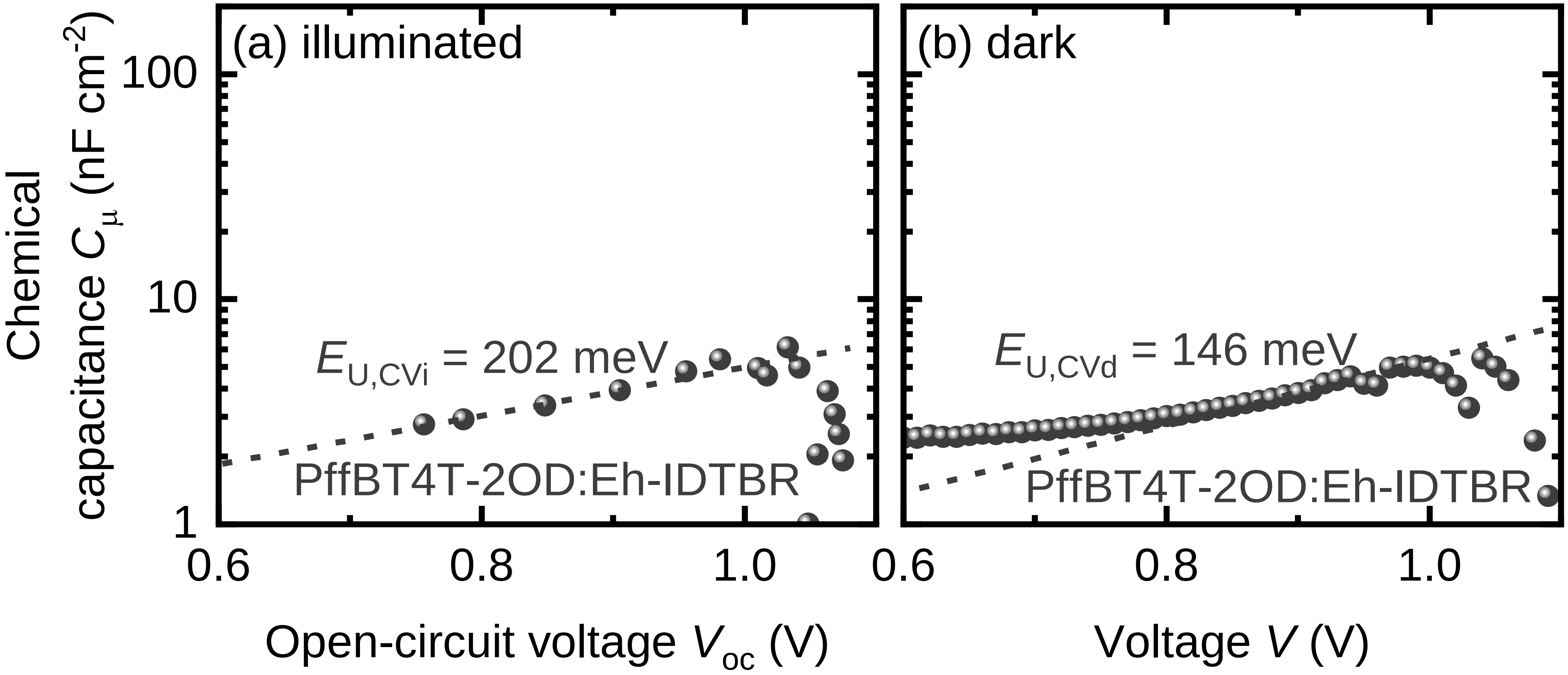


"high" disorder

low disorder

Voltage-dependent measurements

Experimental results

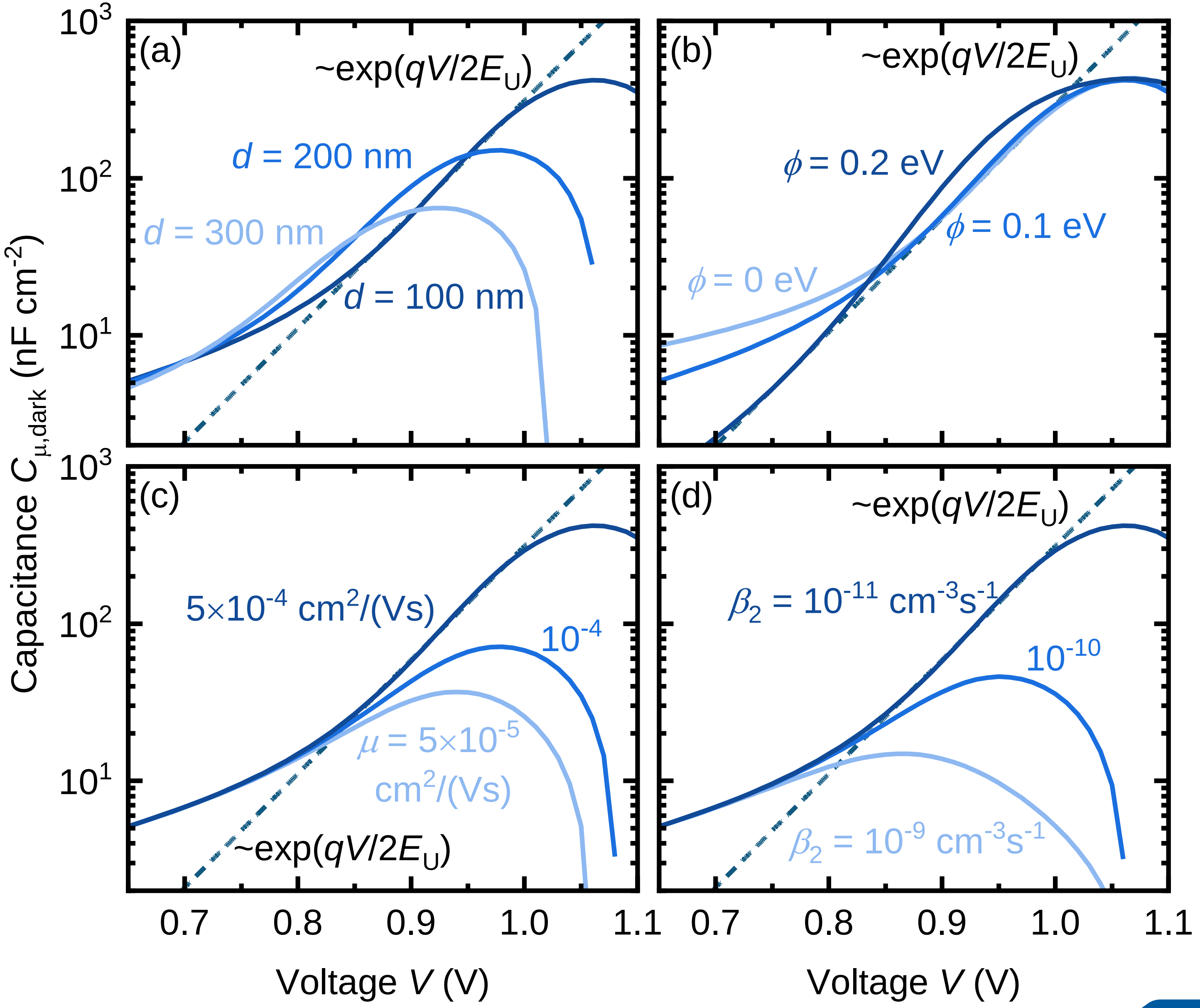


"high" disorder

low disorder

Simulation results

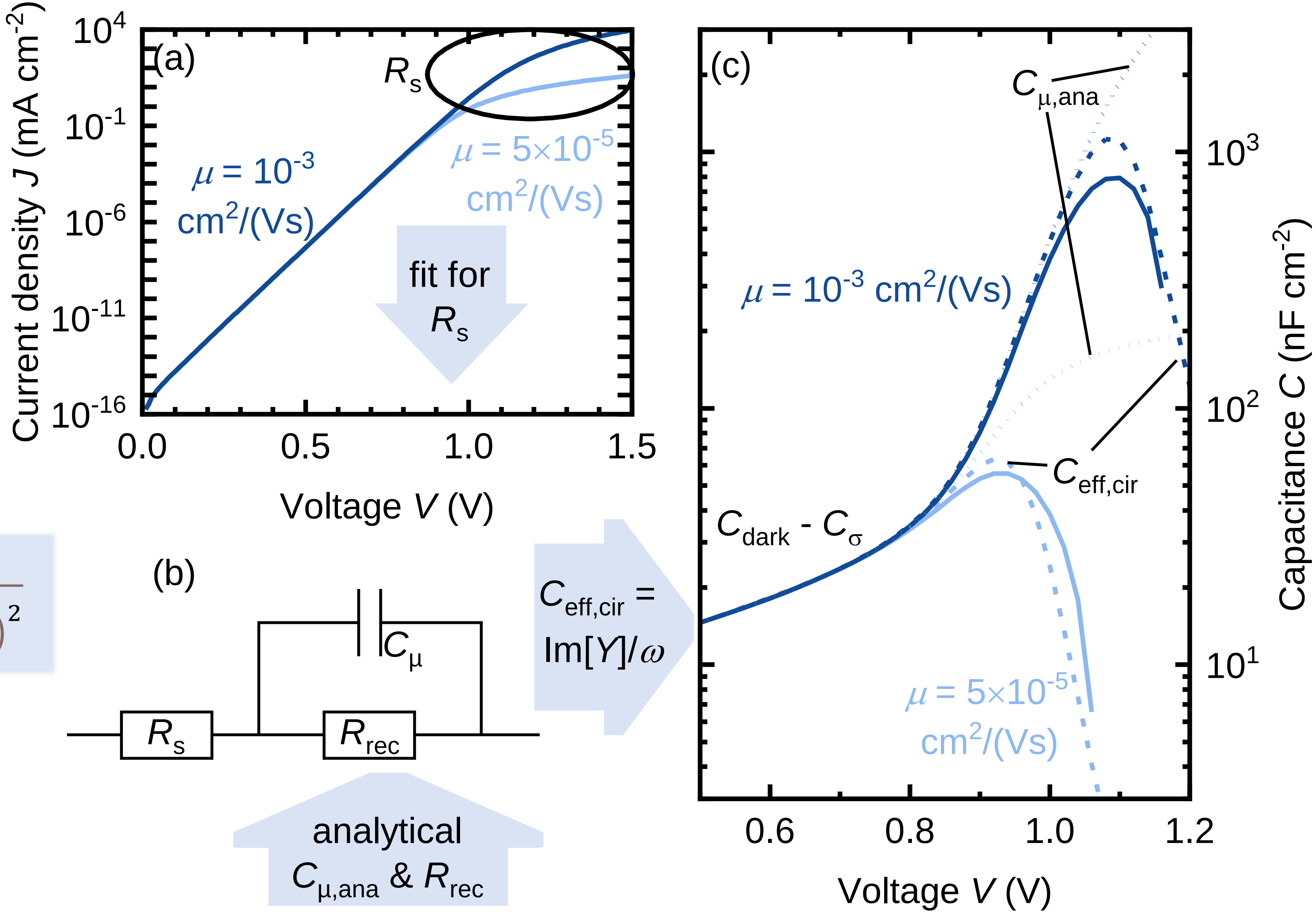
Limitations



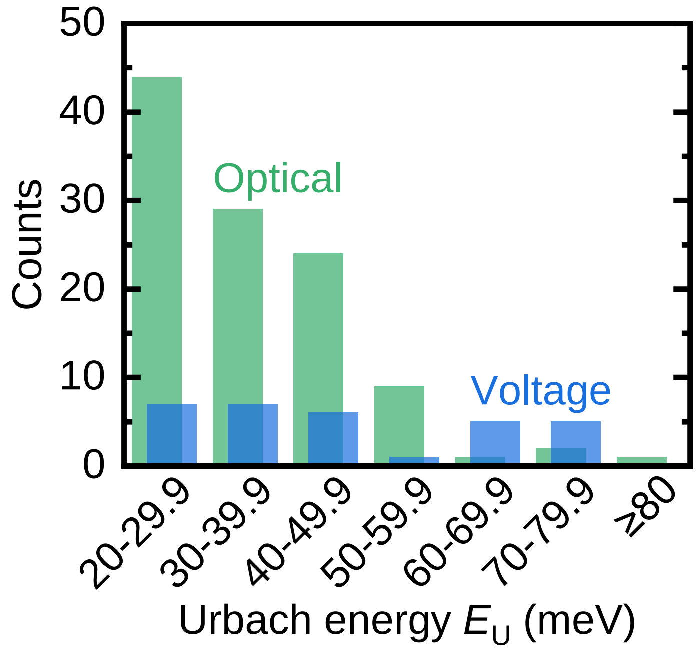
Simulation results

Limitations

$$C_{\mathrm{eff,cir}} = \frac{\mathrm{Im}[Y]}{\omega} = \frac{C_\mu}{\left(\frac{R_\mathrm{s}}{R_\mathrm{rec}} + 1\right)^2 + \left(\omega R_\mathrm{s} C_\mu\right)^2}$$

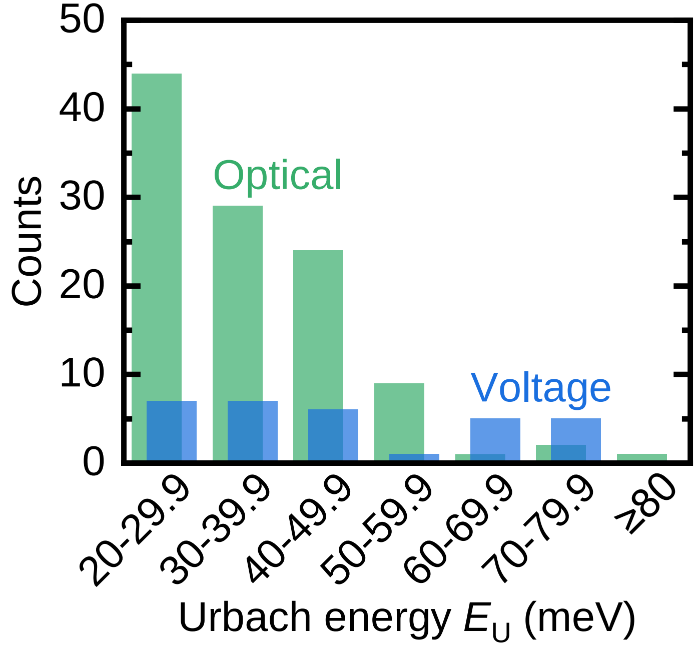


Summary

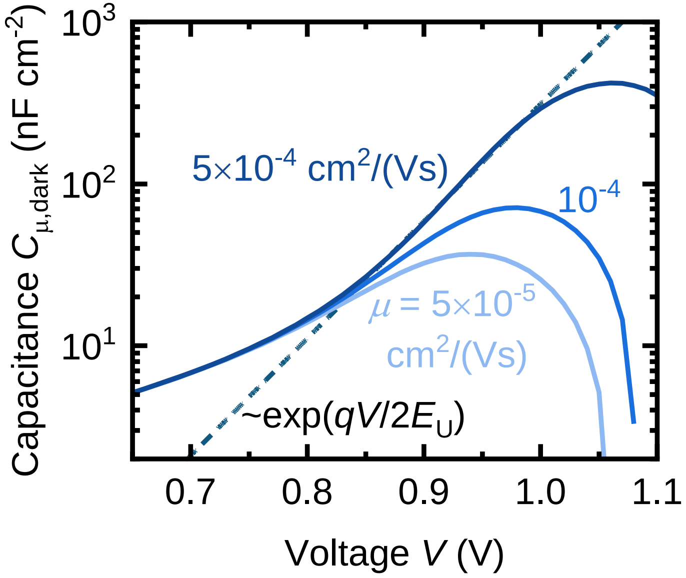


→ Difference in reported disorder between measurement techniques

Summary

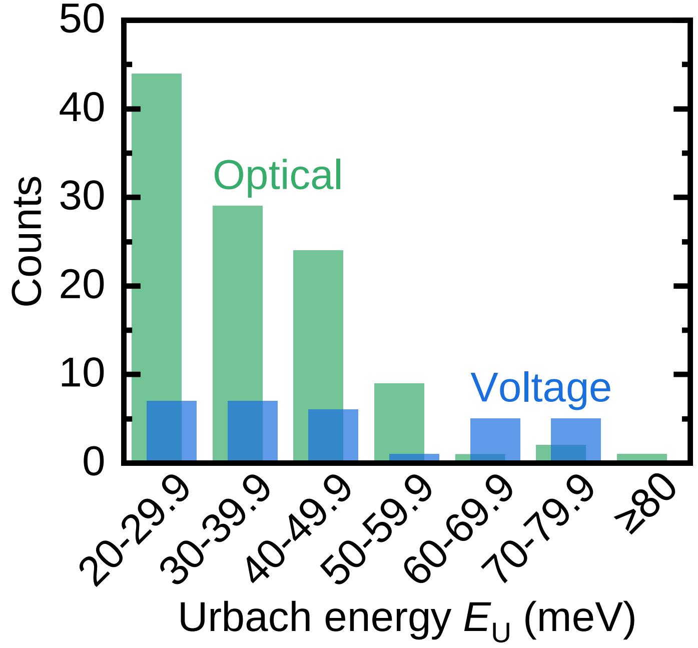


→ Difference in reported disorder between measurement techniques

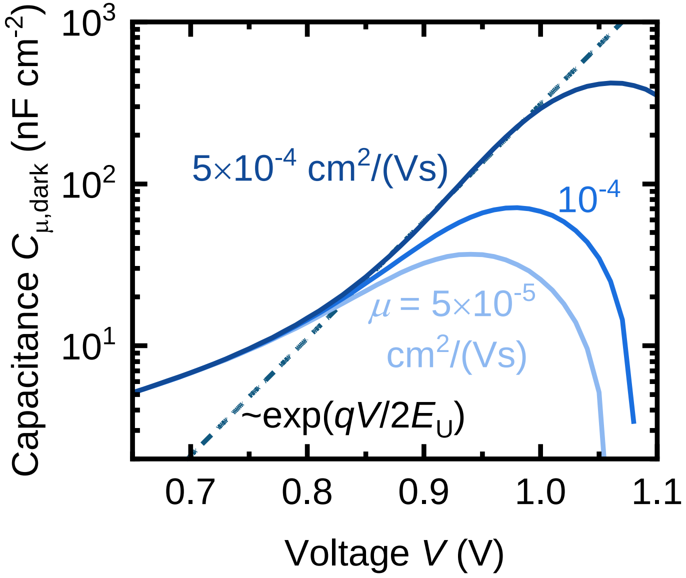


→ Different sensitivity to bad device parameters

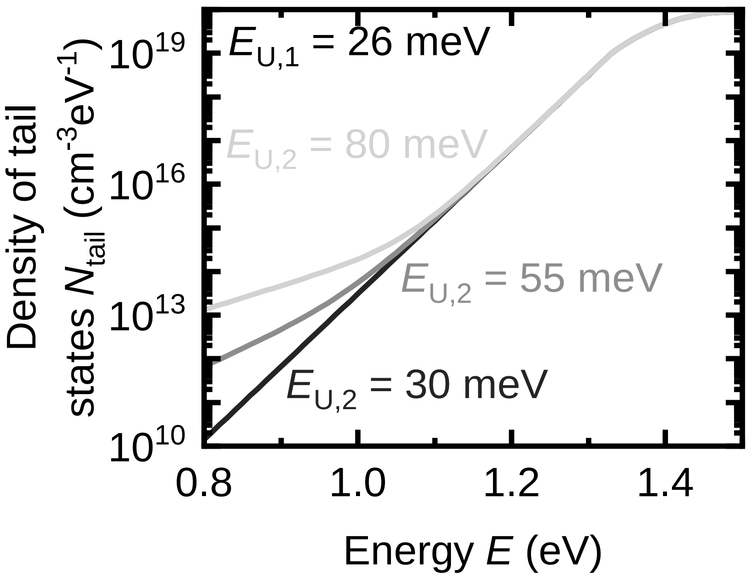
Summary



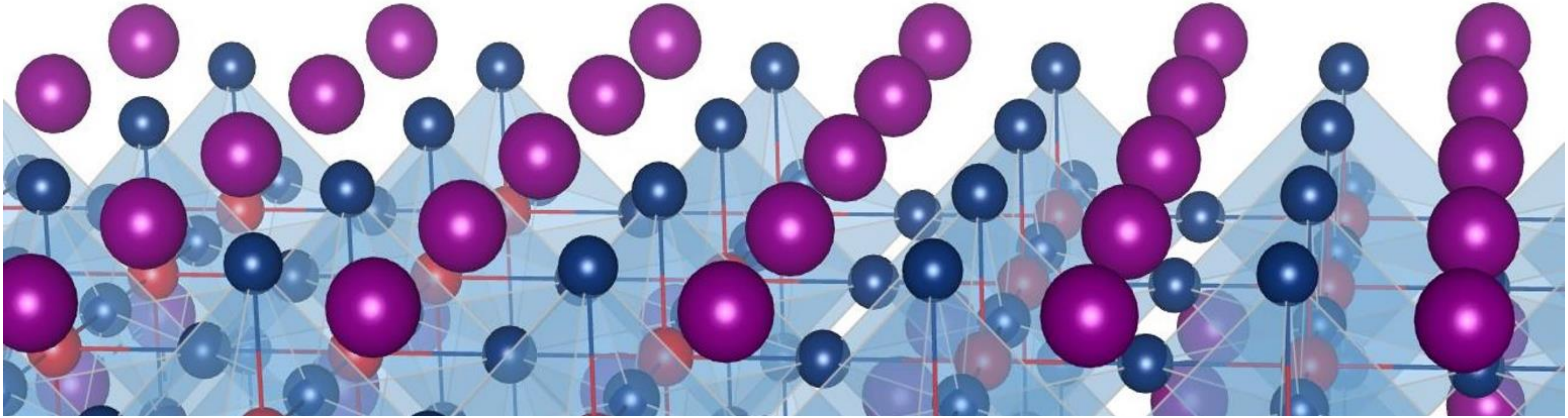
→ Difference in reported disorder between measurement techniques



→ Different sensitivity to bad device parameters



→ Different measurement regimes



Part 2: Inference of Electronic Properties

Thickness and Light Intensity Dependent Data

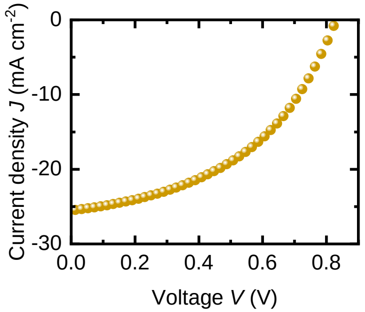
HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

JÜLICH Forschungszentrum

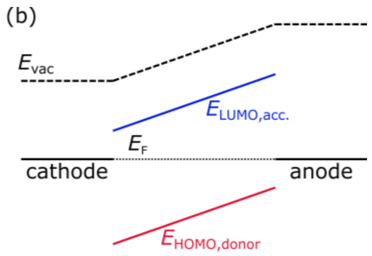
Bayesian Inference

Method

Experimental Data



Device model

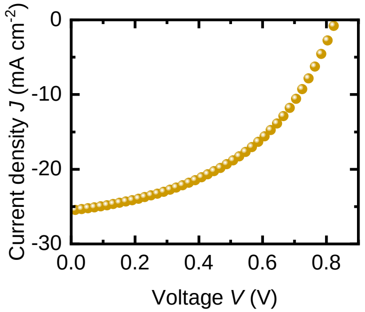



```
variables: mu, tau, …
lower boundaries: 1e-9, 1e-7, …
upper boundaries: 1e-6, 1e-5 , …
```

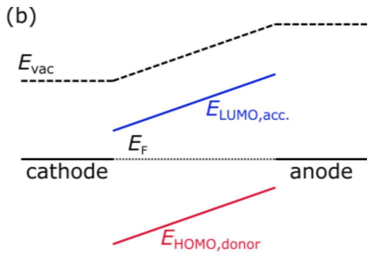
Bayesian Inference

Method

Experimental Data



Device model




```
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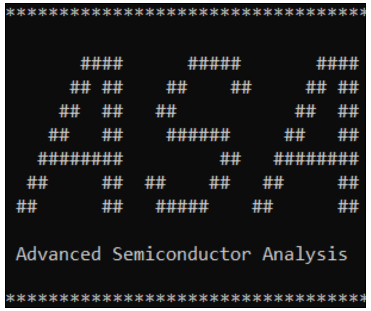
```

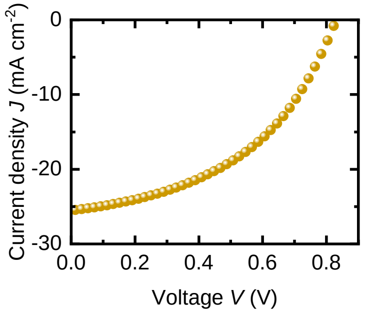
Generate Training Data



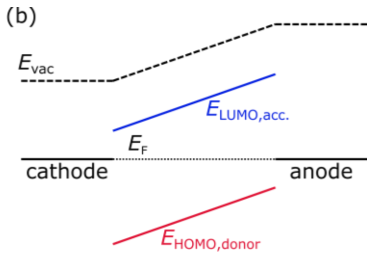
Bayesian Inference

Method

Experimental Data



Device model

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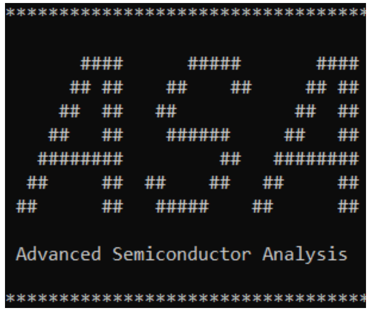
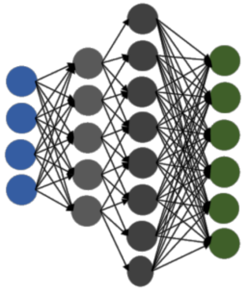
Generate Training Data

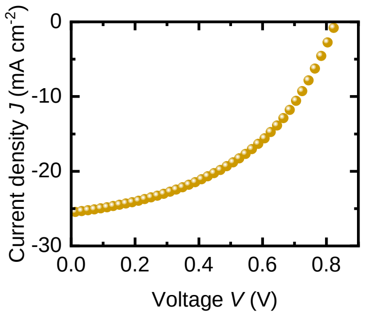


Train Neural Network (NN)

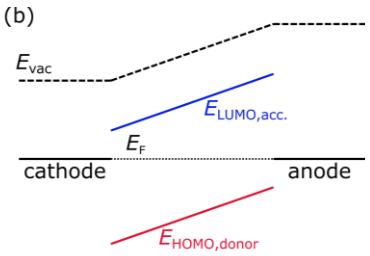


Bayesian Inference
Method

Experimental Data



Device model

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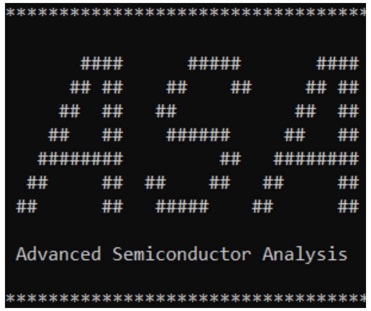

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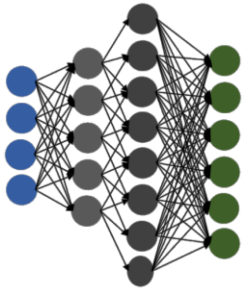
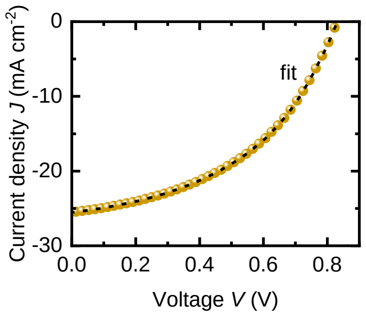
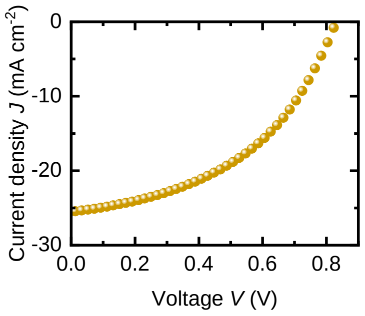


Find Best Fit

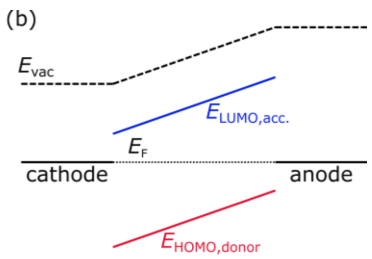


Bayesian Inference
Method

Experimental Data



Device model

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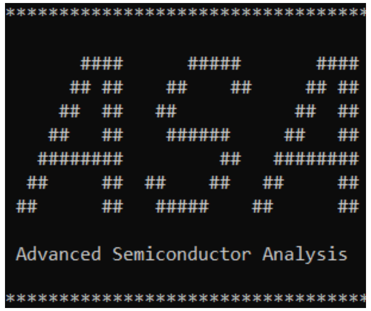

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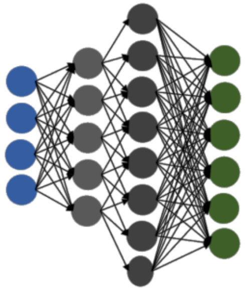

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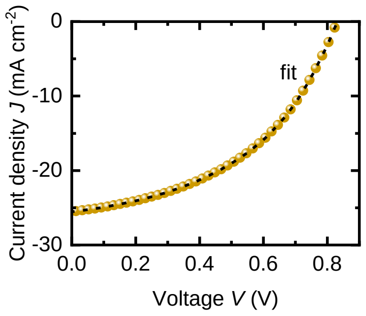
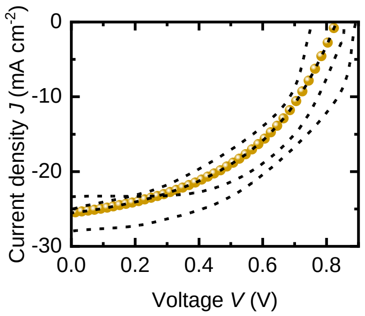


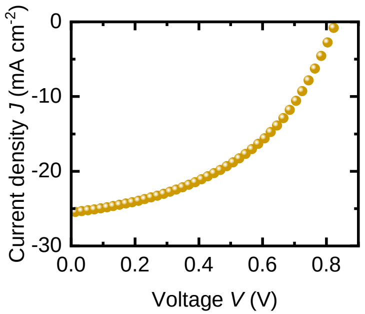
Sample Parameter Space



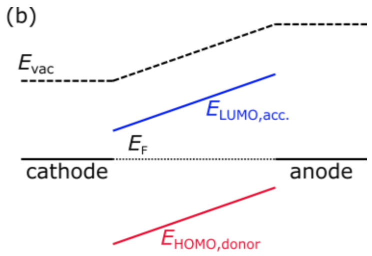
Bayesian Inference

Method

Experimental Data

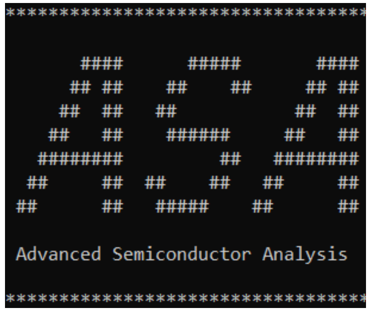


Device model

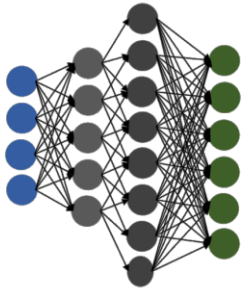



```
variables: mu, tau, …
lower boundaries: 1e-9, 1e-7, …
upper boundaries: 1e-6, 1e-5 , …
```

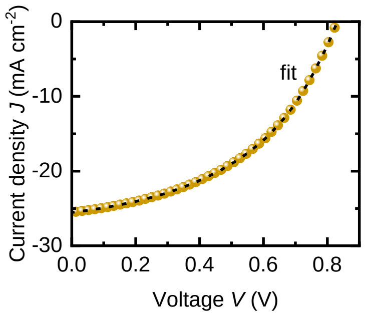
Generate Training Data



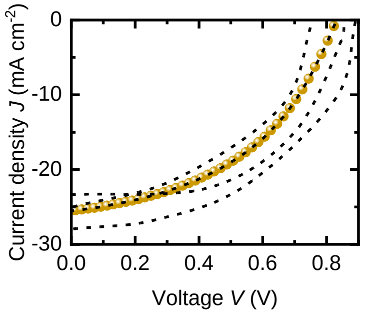
Train Neural Network (NN)



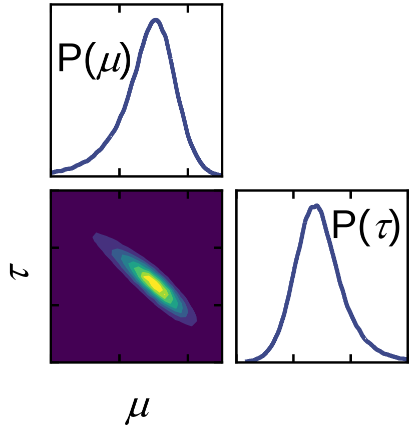
Find Best Fit



Sample Parameter Space



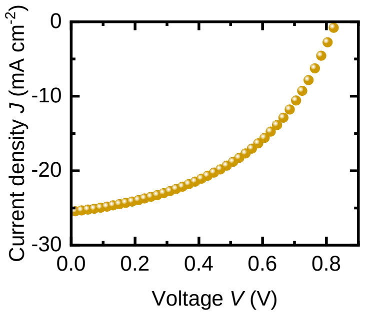
Calculate Probabilities



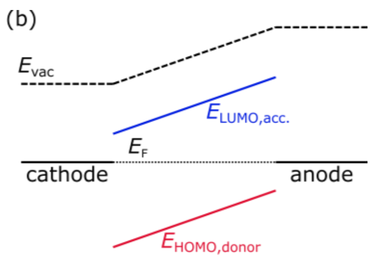
Bayesian Inference

Method

Experimental Data



Device model

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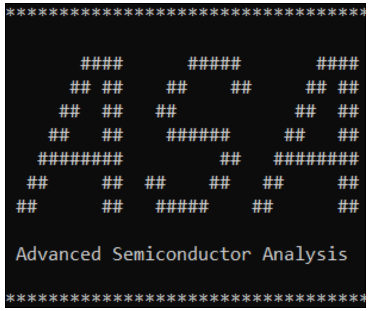
```

Generate Training Data



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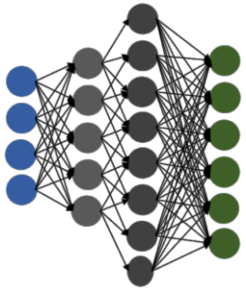

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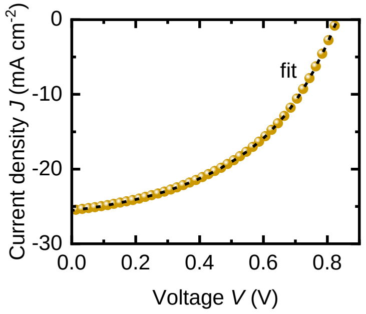
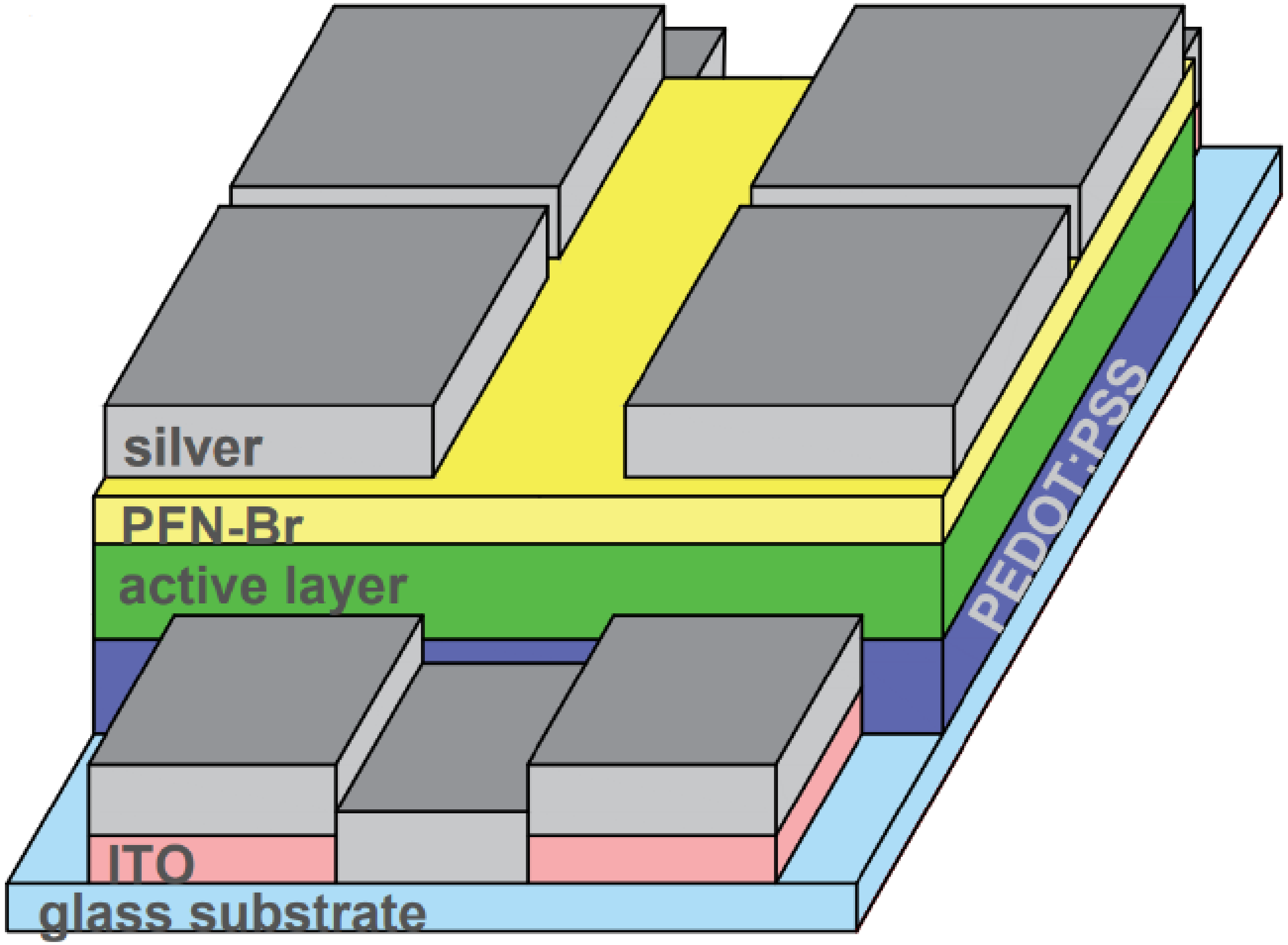


Sample Parameter Space

Calculate Probabilities

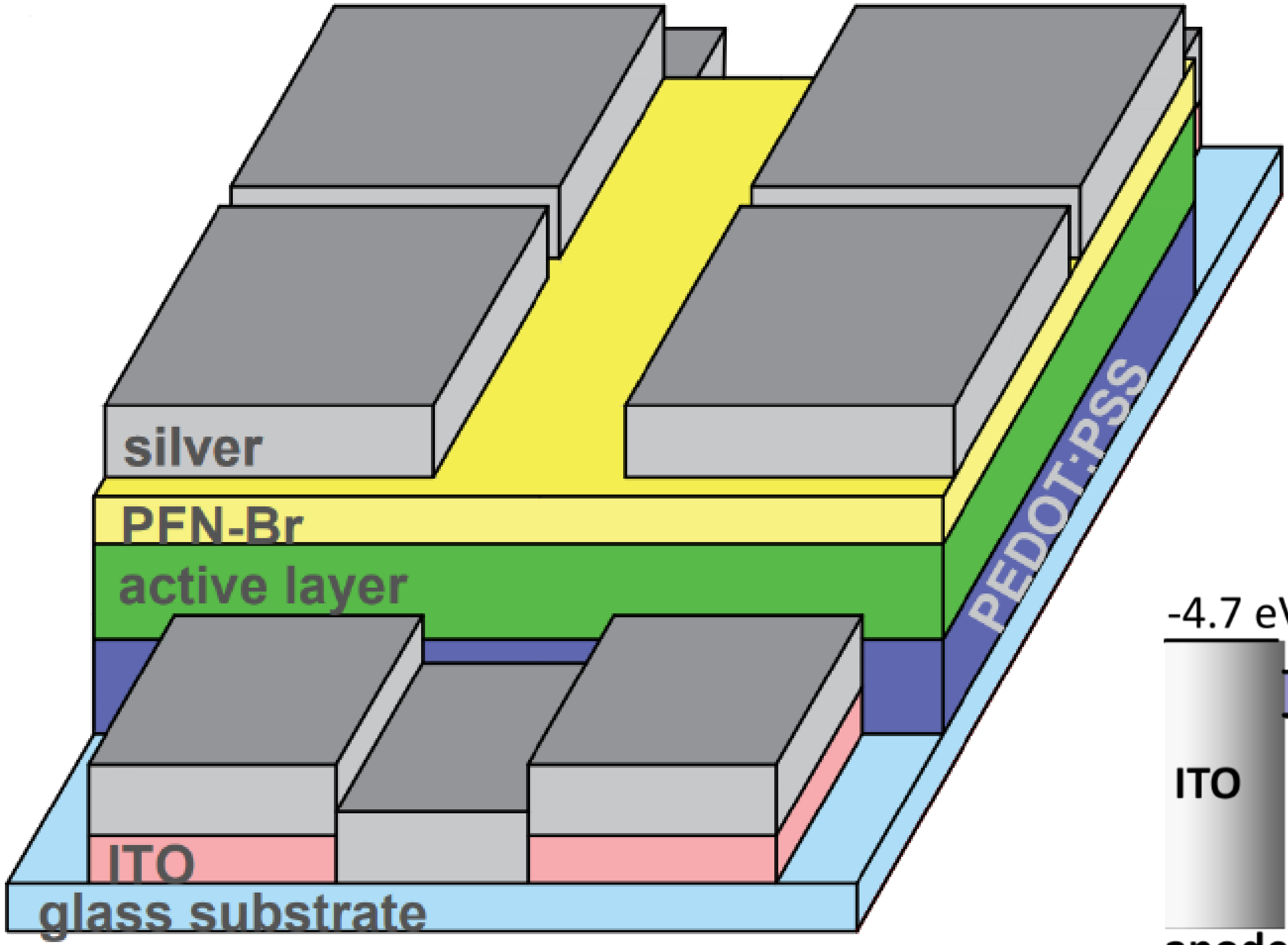
Experimental Data

Device Architecture



Experimental Data

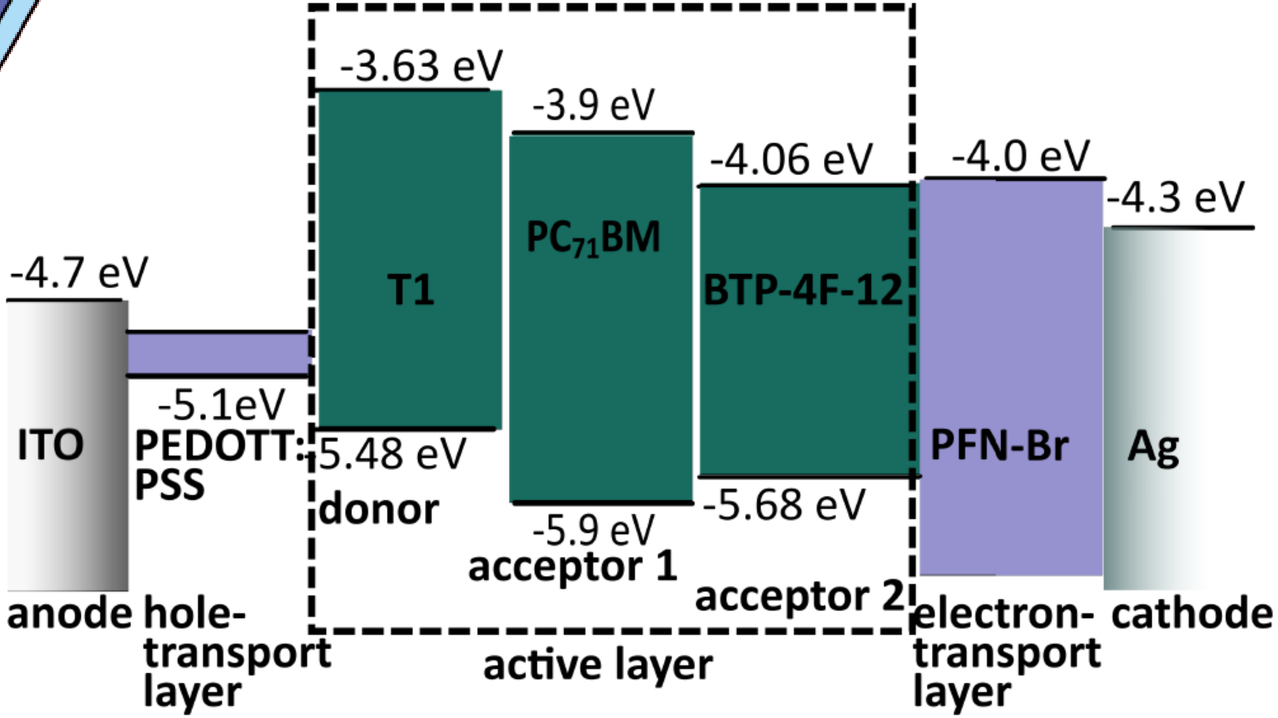
Device Architecture



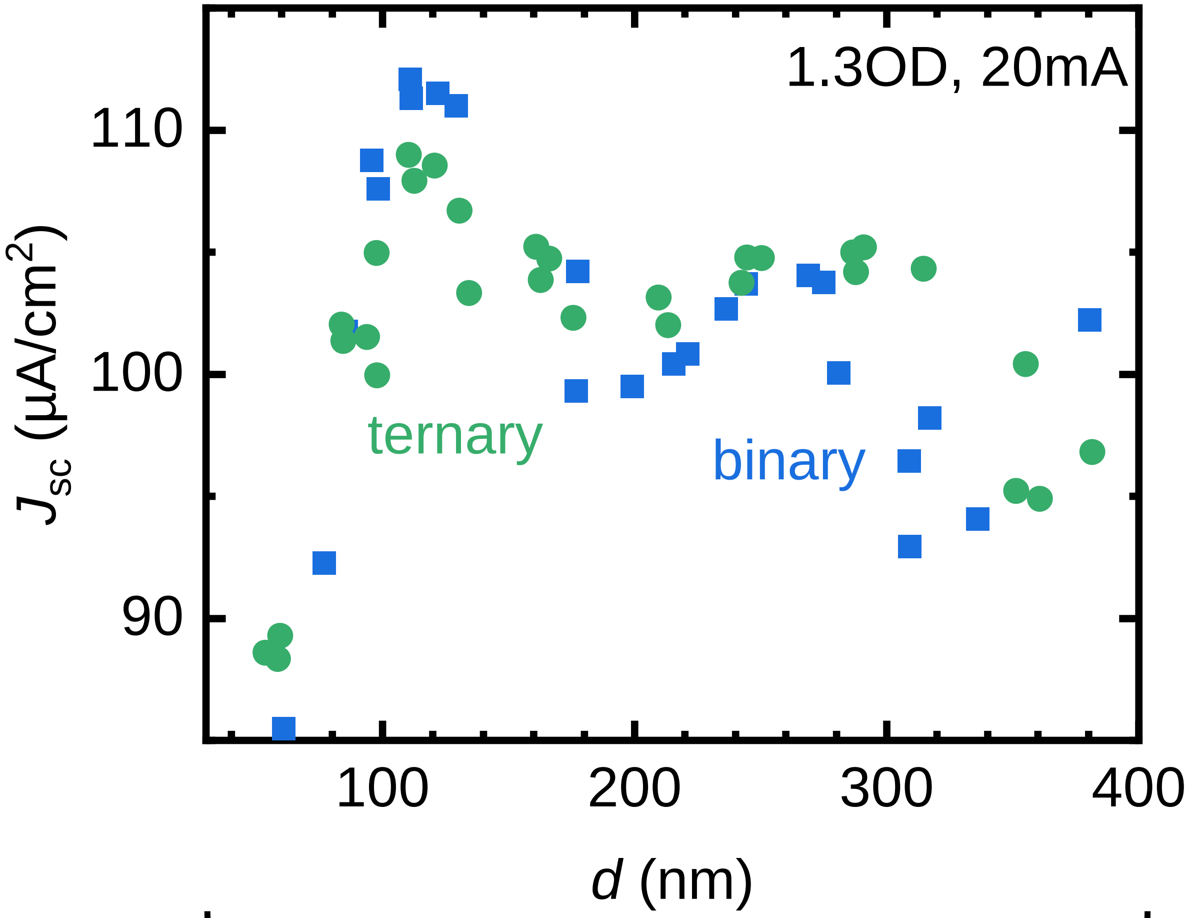
Cell area: 0.16 cm²

Active layers:

- Binary blend (BTP-4F-12: PBDB-TF-T1)
- Ternary blend (BTP-4F-12: PBDB-TF-T1:PC71BM)

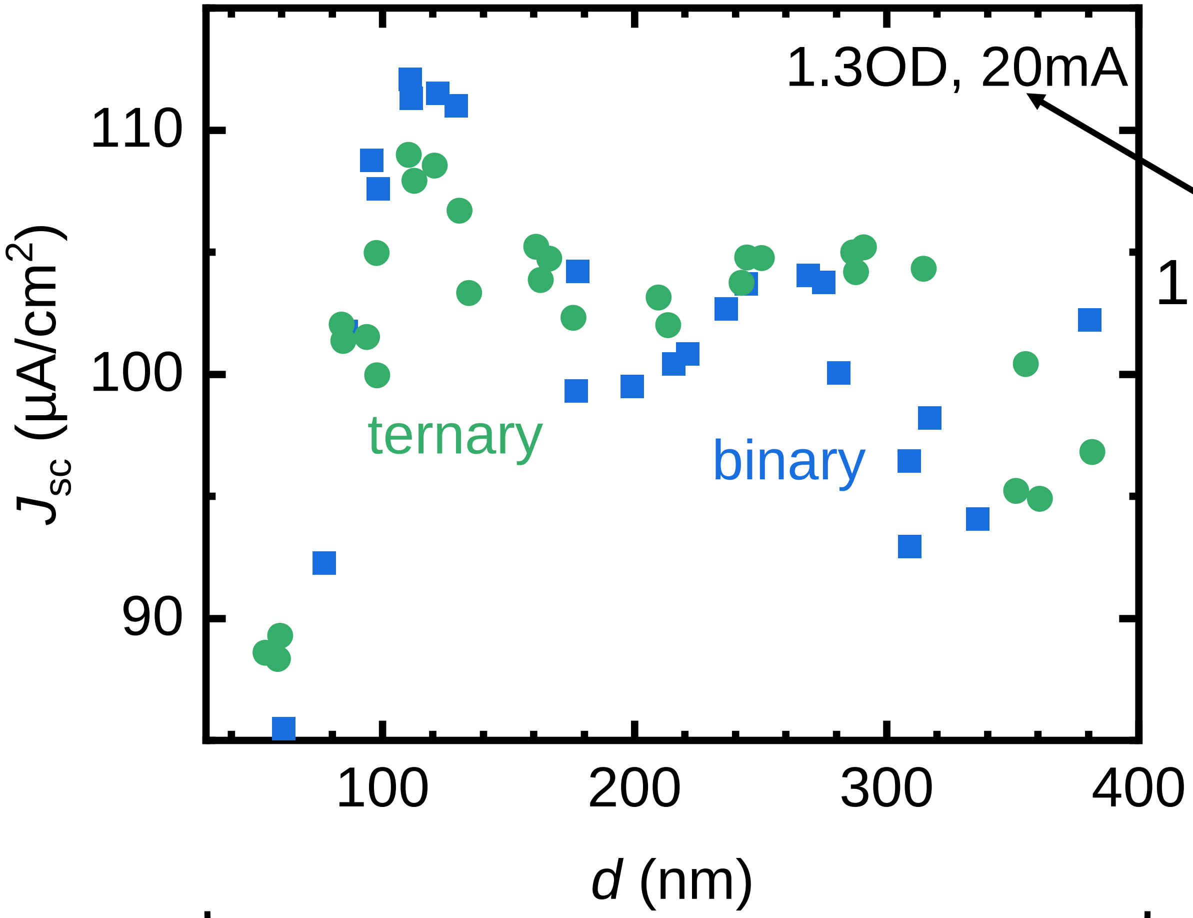


Experimental Data



Wide range of thicknesses

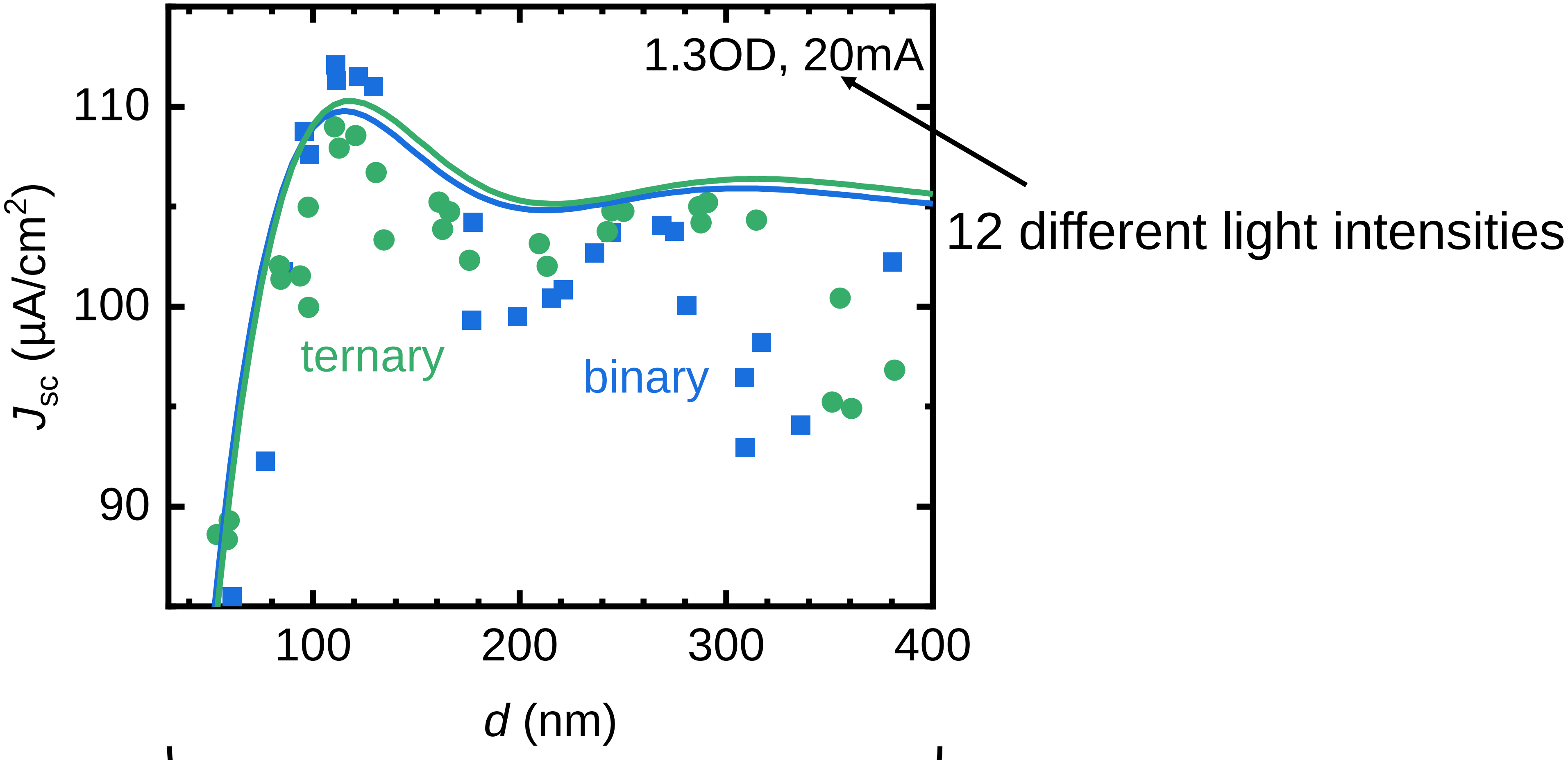
Experimental Data



12 different light intensities

Wide range of thicknesses

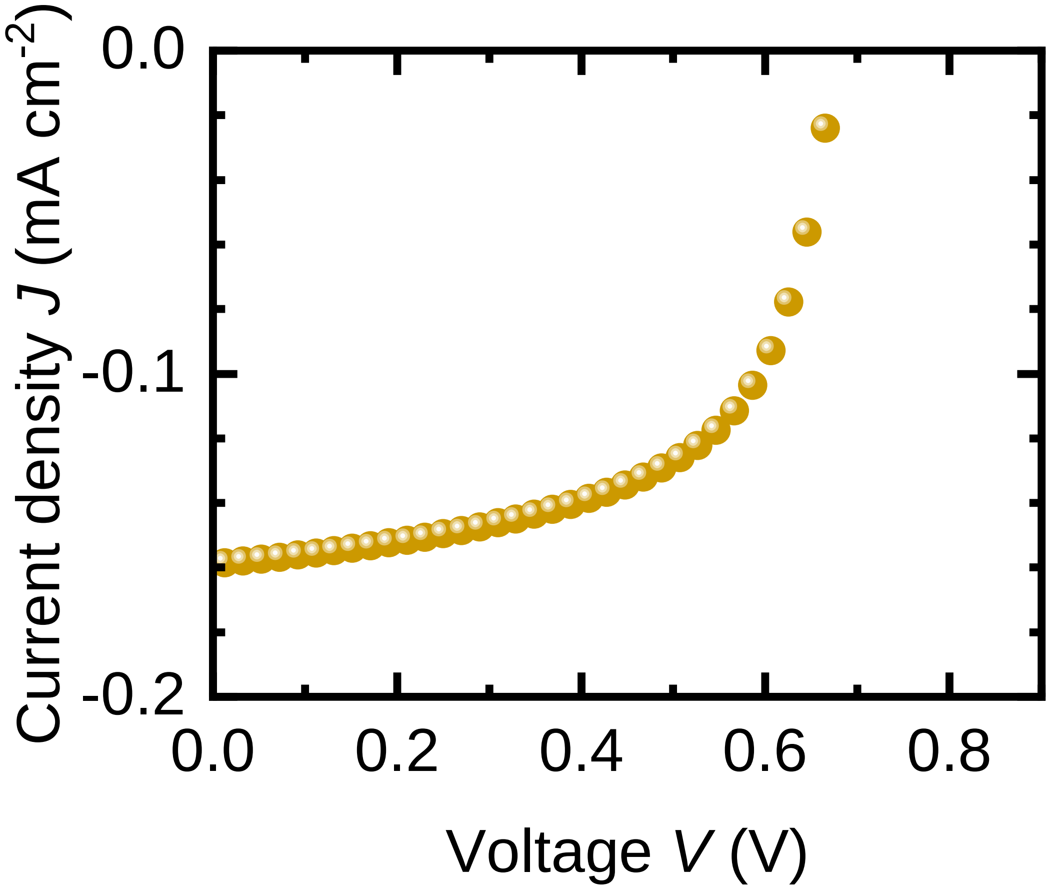
Experimental Data



Wide range of thicknesses

Experimental Data

What Do We Fit?

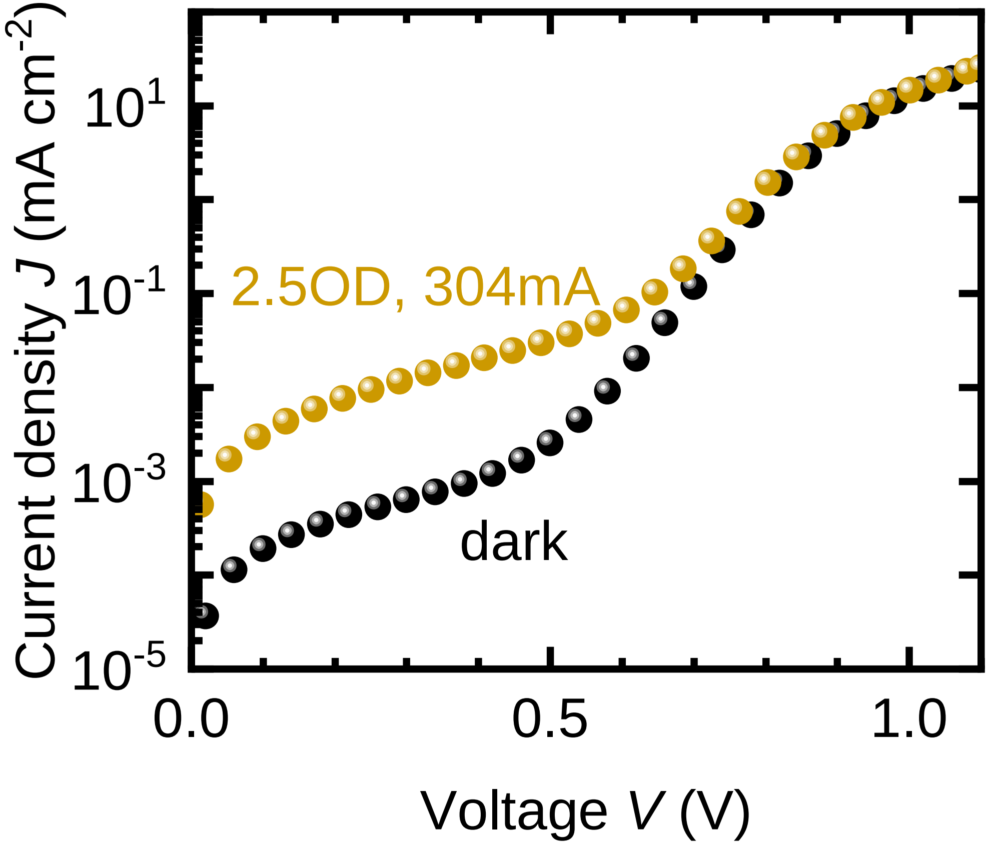


$$\Delta J_{\mathrm{rec}}(V,\Phi) = J_{\mathrm{illu}}(V,\Phi) - J_{\mathrm{sc}}(\Phi)$$

Experimental Data

What Do We Fit?

- Independence of inaccuracies in optical model

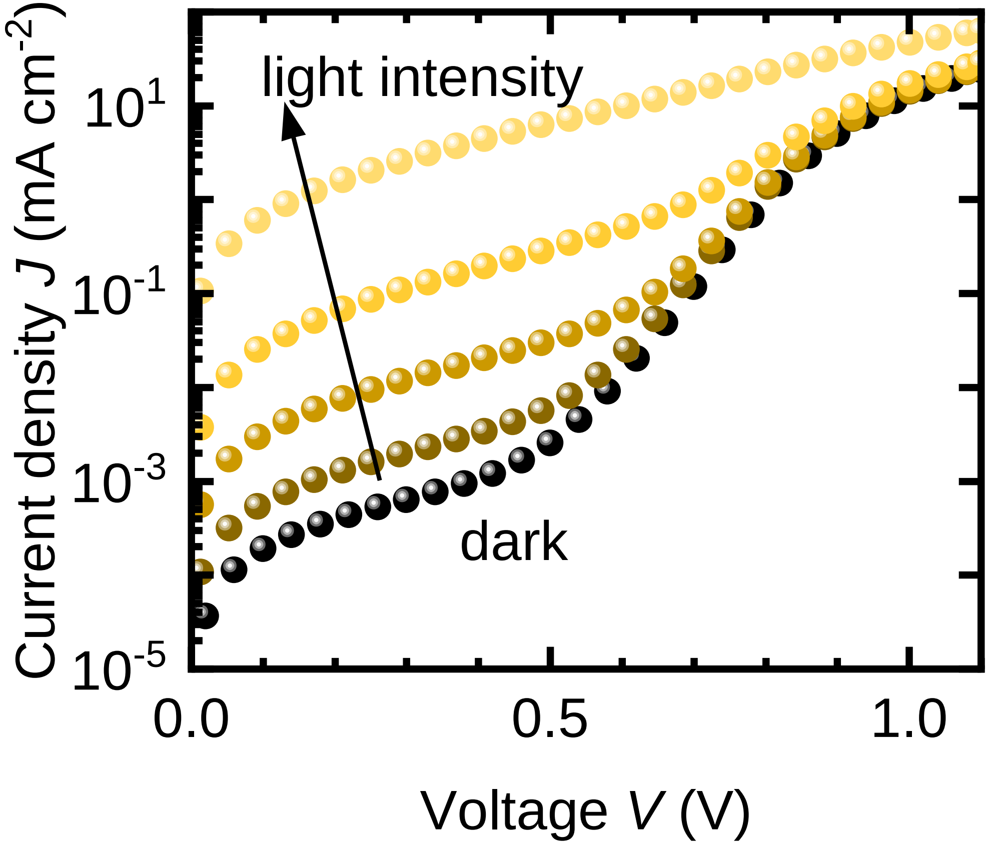


$$\Delta J_{\mathrm{rec}}(V,\Phi) = J_{\mathrm{illu}}(V,\Phi) - J_{\mathrm{sc}}(\Phi)$$

Experimental Data

What Do We Fit?

- Independence of inaccuracies in optical model
- Emphasis on light-intensity dependent recombination current density

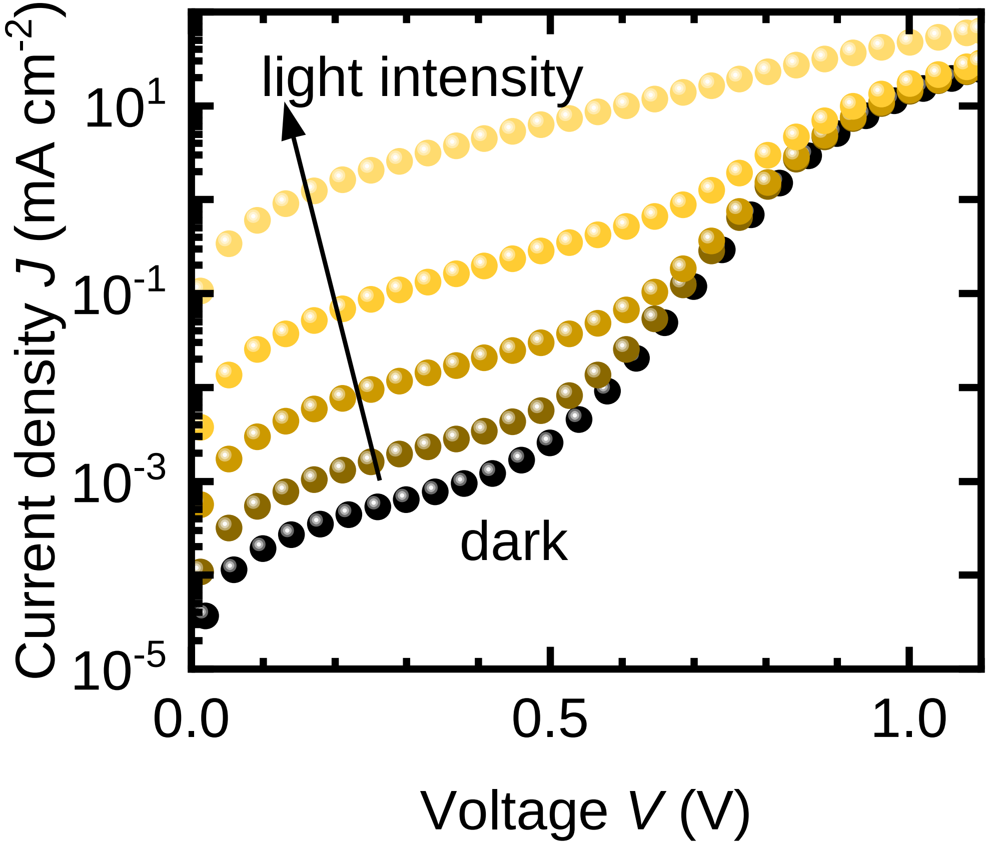


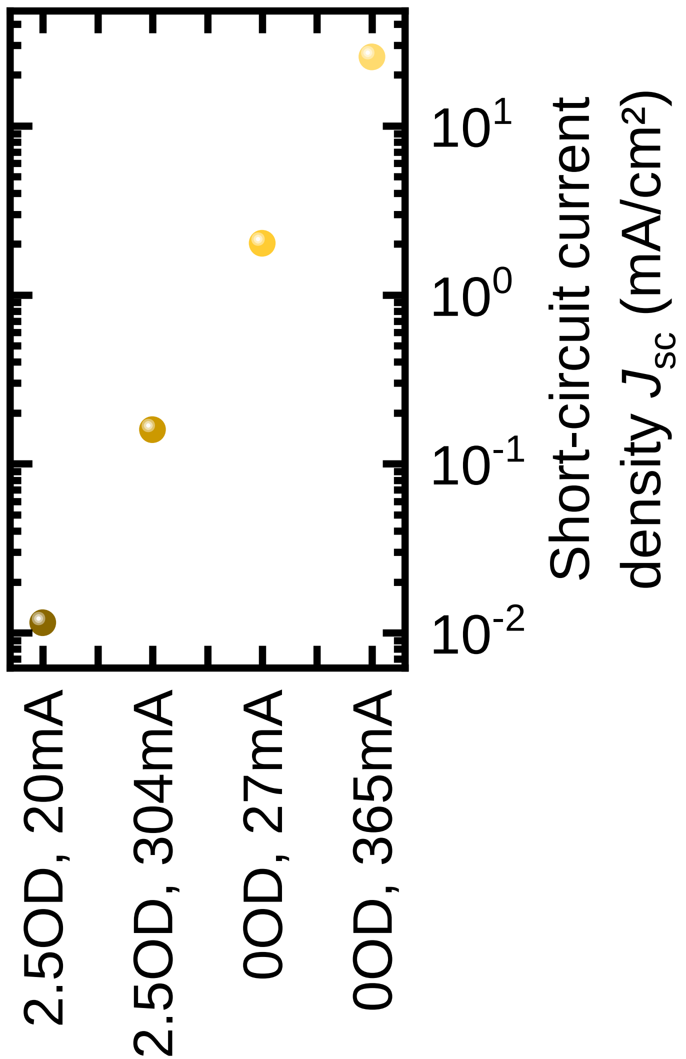
$$\Delta J_{\mathrm{rec}}(V,\Phi) = J_{\mathrm{illu}}(V,\Phi) - J_{\mathrm{sc}}(\Phi)$$

Experimental Data

What Do We Fit?

- Independence of inaccuracies in optical model
- Emphasis on light-intensity dependent recombination current density
- Use *Jsc* in fitting procedure to penalize fits that are too far off





$$\Delta J_{\mathrm{rec}}(V,\Phi) = J_{\mathrm{illu}}(V,\Phi) - J_{\mathrm{sc}}(\Phi)$$

Bayesian Inference: The General Idea

Prior

Jon Snow
(„knows nothing“)

Experiment

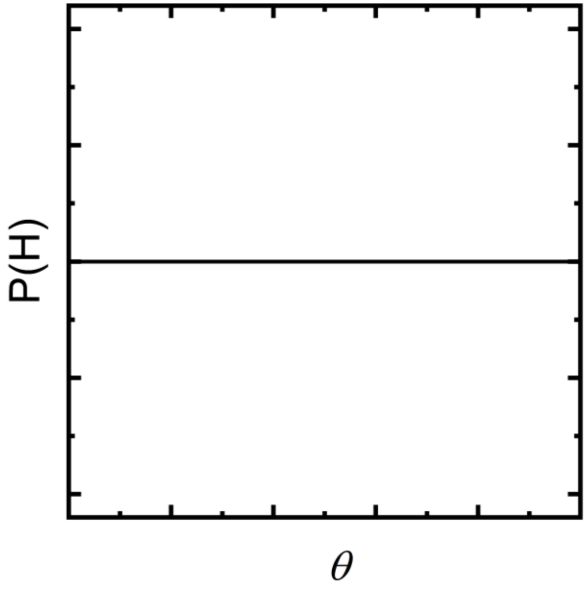
Some explanations
later…

Posterior

Jon Snow
(„knows something“)

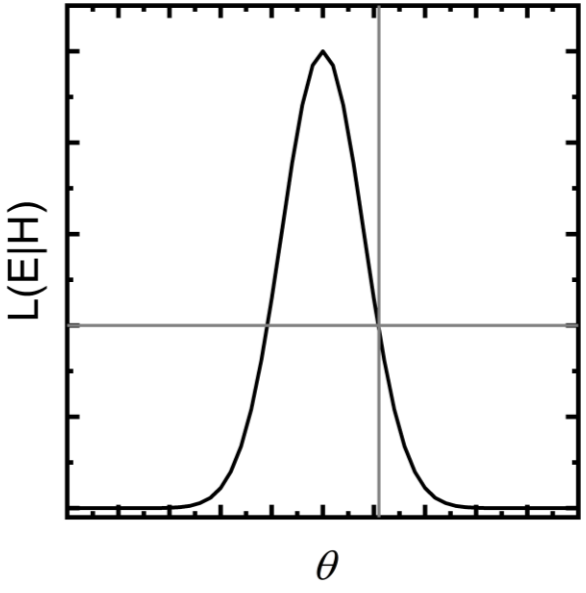
Bayesian Inference: The General Idea

Prior



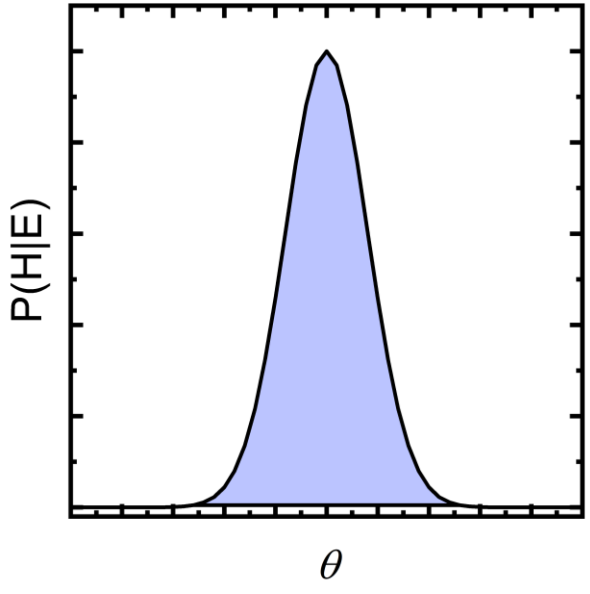
Jon Snow
(„knows nothing“)

Experiment



Some explanations later…

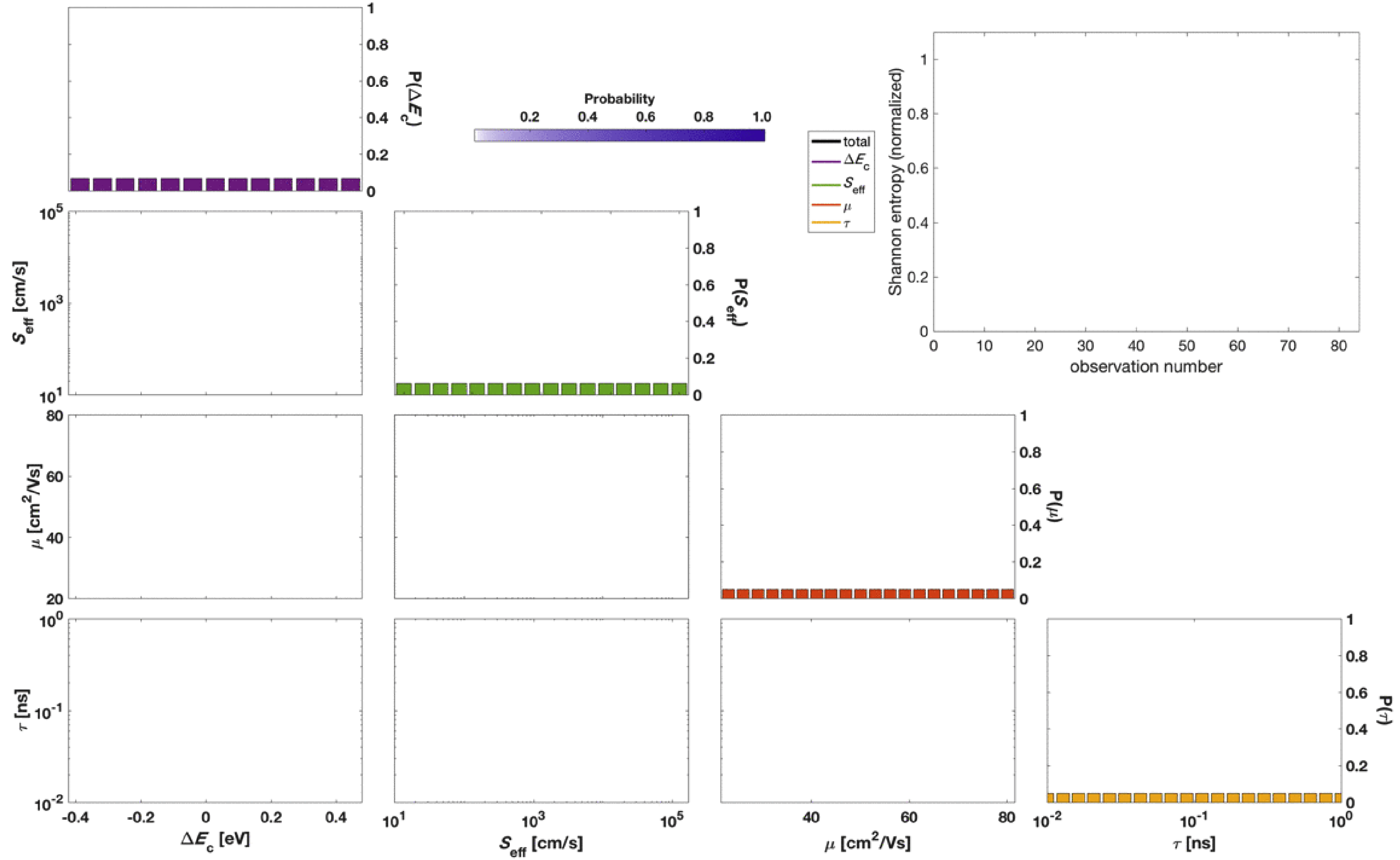
Posterior



Jon Snow
(„knows something“)

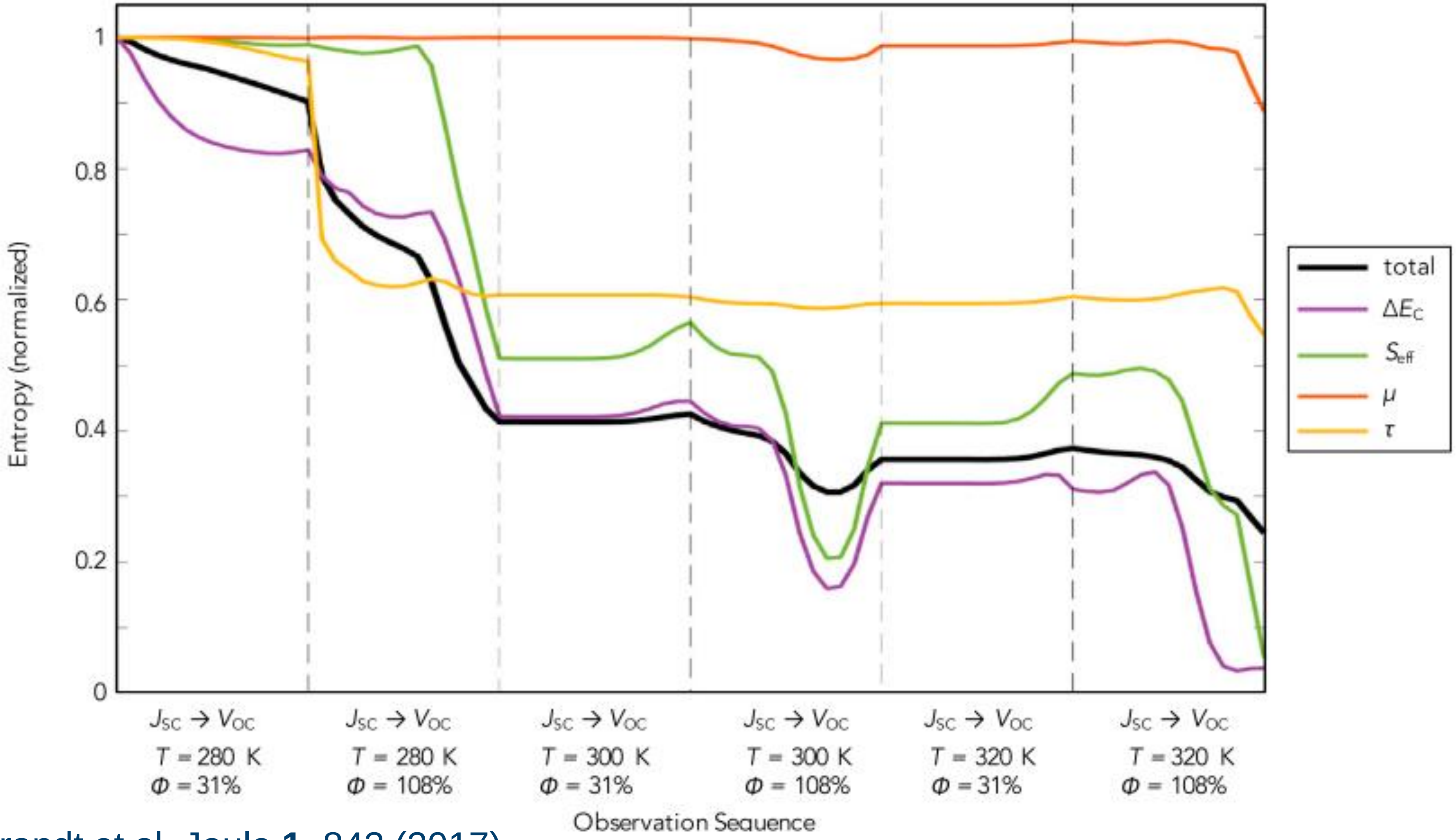
Example from Literature: SnS Solar Cells

Corner Plots and Information Entropy



Brandt et al. Joule **1**, 843 (2017)

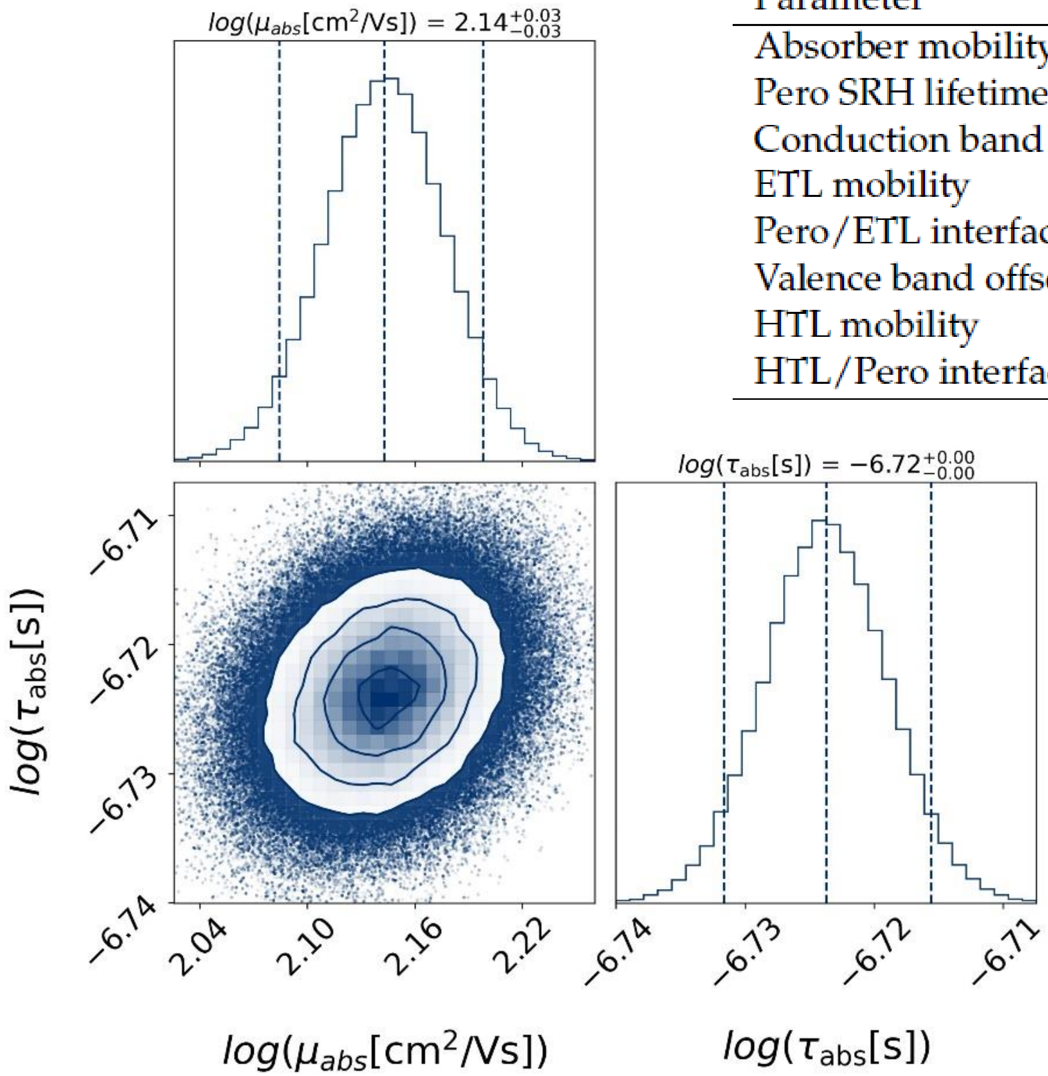
Which Experiments Identify Which Parameter?



Brandt et al. Joule **1**, 843 (2017)

(mini) Cornerplot for some Perovskite Data

Parameter	Symbol	Value
Absorber mobility	μ_{pero}	$138\ \text{cm}^2/\text{Vs}$
Pero SRH lifetime	$\tau_{SRH}(\text{Pero})$	$1.8 \times 10^{-7}\ \text{s}$
Conduction band offset	ΔE_{C}	15 meV
ETL mobility	μ_{ETL}	$0.1\ \text{cm}^2/\text{Vs}$
Pero/ETL interface recombination velocity	S_{ETL}	35.7 cm/s
Valence band offset	ΔE_{V}	39 meV
HTL mobility	μ_{HTL}	$1.3 \times 10^{-4}\ \text{cm}^2/\text{Vs}$
HTL/Pero interface recombination velocity	S_{HTL}	2 cm/s

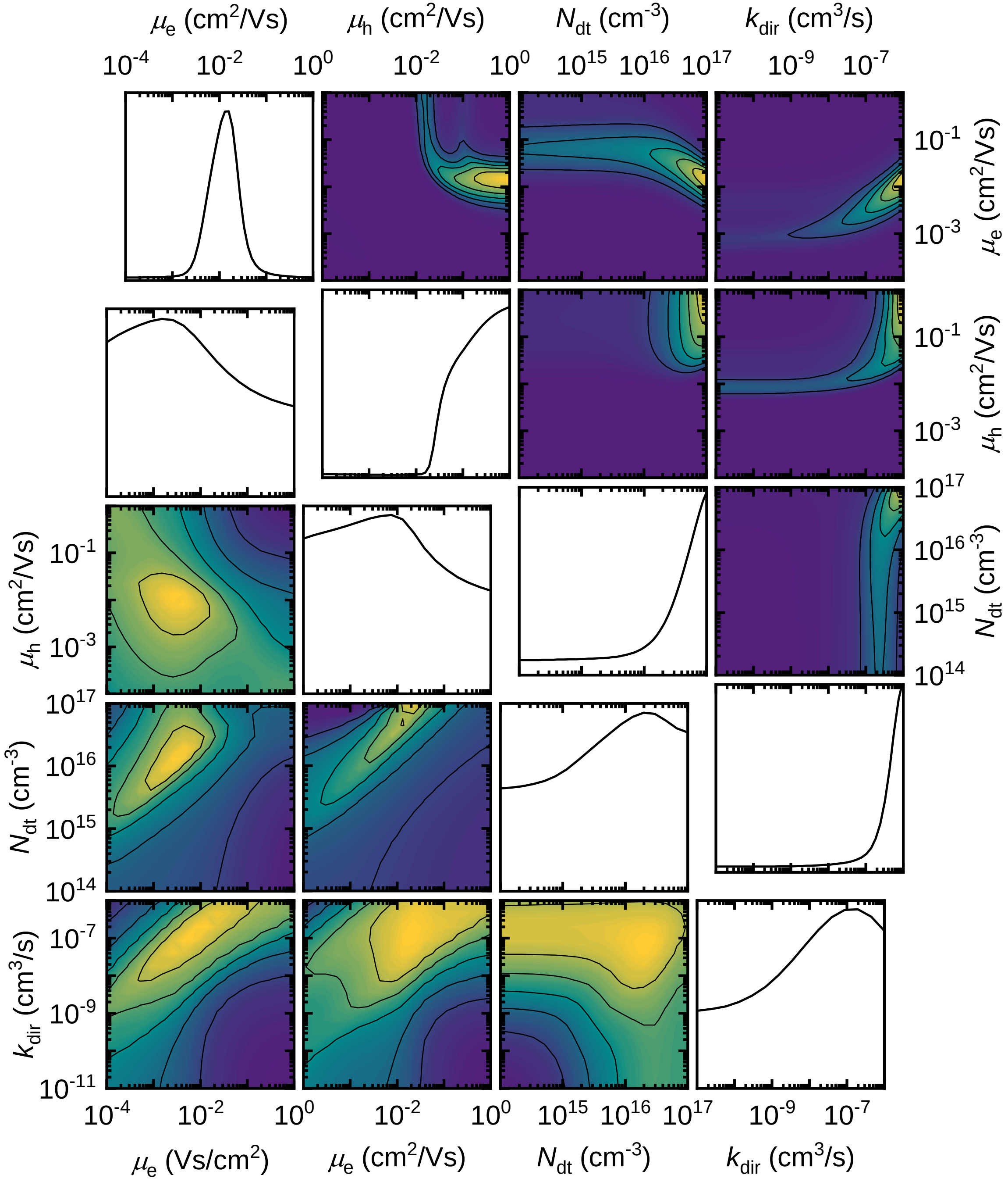


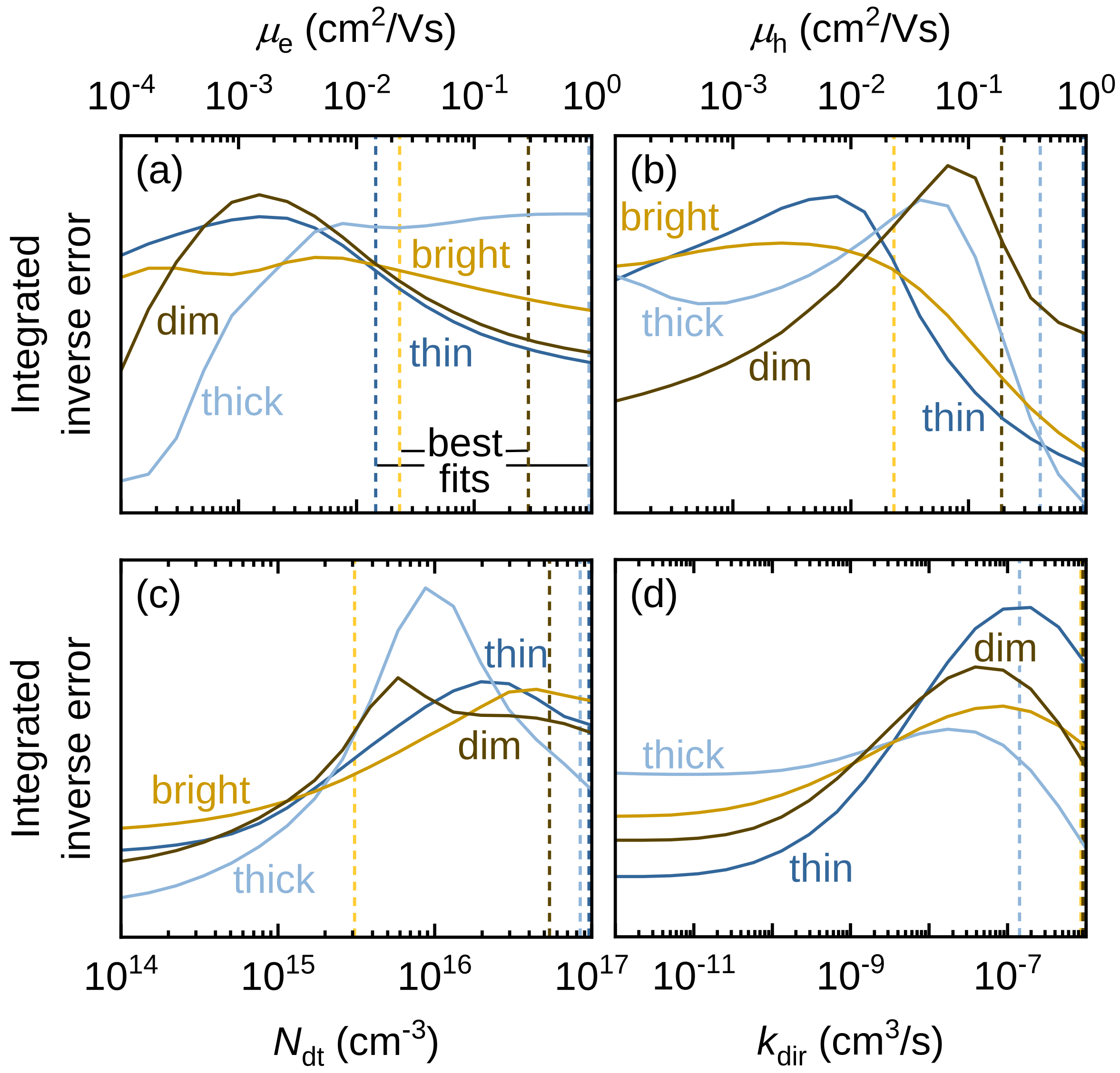
$$\tau = 180\text{ns}$$

$$\mu_{\text{abs}} = 138\text{cm}^2 / \text{Vs}$$

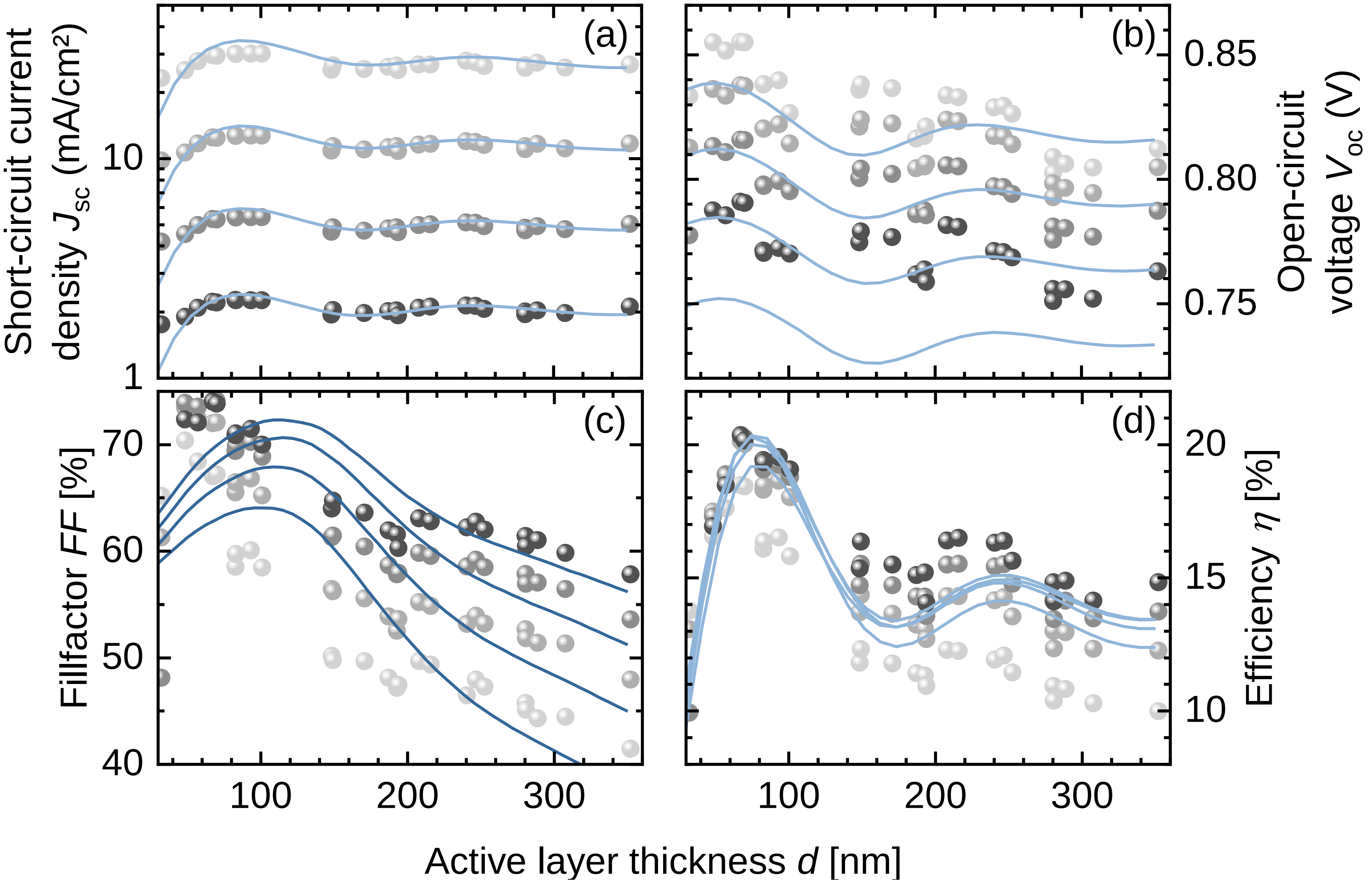
Problems with MCMC

Cornerplots





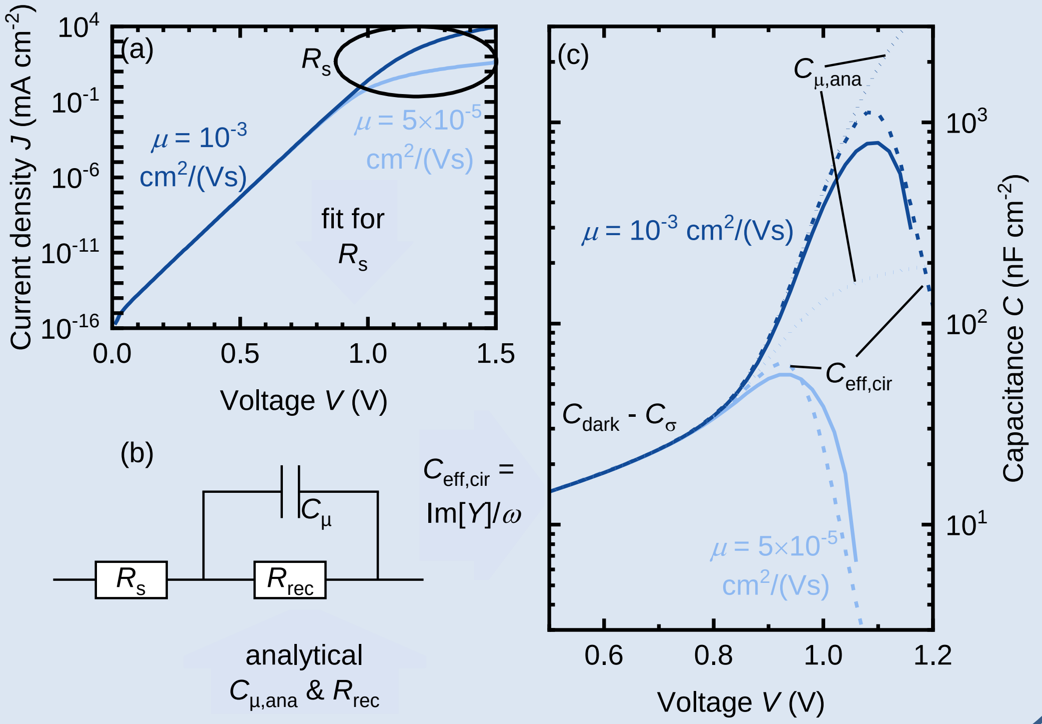
Trends with Thickness



Conclusions

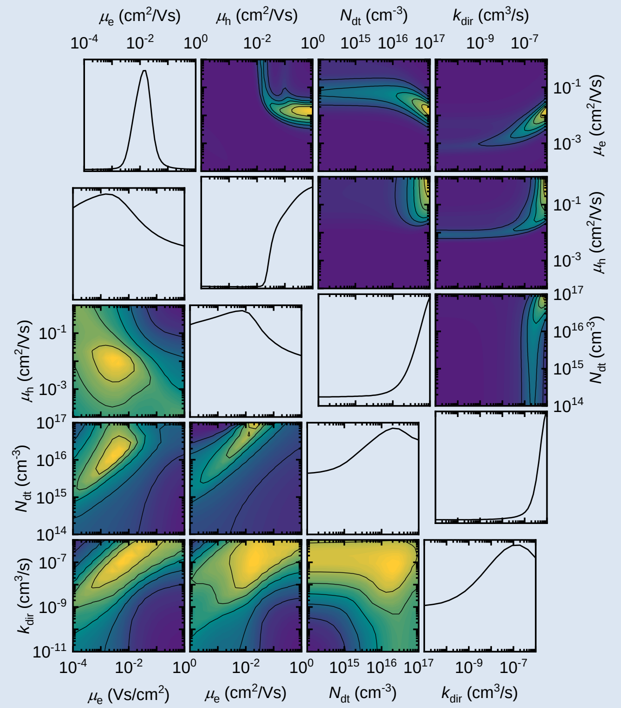
1) Measuring Disorder

- Differences in optical and electrical methods
- Electrical methods often affected by resistive effects



2) Inferring Parameters

- NNs interpolate well and look up well
- Trained NNs can tehrefore accelerate simulations by a factor ~10^4 for JV curves
- Allows finding likely material parameters



Funding from
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DFG
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Thank you for your attention

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES

JÜLICH Forschungszentrum