Identifying crucial device parameters in emerging photovoltaics: towards a digital twin

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Sino-German workshop, Erlangen 22.05.2024

Grand challenges in emerging PV

Voltage losses
- Level matching at interfaces
- QFLS in bulk

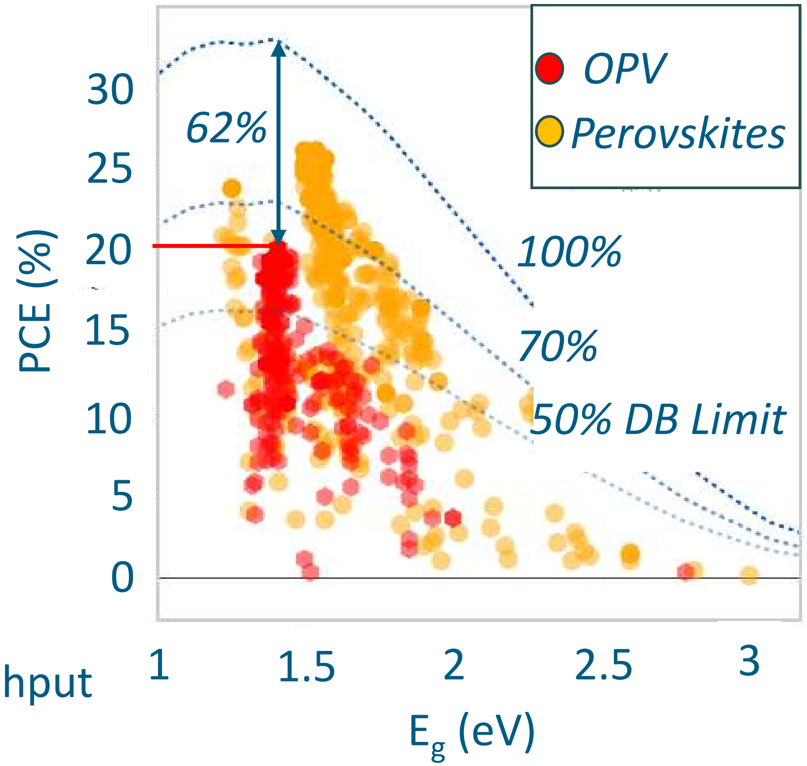
Operational stability:
- Re-orientation at interfaces
- De-mixing in bulk
- Photochemistry

20 % PCE now reached by OPV!

See Guan et al., https://doi.org/10.1002/adma.202400342

Still, large headroom For improvement

Improvement headroom of emerging PV towards DB Limit



emerging-pv.org

Understanding the essential device physics
- **Well established model**: compare two samples
- Model search **along one dimension**: encounter trends
- Model search **along several dimensions**: use machine learning
- Find **needle in haystack** (quasi-infinite dimensionality): towards a digital twin

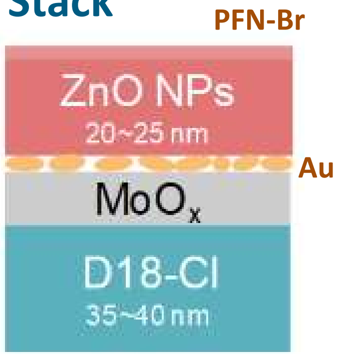
Low throughput

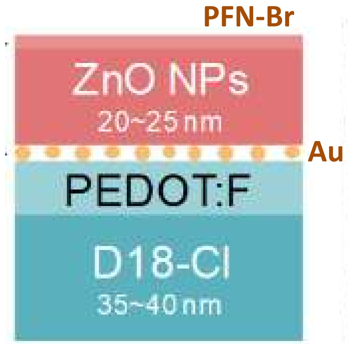
High throughput (one campaign)

Outlook:
Big Data (across campaigns & labs)

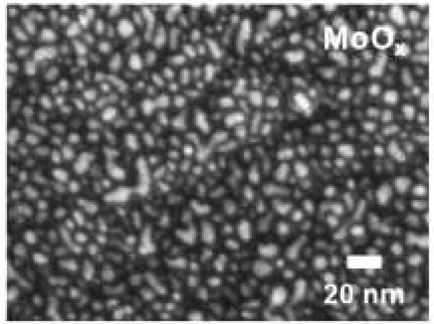
Novel ICL for n-i-p Perovskite-Organic Tandem Solar Cells

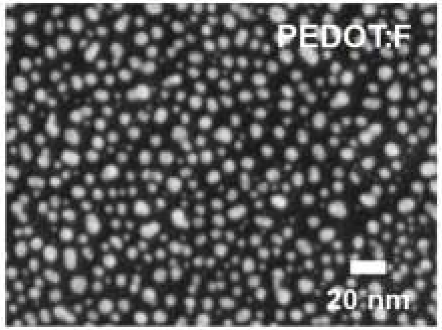
Stack



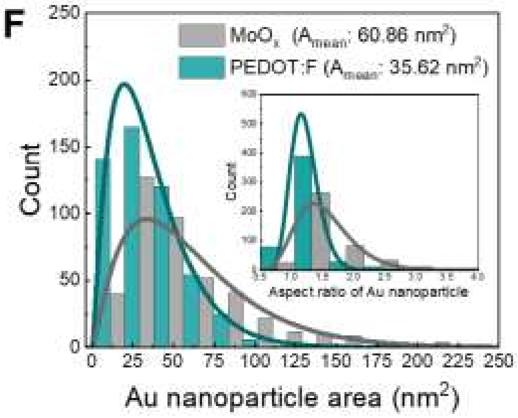


Microstructure: SEM

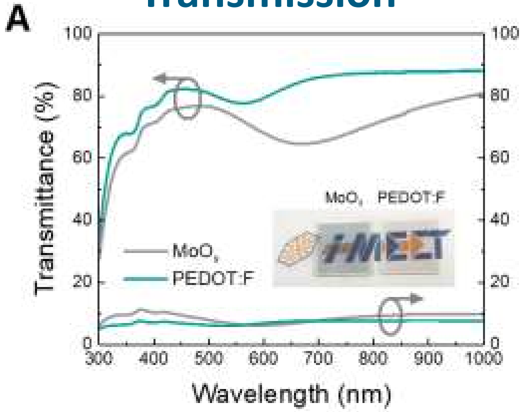


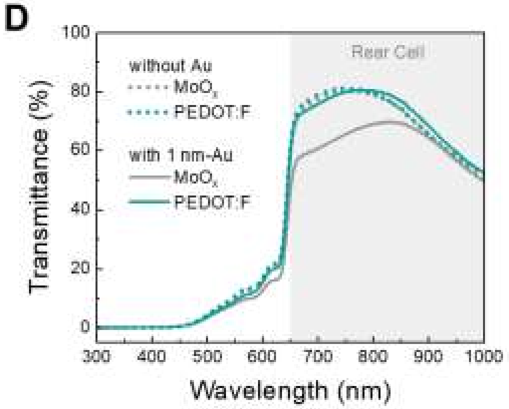


Featurization: Size distribution



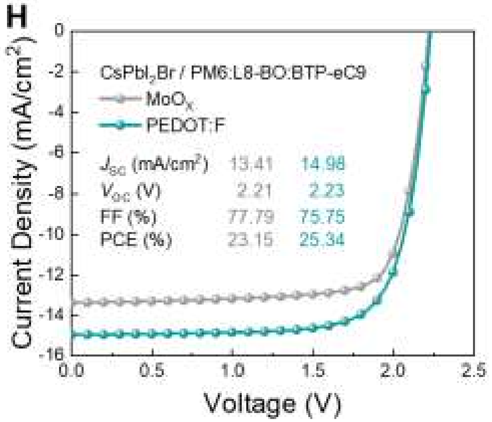
Transmission





Jingjing Tian
\+ Chao Liu

Electrical performance



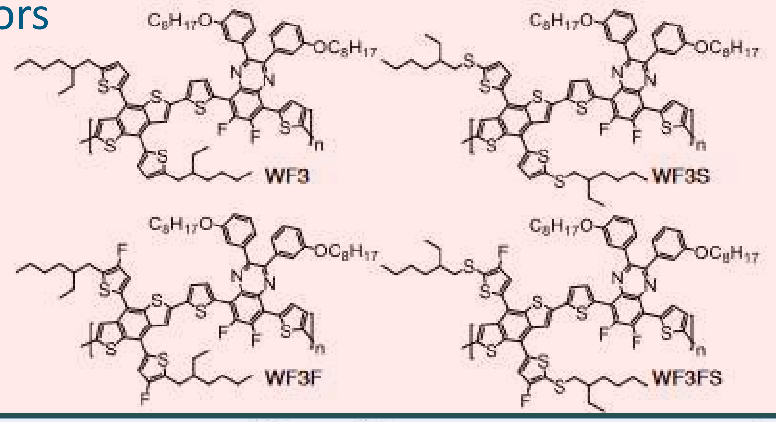
Causal structure of Material Science

High fidelity study along the causal chain: two samples are enough for clear cut result

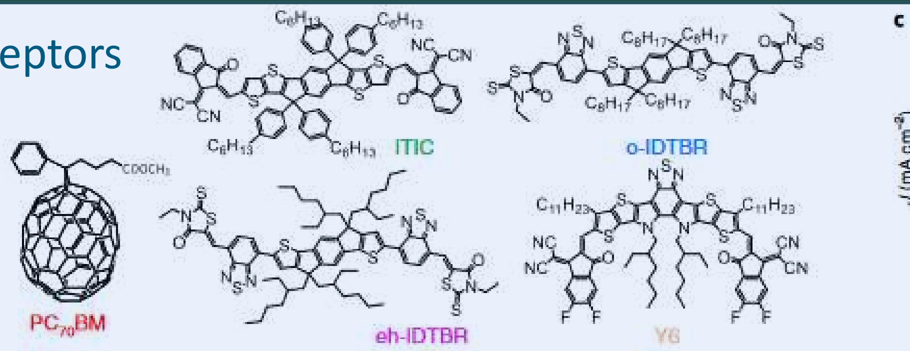
Jingjing Tian, Chao Liu, et al., in submission (2024)

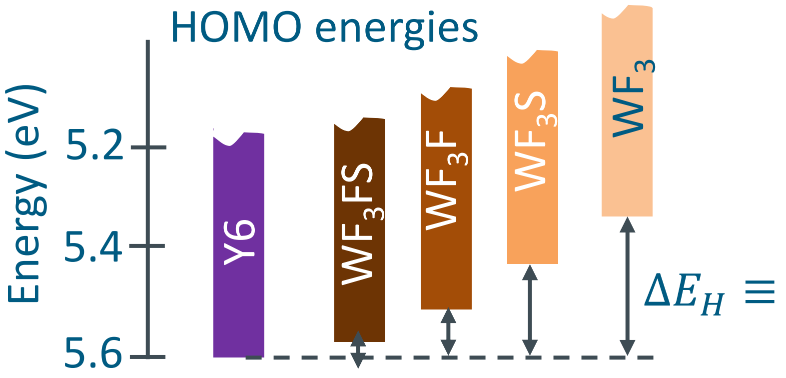
How to minimize voltage losses: driving forces and dual EL

Donors



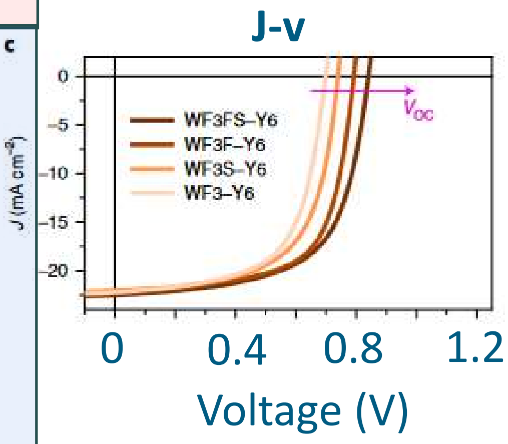
Acceptors

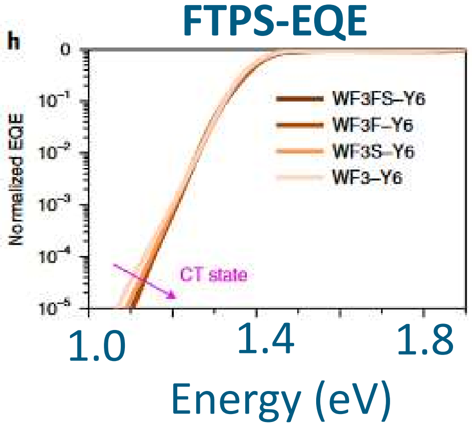


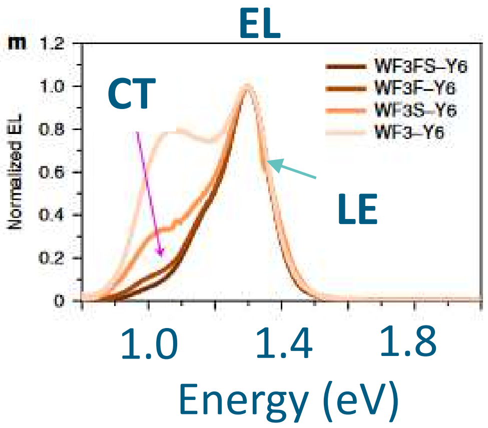


Driving force for dissociation of acceptor excitons into free carriers

Andrej Classen







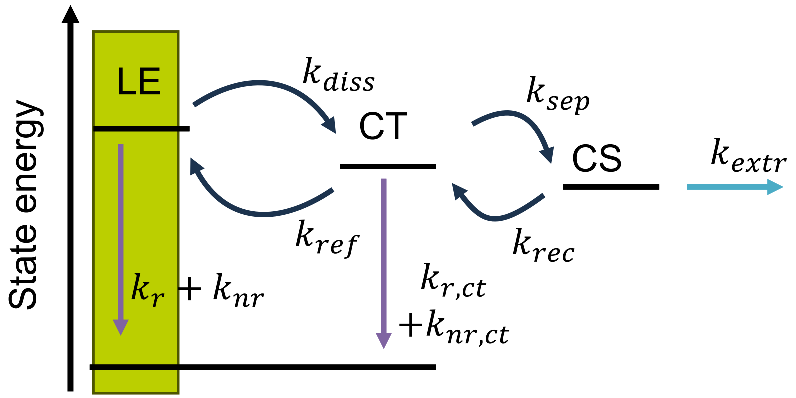
20 D:A pairs
Same process conditions

A Classen et al., Nat. En. 2020

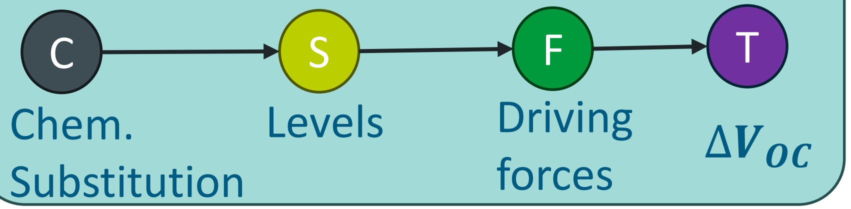
Ratio EL from CT and LE controlled by driving force

Voltage losses: driving forces and LE lifetime

Analytical treatment: Three-states model



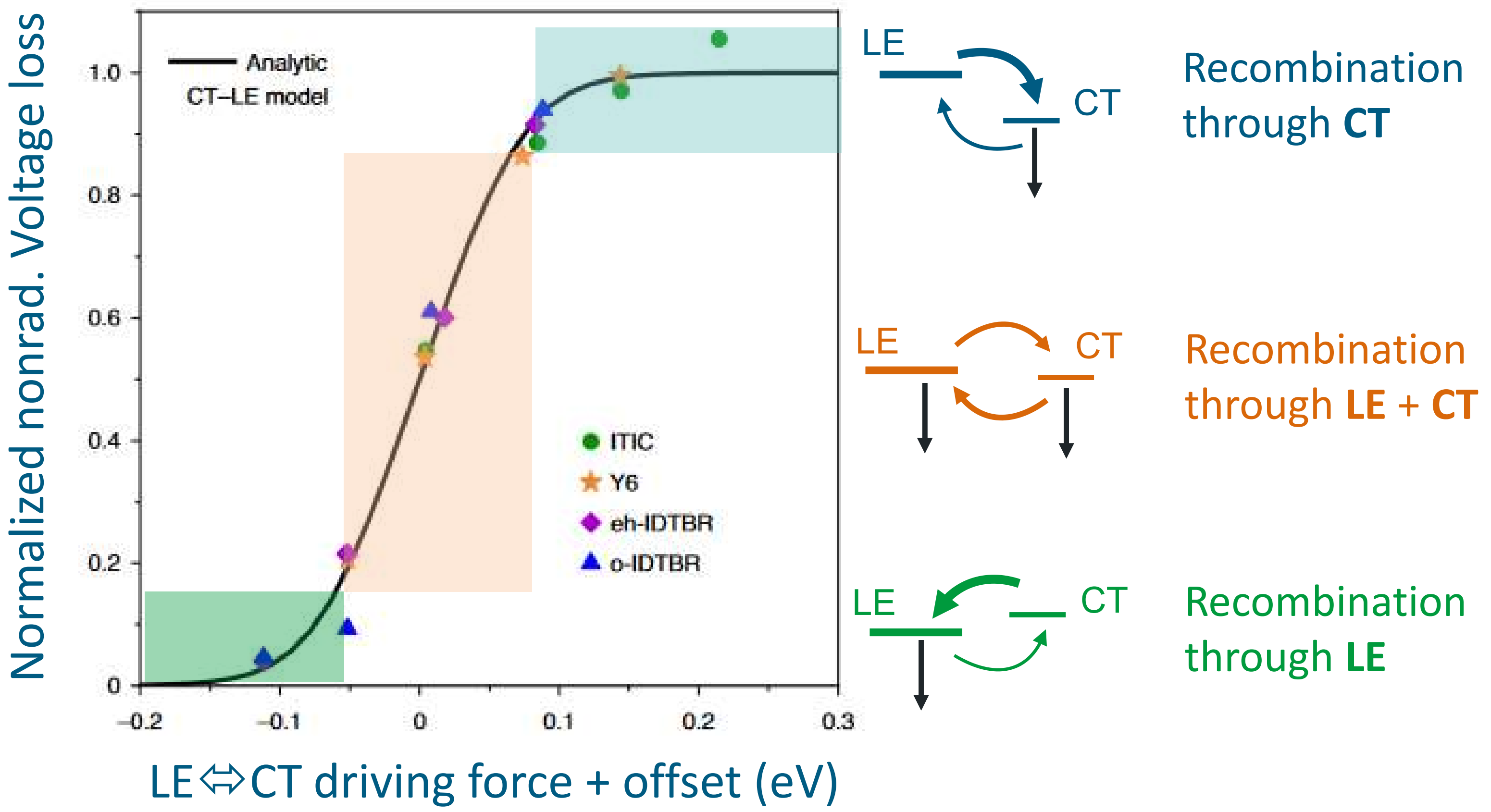
Causal structure



A. Classen et al., Nat. En. 2020

Voltage losses follow **universal trend**

Explained by LE ⇔ CT equilibrium



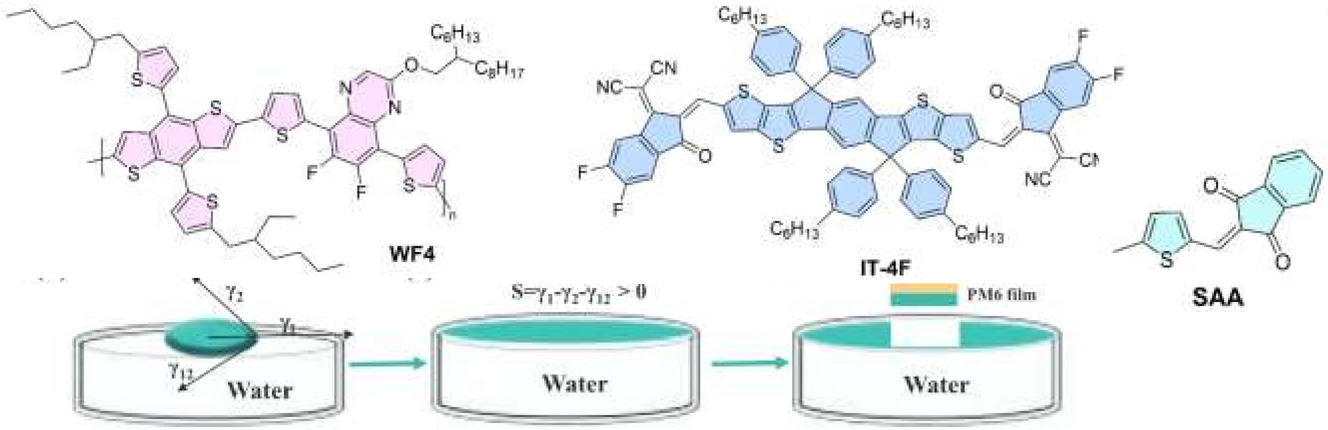
At low driving force, LE lifetime is crucial!

Voltage losses: the role of interfacial disorder

Only 1 D:A pair: WF4:IT-4F bi-layers
16 different process conditions

Method: transfer printing

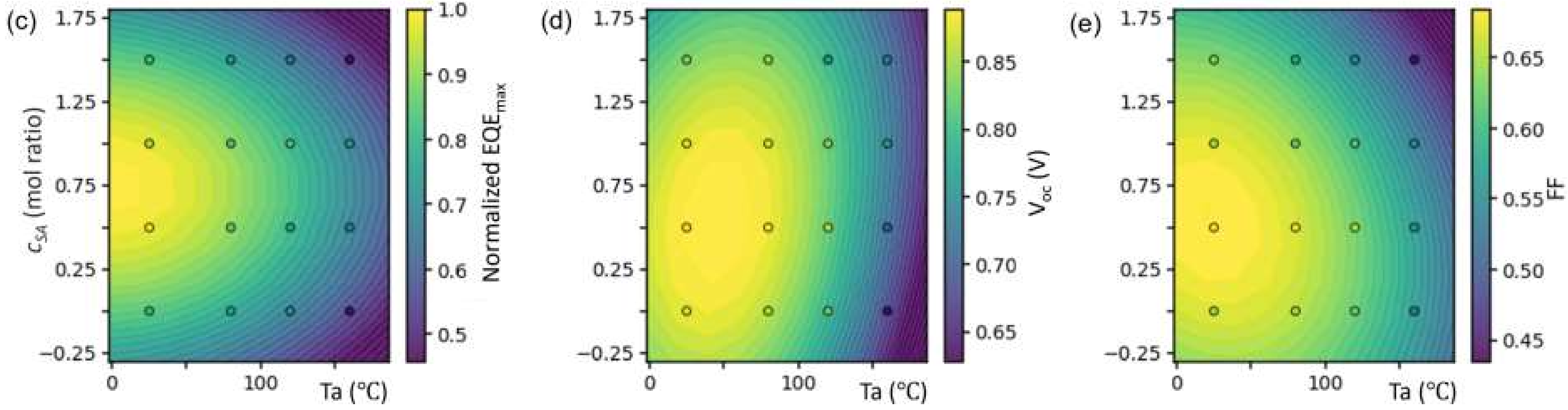
- abrupt interface
- Individual morphology control



Rong Wang

Charge… **…Generation: EQE** **…Recombination: V_{OC}** **…Extraction: FF**

SAA molar fraction



Excellent reproduction of exptl. results by GPR surrogate function

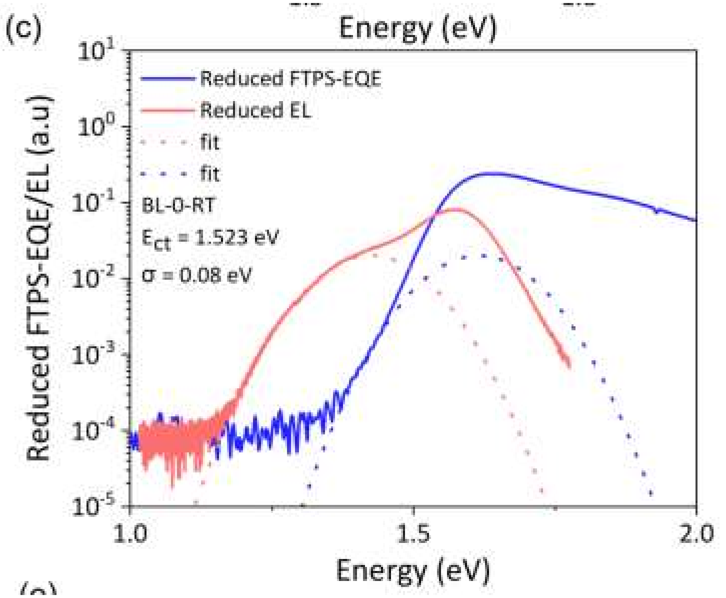
Annealing temperature

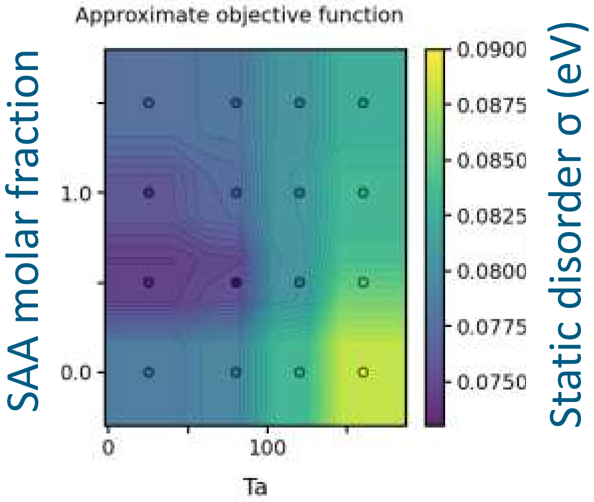
R. Wang et al., Adv. En. Mat 2024

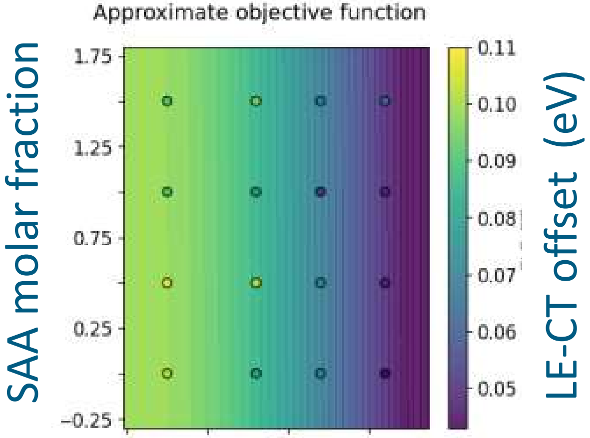
Different optima for charge generation and recombination

CT energies and static interfacial disorder

Marcus equation:
- Dynamic disorder – el-ph coupling
- Static disorder – interfacial DOS

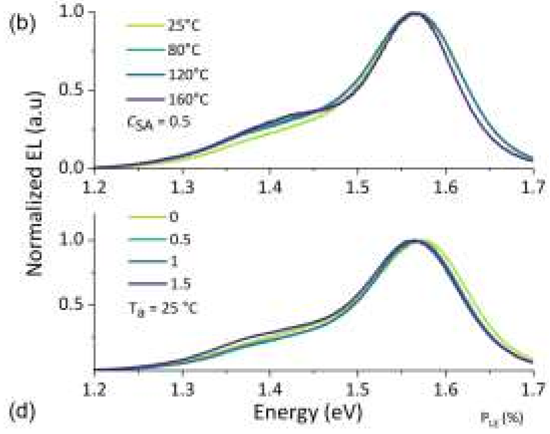


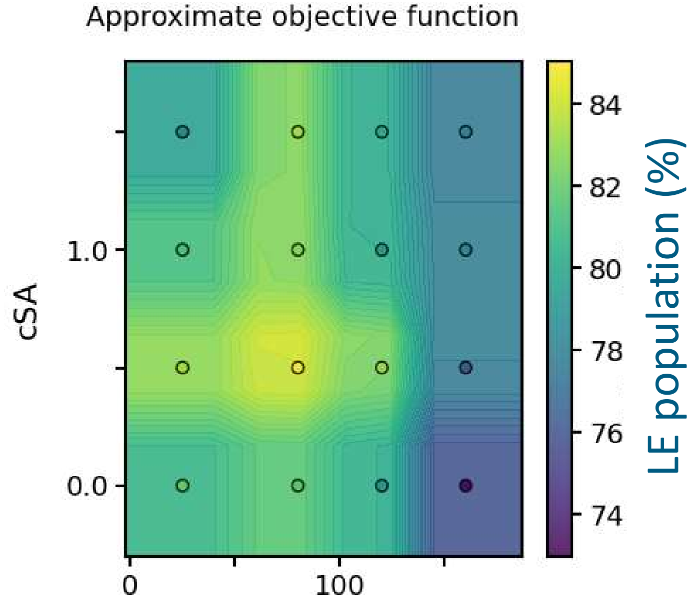




Annealing temperature (°C)

EL spectra





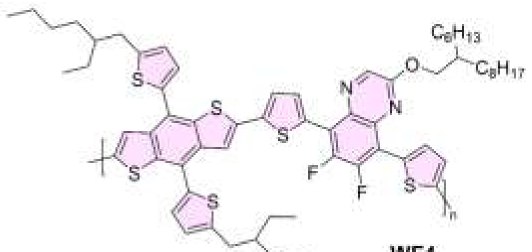
Annealing temperature (°C)

R. Wang et al., Adv. En. Mat 2024

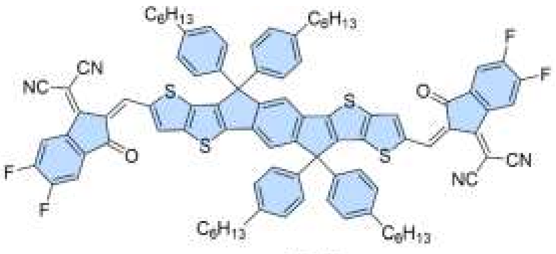
Surprisingly small variation of EL spectrum

Voltage losses: the role of interfacial disorder

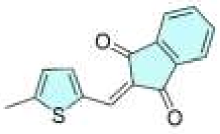
Only 1 D:A pair: WF4:IT-4F bi-layers
16 different process conditions



WF4

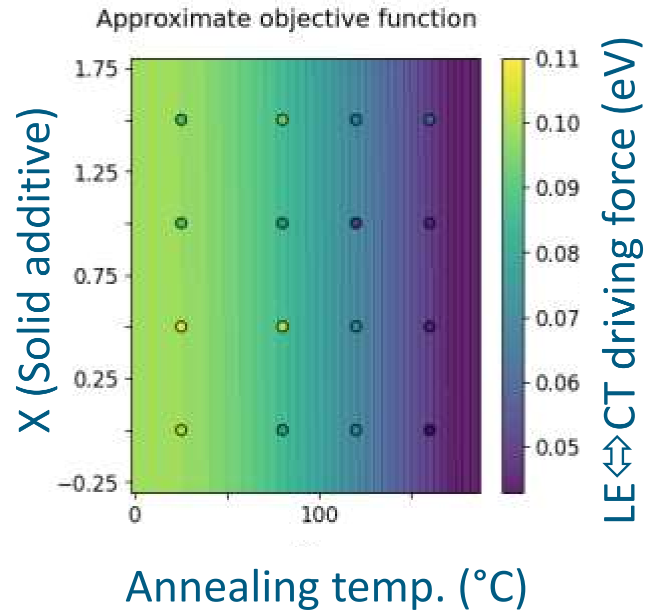


IT-4F

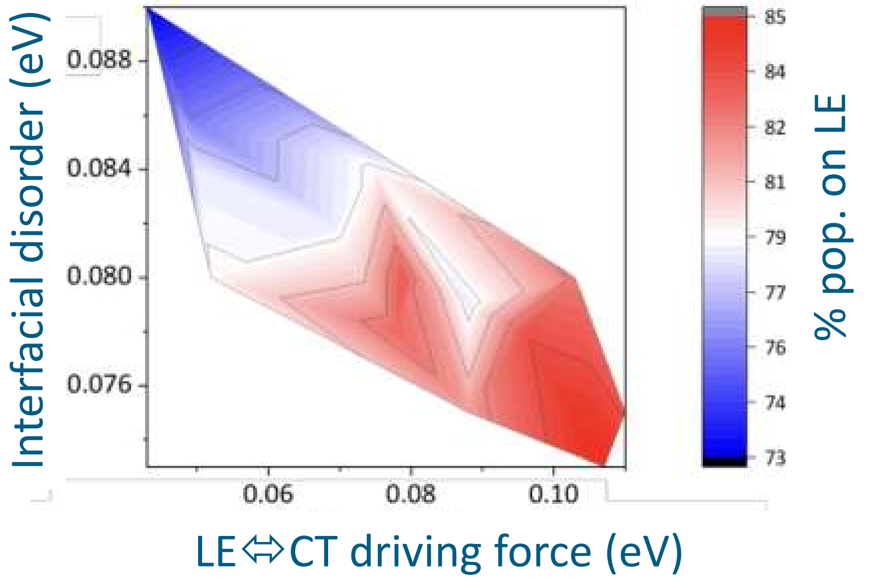


SAA

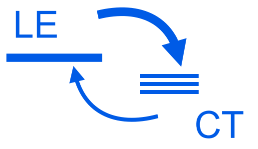
Strong effect of process conditions on driving force **within same D:A pair!**



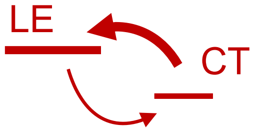
High driving force – high LE population!
“Anti-Boltzmann” behavior??



Caused by **interfacial disorder**



High disorder – high CT degeneracy

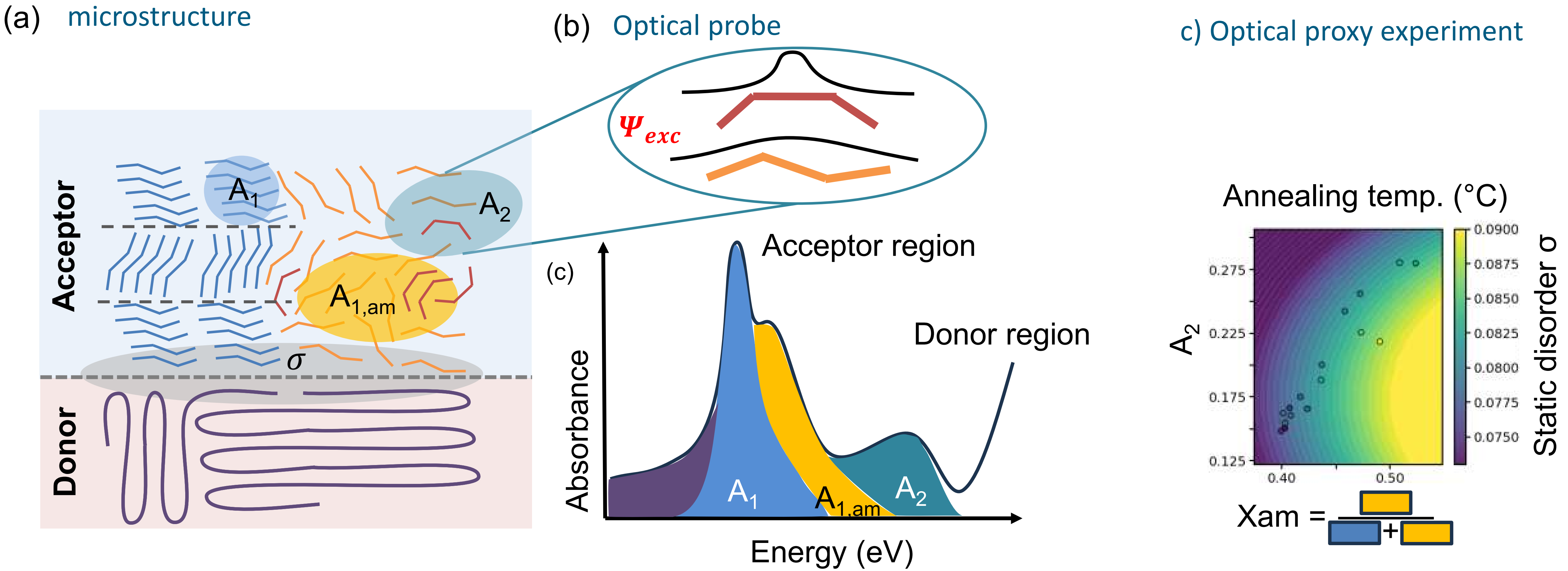


Low disorder – low CT degeneracy

R. Wang et al., Adv. En. Mat 2024

Interfacial disorder increases effective degeneracy of CT states

Training optical proxy experiments on the fly



R. Wang et al., Adv. En. Mat 2024

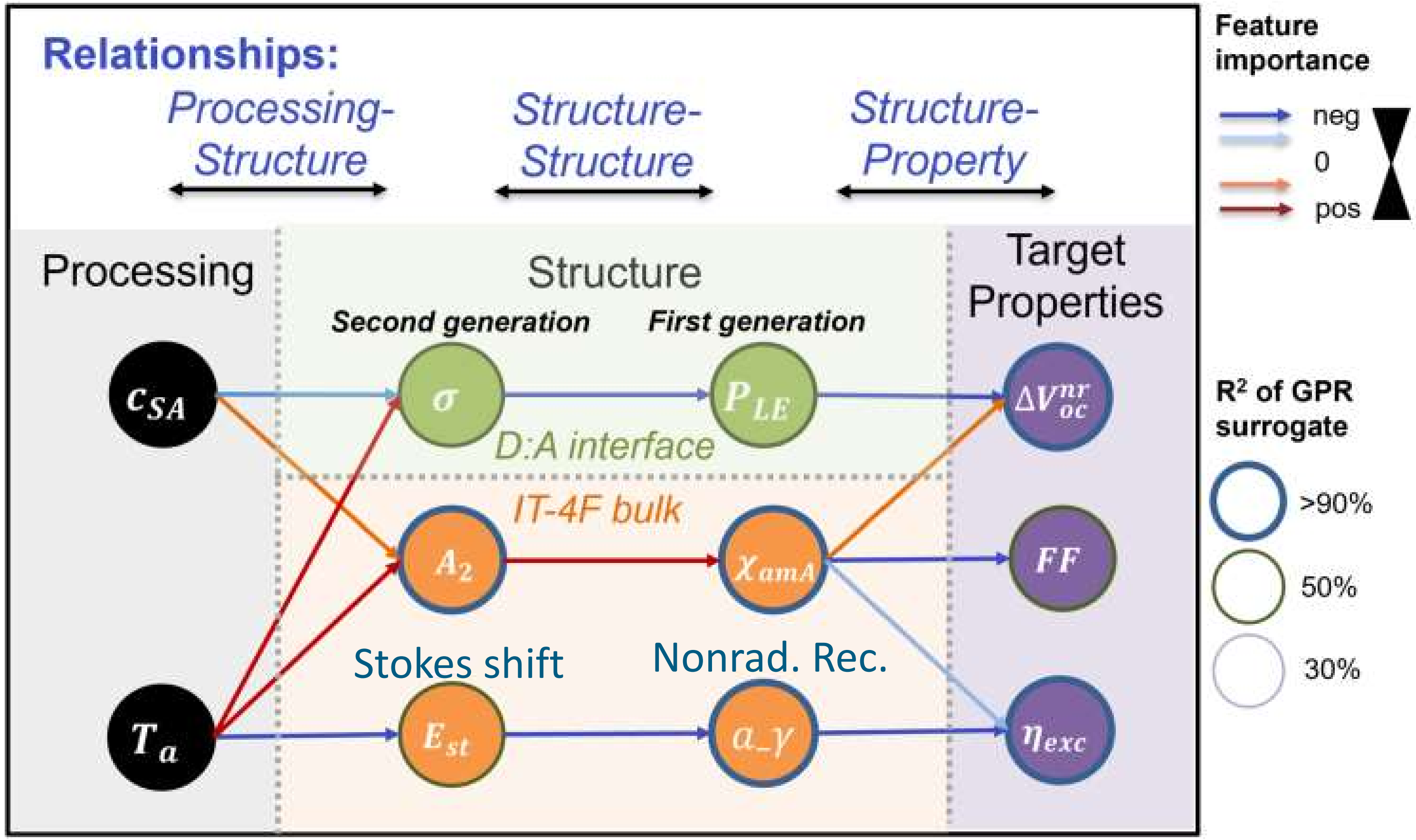
Bulk disorder controls interfacial disorder

UV-Vis is much easier than EL/FTPS/EQE! Proxy experiment!

Towards interfacial disorder management

Method: hierarchical mRMR embedded GPR (C. Liu et al., Adv. Mat 2023)

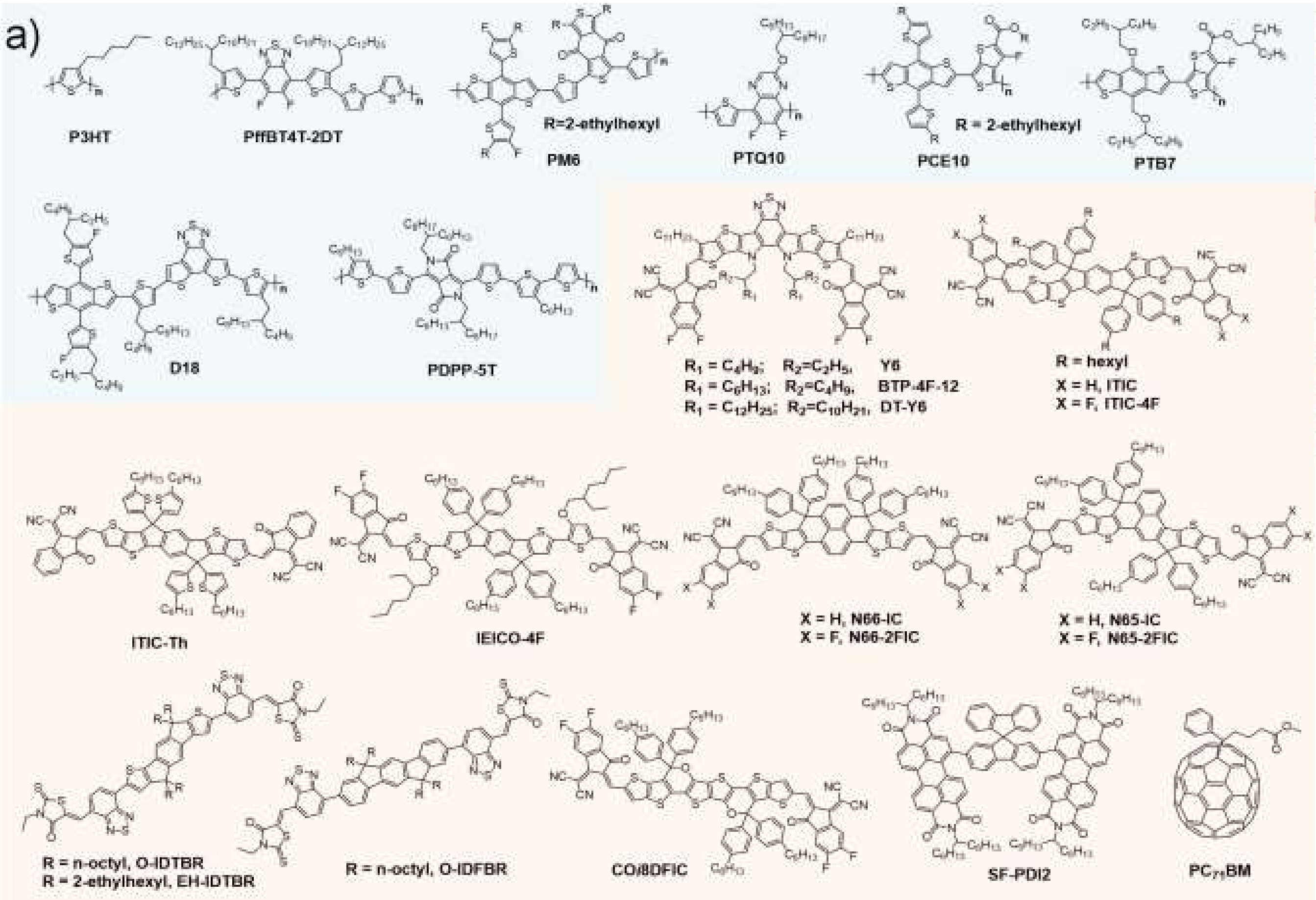
Result: Knowledge graph of essential predictors for target properties

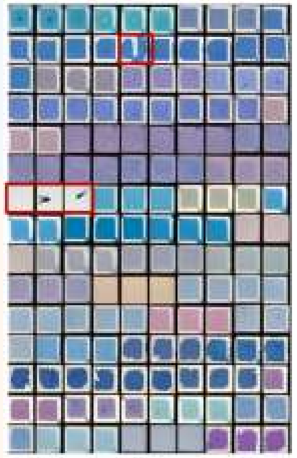


R. Wang et al., Adv. En. Mat 2024

Target properties controlled by different disorder motifs

Can OPV be processed in ambient conditions?



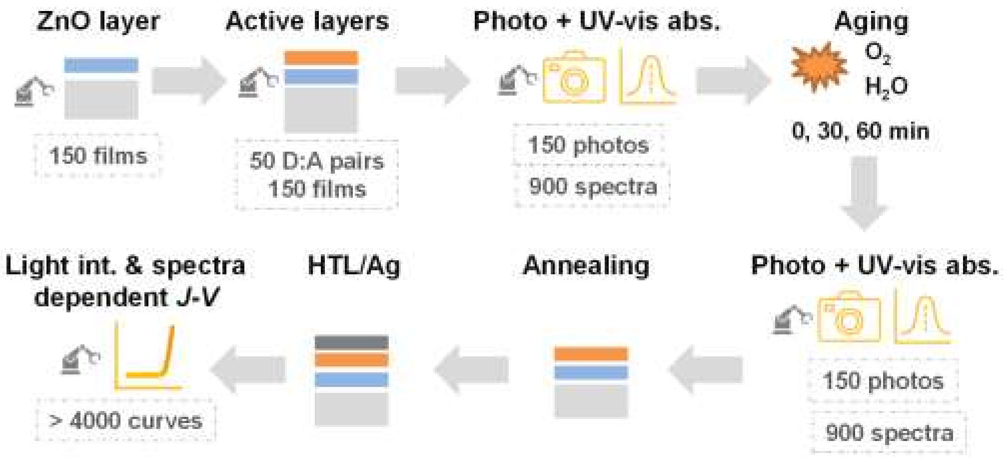


50 D:A pairs
150 devices

30 mins air/room light onto active layer before electrode deposition to simulate processing in ambient

Xiaoyan Du et al., INFOMAT (accepted) 2024

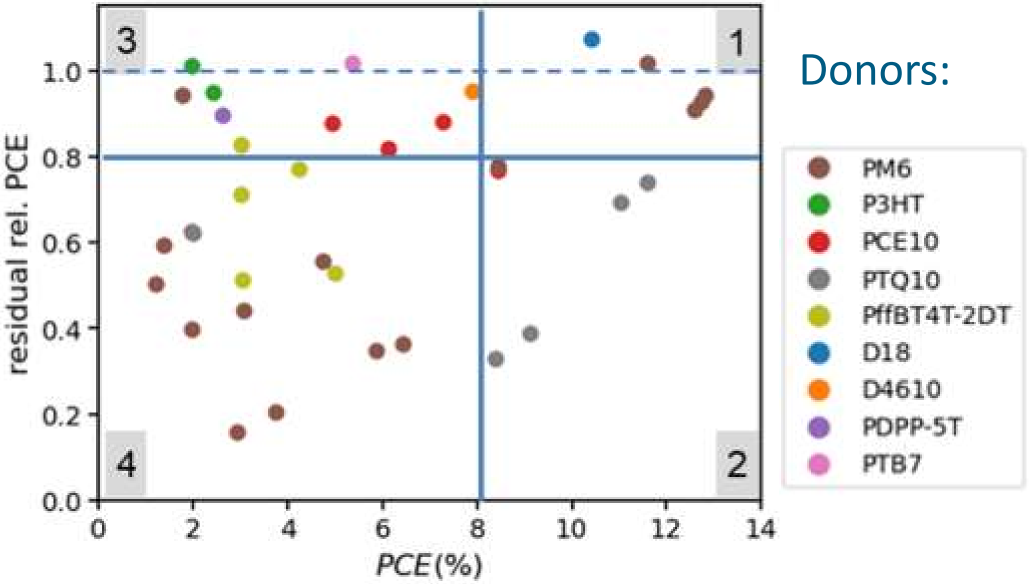
Xiaoyan Du



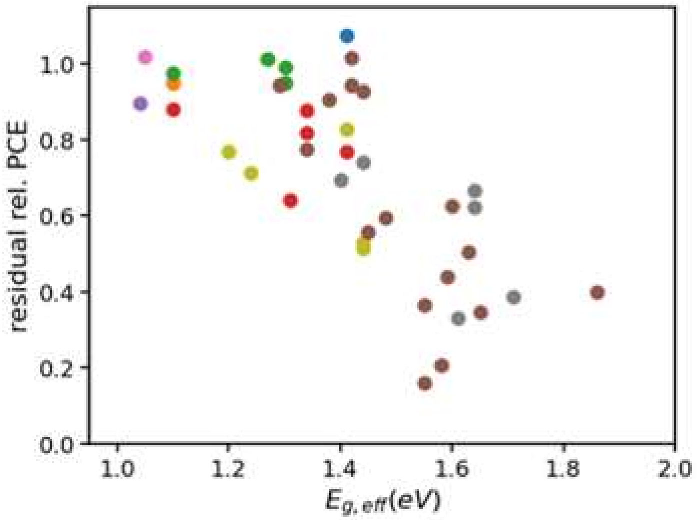
Air-light resilience during production allowing upscaling without need for vacuum/inert gas

Stability trends with frontier orbital levels

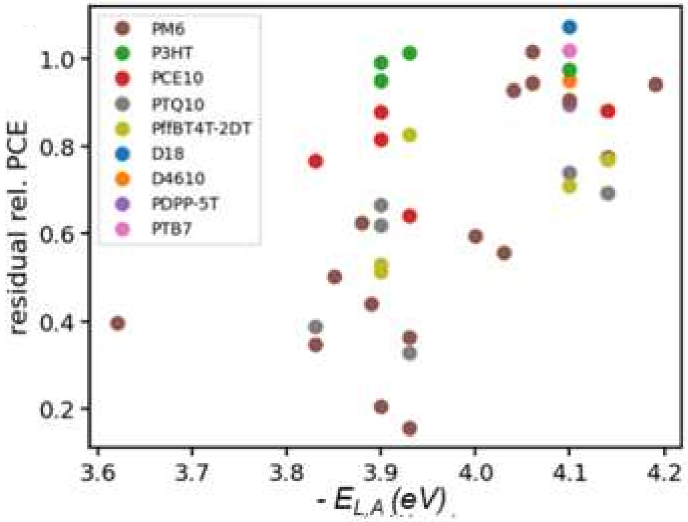
Air/light resilience after 30 mins



Effective gap



LUMO (acceptor)



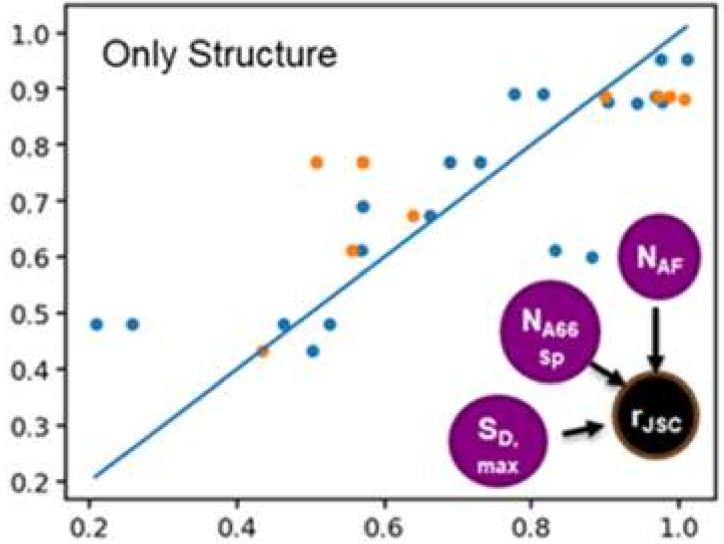
- **Air / light resilience scales with $E_{g,eff}$ rather than $E_{LUMO,A}$!**
- Points to energy transfer $$CT^* + {}^3O_2 \rightarrow {}^1O_2$$
- Rather than charge transfer $$A^* + {}^3O_2 \rightarrow A^+ + O_2^-$$

Xiaoyan Du et al., INFOMAT (accepted) 2024

Stability trends with molecular structure

Gaussian Process Regression (**GPR**) with embedded **mRMR** feature selection (C. Liu et al., Adv. Mat 2023)

Target: residual relative Jsc after 30 mins air/light



Predicted $\mathbf{r}_{JSC}$

Experimental $\mathbf{r}_{JSC}$

Relevant and non-redundant predictors:		**Known relation to physics**	**Tentative action on stability**
N_{AF}	# F on Acceptor	Frontier orbital levels	**Photochemistry**
$N_{A66,sp}$	# spiro bridges on 6-rings on Acceptor	Torsional mobility	**Diffusion**
$S_{D,max}$	Longest side chain on Donor	Hansen parameters	**Microstructure**

- **Machine-learned predictors** for air/light resilience agree with known physics
- For breakthrough innovation, we must **go beyond the known**.

Xiaoyan Du et al., INFOMAT (accepted) 2024

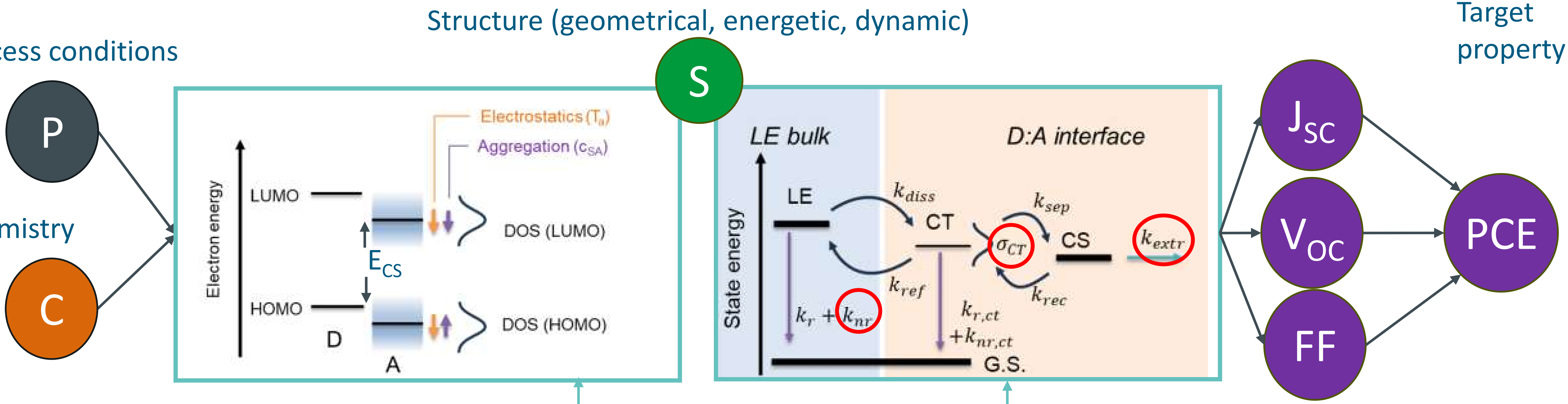
Fast parameter inference by multi-evidence fitting

Why not rely on blackbox optimization?

Quasi-infinite
dimensionality!

Fast parameter inference by multi-evidence fitting

Why not rely on blackbox optimization?



Effective gap

Stationary carrier density under 1 sun

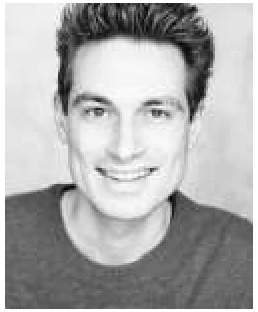
Quasi-infinite dimensionality!

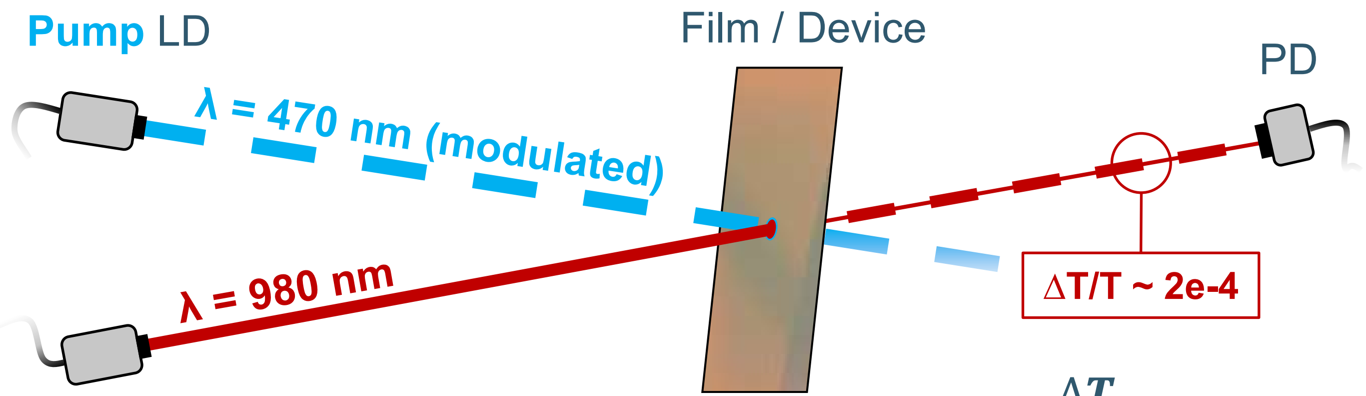
$$V_{OC} = E_{CS} + k_B \cdot T \cdot ln\left(\frac{c_{st}^2}{N_{eff}^2}\right)$$

Linking P and C directly to material parameters dramatically reduces search space

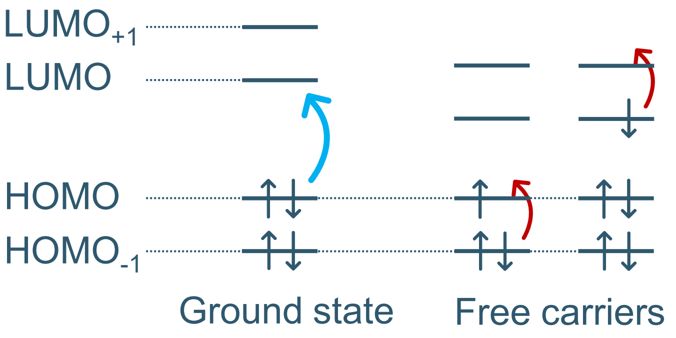
A novel implementation of transient pump-probe kinetics

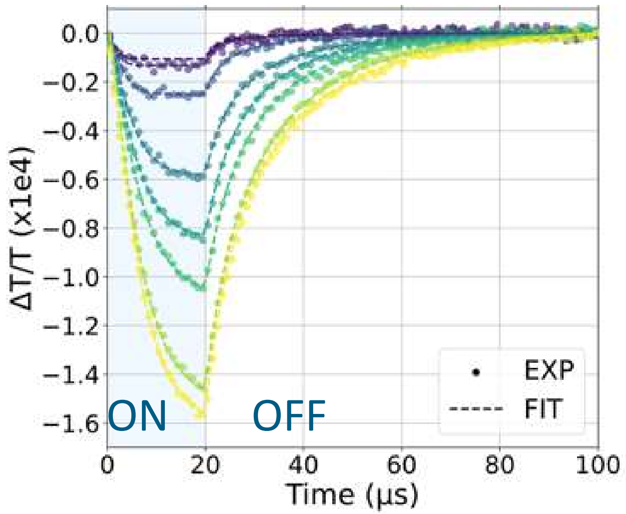
Julian Haffner-Schirmer
+ Vincent Le Corre





$$\frac{\Delta T}{T} \approx \sigma c d$$





$$\frac{dc}{dt} = G_{CS} - k_1 \cdot c - k_{r,eff} \cdot c^2$$

Intrinsic determination of σ due to square pulses:

- From initial slope $\frac{dc}{dt} = G_{CS}$ = photon flux X EQE!
- From stationary value $c_{st} = G_{CS}/k_1$

Global fitting yields k1, $k_{r,eff}$,σ with very little cross-talk

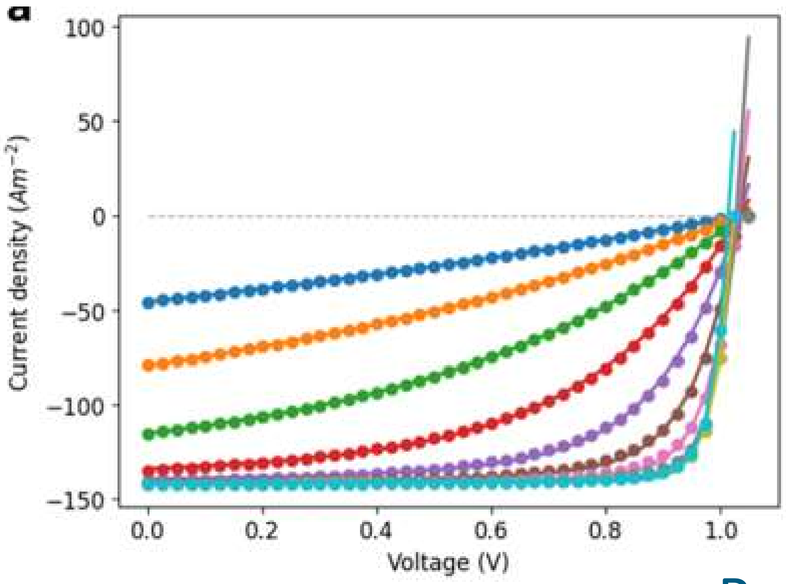
Fitting J-V traces with the same rate equation

Benchmarking against simulated data

$$\frac{dc}{dt} = G_{CS} - (k_1 + k_{extr}) \cdot c - k_{r,eff} \cdot c^2$$

$$k_{extr} = \frac{1}{\tau_{tr}} = \frac{2 \cdot \mu_{extr} \cdot (V_{OC} - V_{ext})}{L^2}$$

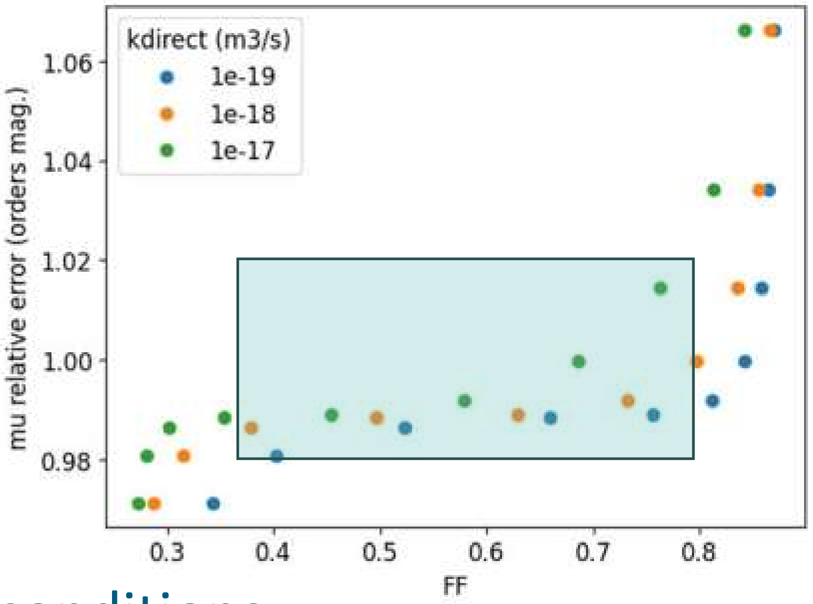
Drift-diffusion simulations



simsalabim

Rate equations

Mobility: fitting error

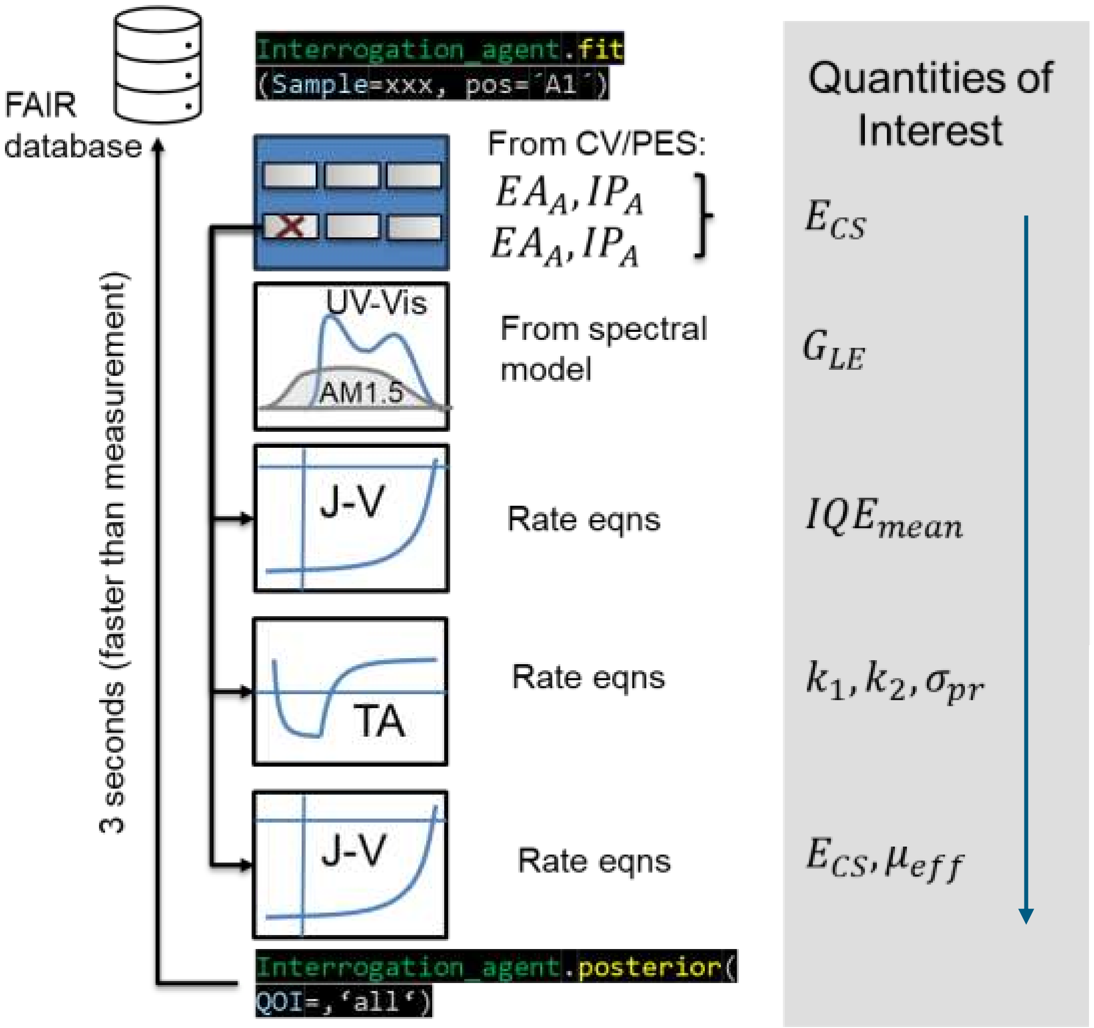


Rate equations OK under 2 conditions:

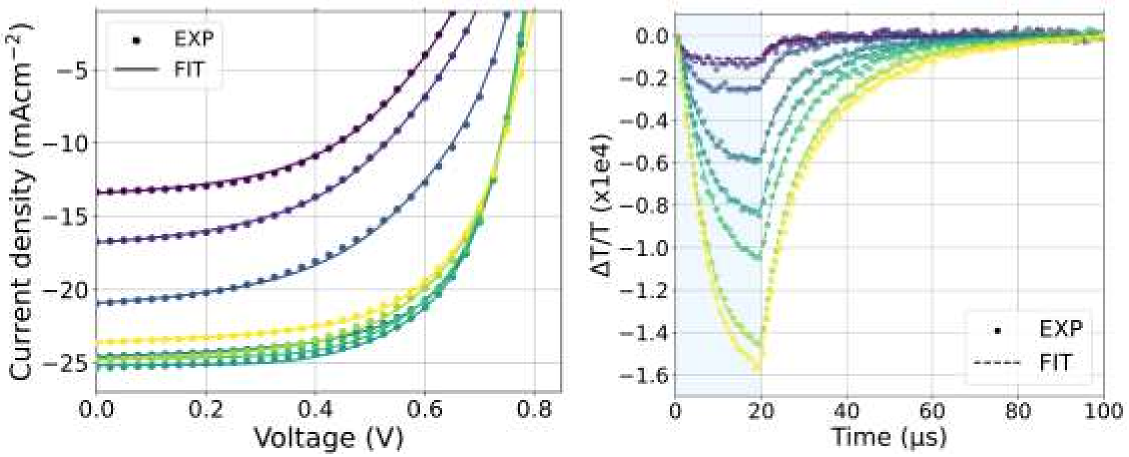
- FF>80%
- No extraction barriers

Fast parameter extraction by self-learning agents

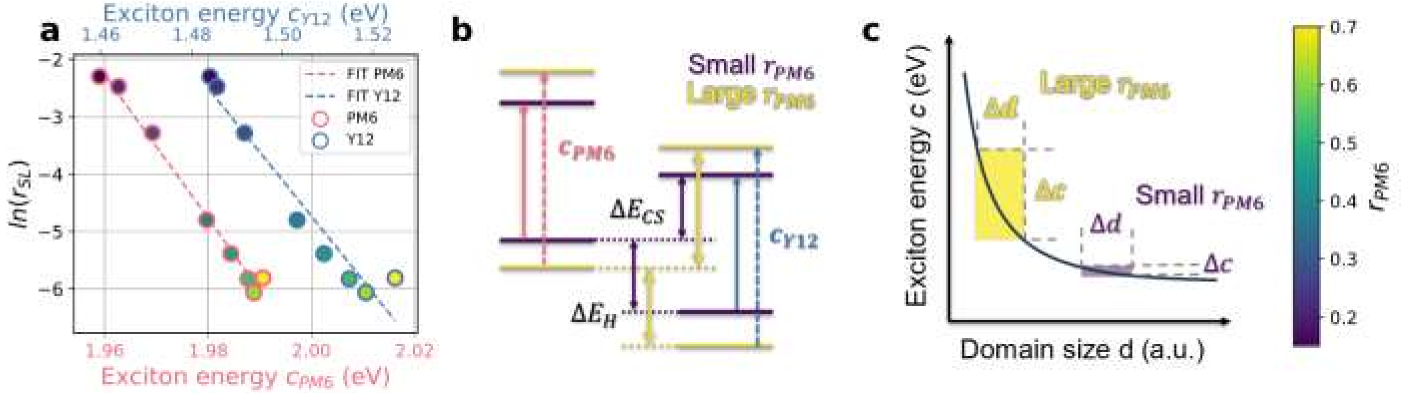
Understanding FF losses in acceptor-rich blends of PM6:BTP-4F-12



Excellent multi-evidence fits



Langevin reduction factor r_{SL}: scaling with energy disorder



J. Haffner-Schirmer et al., in submission (2024)

OPV beyond 20% PCE: accelerating the next breakthrough

Problem statement:

- Discovery of unseen trends requires big data: **volume, variety, veracity:**
  - **1000s of D:A pairs**
  - **Millions of microstructure variations**

Impossible for a single lab
$\Rightarrow$ **Veracity** issues

Can't provide 10^6 cross-section TEMs
$\Rightarrow$ **Black box** optimization without using human capacity for abstract thinking

Needed:

Probabilistic method to **learn** (attain predictive power) from

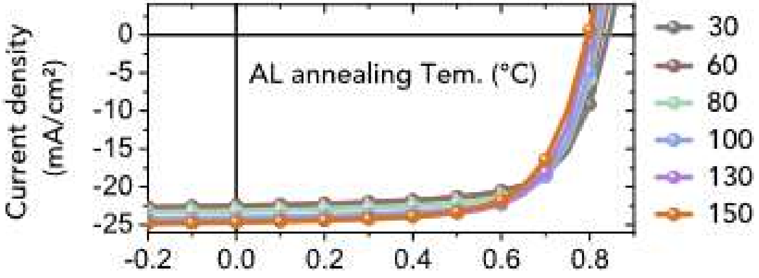
- **incomplete,**
- **indirect,**
- **uncertain evidence**

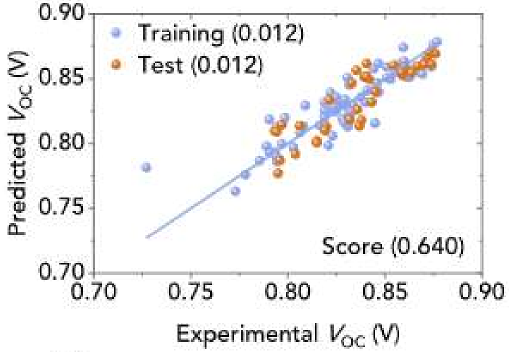
This is exactly the realm of a digital twin

How does a digital twin achieve acceleration?

Featurization allows redundancy rejection

J-v curves





Process conditions

Microstructure

Observables

cause

S

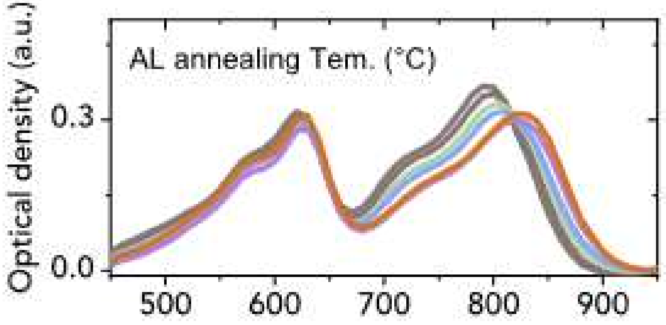
cause

O

GPR

High fidelity prediction of V_{OC}

UV-Vis spectra

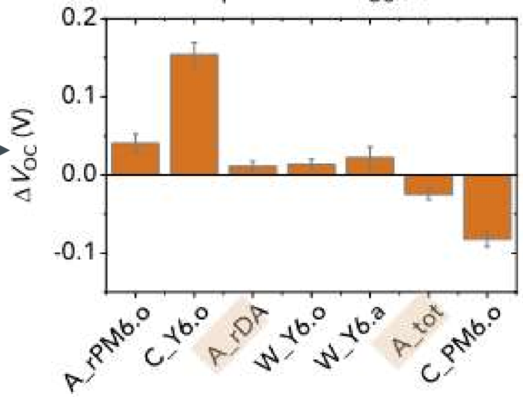


Morphological Features:

Domain size

Order

Film thickness



X. Du et al., Joule 2020

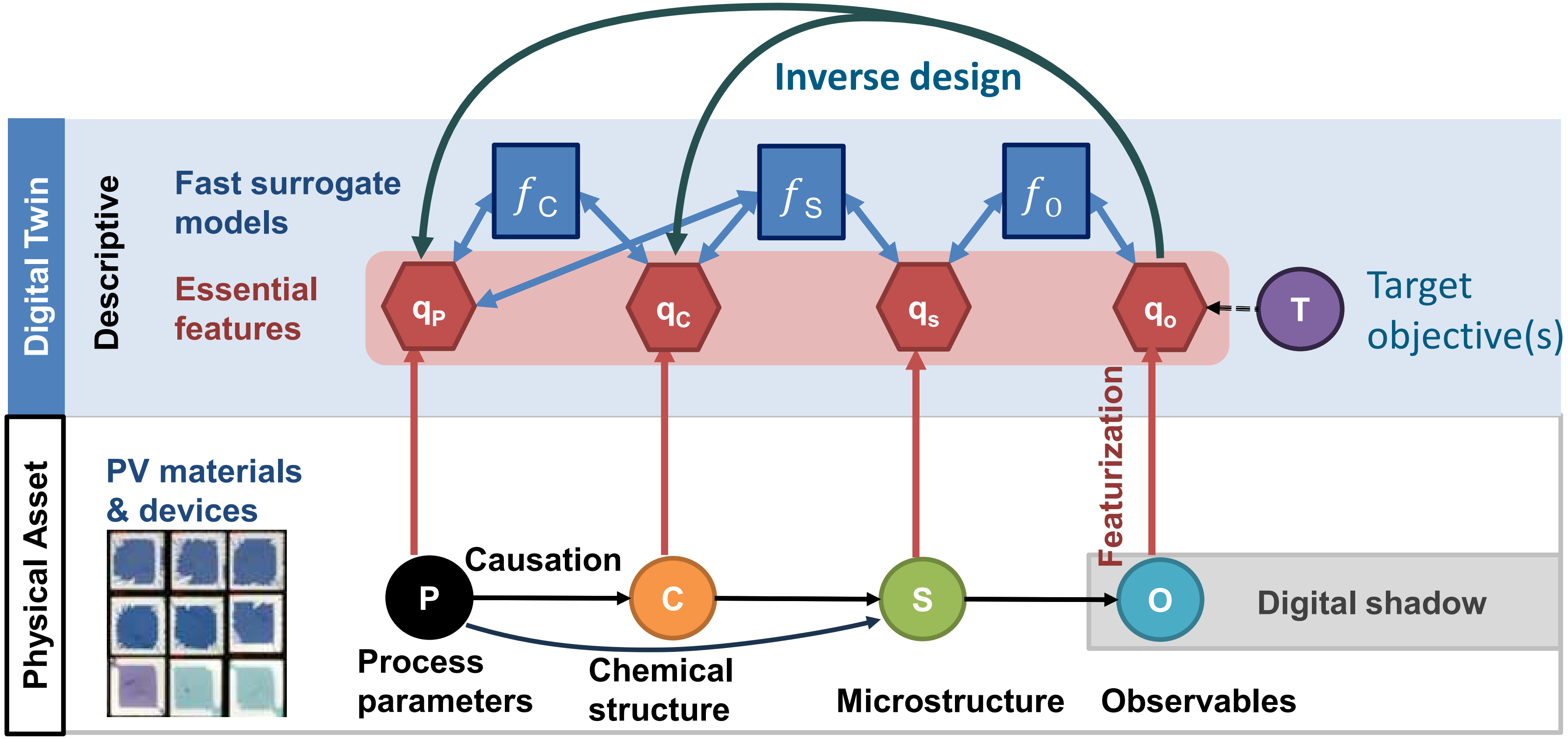
UV-Vis spectra: **essential microstructure features** for correct V_{OC} prediction

Redundancy rejection is not approximation!

Digital twin: redundancy rejection enables inverse design

$$q_C = f_S^{-1}(q_S) = f_S^{-1}\left(f_o^{-1}(q_o)\right)$$

Find optimal chemical structure given a set of target objectives



Essential optical features to predict target

Essential structural features to predict optical features

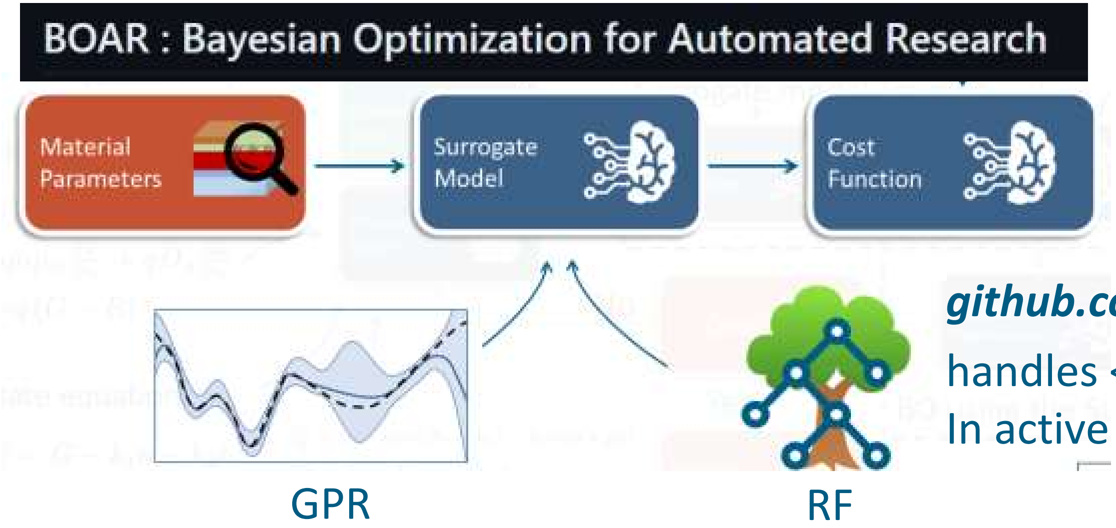
Essential chemical features to predict structural features

L. Lüer et al., Joule 8, 1–17, 2024

Challenges

Current hot topics in data science:

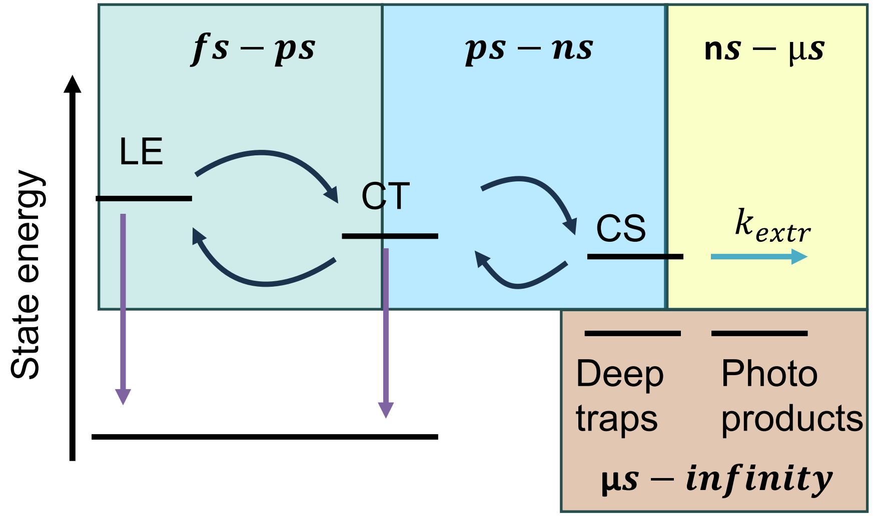
- Uncertainty quantification
  - Requires posterior calculation > 15 parameters
- Cascading surrogates
  - From femtoseconds to hours
  - From Angstroms to meters
- Mixed integer optimization



github.com/i-MEET/boar

handles <=10 dimensions

In active development



Conclusions

- Big data approach required to further reduce voltage losses and increase operational stability of OPV technology
- A digital twin will allow acceleration of knowledge generation by redundancy rejection and fast surrogates allowing inverse design
- A strategy for attaining inverse design capacity is proposed

Goal: from a set of target objectives, identify the optimal molecular structure and corresponding process conditions

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