Machine Learning For Materials And Device Simulations

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Topics

- Multiscale Models
- Hot carriers and polarons
- Machine Learning insights into Monte Carlo simulations

Practical modelling: students investigating installation of solar on Bath Abbey roof
https://doi.org/10.1002/ese3.1069

Photovoltaics theory and modelling advances

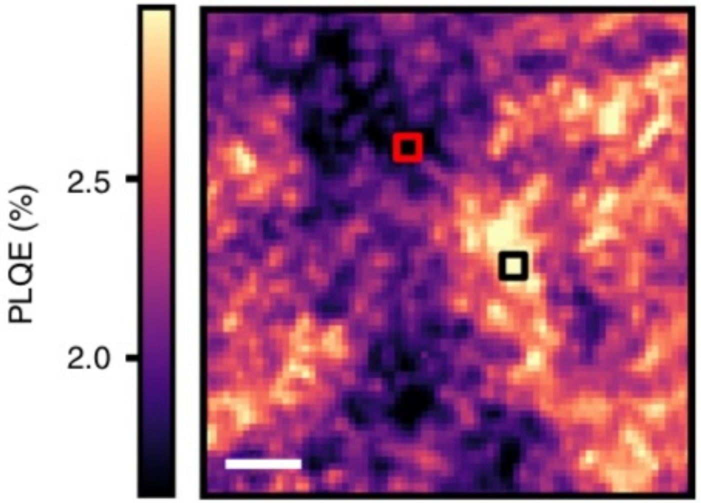
Was descriptive, now predictive
Can model complex and heterogeneous materials

Electron backscattering diffraction plot for halide perovskite
E Tennyson et al *Nature Rev Mater* (2019)

Highlights

- Machine Learning to accelerate simulations
- Publicly available databases: Materials Project
- Electronic structure-based *ab-initio* molecular dynamics beyond 100 million atoms
- Model temperatures > 0K through free energies
- DFT inclusion of many body effects - band aligning
- Device Models that include mobile ions
- Mesoscopic models of charge transport

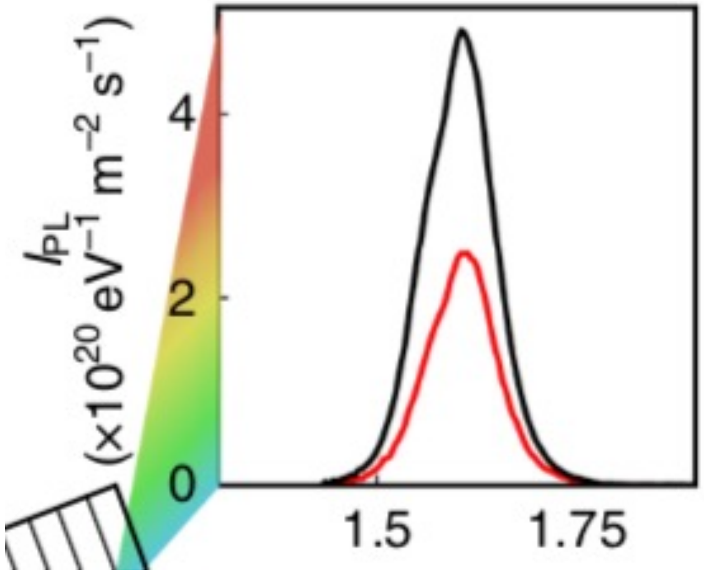
Multiscale Models: Why?



Photoluminescence Quantum Efficiency PLQE

$Fa_{0.79}Ma_{0.16}Cs_{0.05}Pb(I_{0.83}Br_{0.17})_3$

Scale bar: 2 μm



PL spectra from black and red highlighted regions

K Frohna et al *Nature Nano Tech* (2022)

Heterogeneity on submicron length scales dominates optoelectronic response for alloyed perovskites. Implications for hot carrier cooling.

Models at varying length and timescale

time s

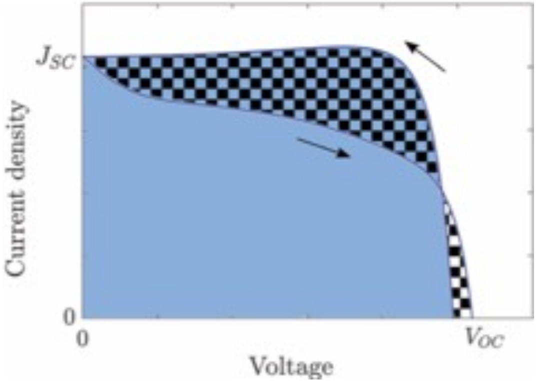
10^0 to 10^2

10^{-6}

10^{-8}

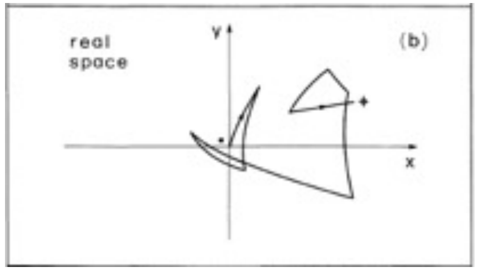
10^{-10}

Macroscopic



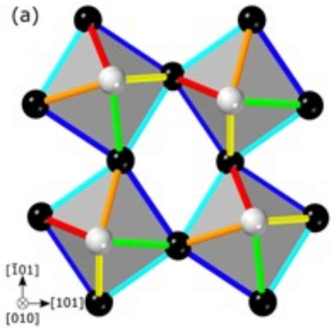
J M Cave et al, *J Appl Phys* **128**, 184501 (2020)

Mesoscopic



L A D Irvine et al, *Phys Rev B* **103**, L220305 (2021)
McCallum et al *APL Machine Learning* accepted (2023)

Electronic/ atomistic



T J A M Smolders et al, J Phys Chem Lett, **12**, 5169 (2021)

length m

10^{-10} to 10^{-9}

10^{-9} to 10^{-6}

10^{-6} -10^0

Hot Carriers and Polarons

Boltzmann Transport Equation, BTE

Statistical behaviour of a thermodynamic system away from equilibrium

Jamie Lerpiniere

Lewis Irvine

Matthew Wolf

William Saunders

$$\frac{\partial f}{\partial t} + \mathbf{F} \cdot \frac{\partial f}{\partial \mathbf{p}} = \left(\frac{\partial f}{\partial t} \right)_{\mathrm{scatt.}}$$

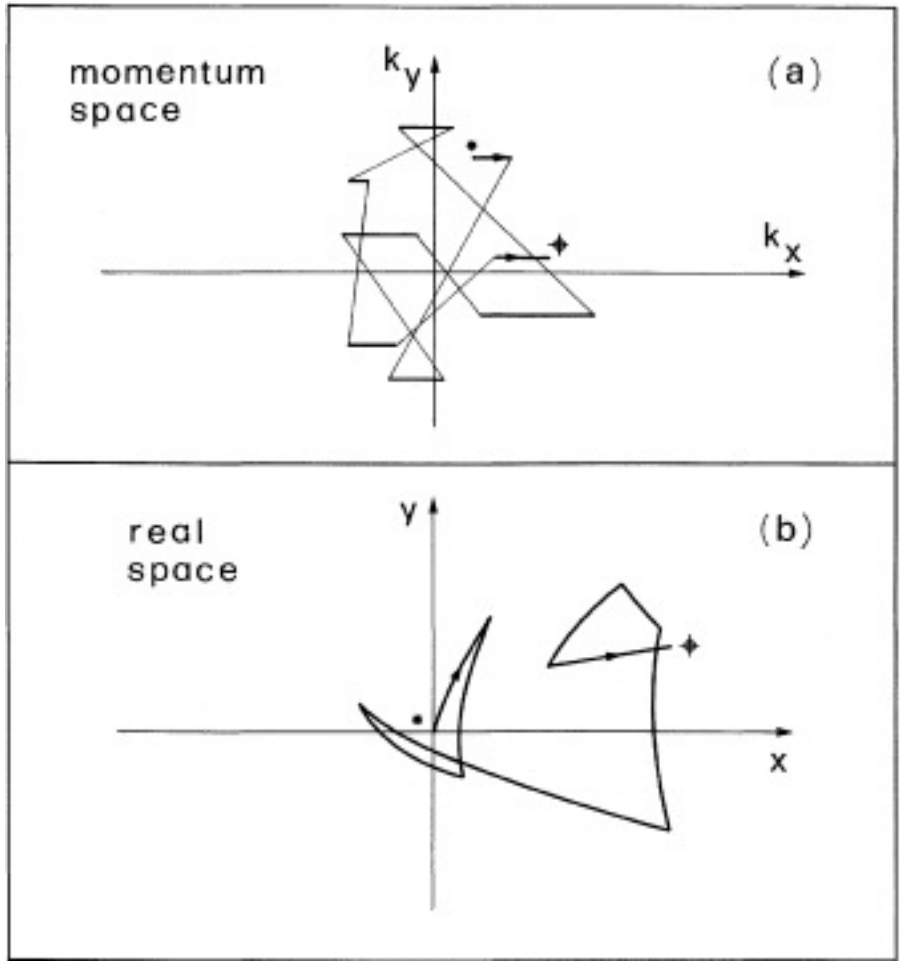
$f(\boldsymbol{p}, t)$ is one-particle probability distribution function

$\boldsymbol{p}$ is momentum, t is time, $\boldsymbol{F}$ is field

All parameters can be calculated from first principles

Can solve BTE using Monte Carlo simulations

Monte Carlo device simulation for delocalized charges



Select free flight time δ*t* in electrostatic field from uniformly distributed random number *r* and total scattering rate Γ

$\delta t = \Gamma^{-1} \log(r)$

Charge carriers are scattered in polar semiconductors by:

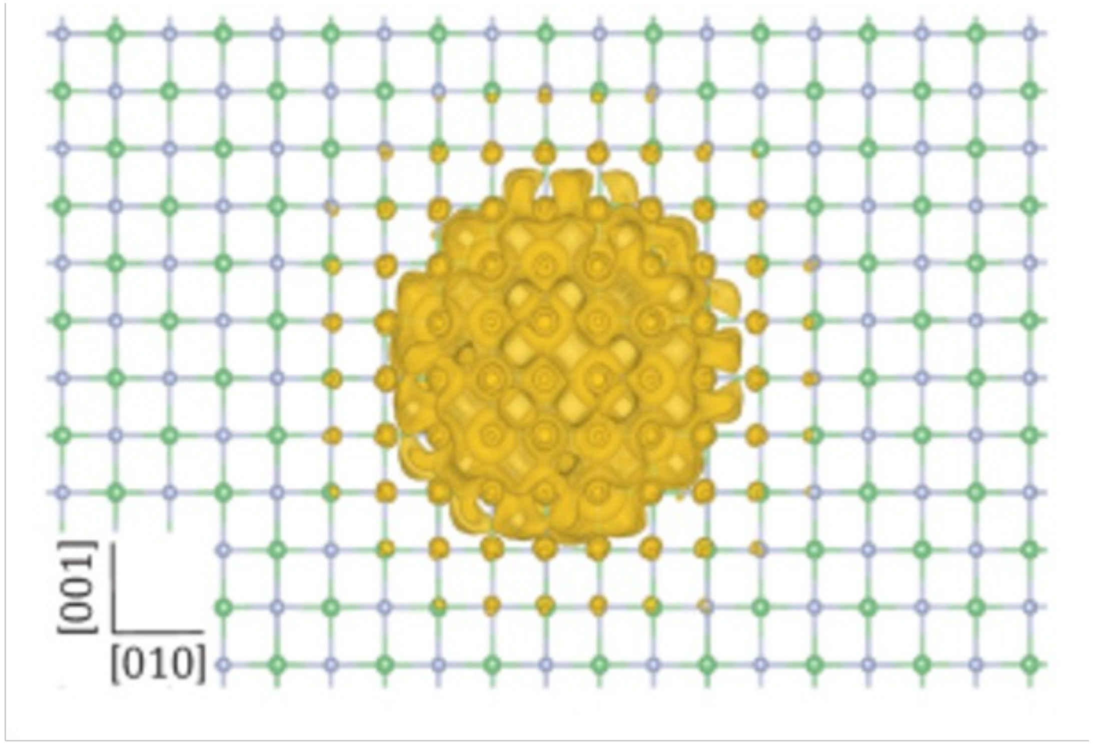
- polar optical phonons
- acoustic phonons
- ionised impurities
- other carriers
- grain boundaries

C Jacoboni et al *Solid State Electronics* **20** 77 (1977), *Rev Modern Phys* **55** 646 (1983)

Hot Carriers

- 2D electronic spectroscopy measures evolution of hot carrier distributions from ~10 fs
- 10-100 fs cooling dominated by carrier-carrier scattering
- As the distribution spreads out and cools, carrier–carrier scattering rate decreases and phonon scattering more important
- Fitting an effective temperature to a distribution can be misleading

Polarons

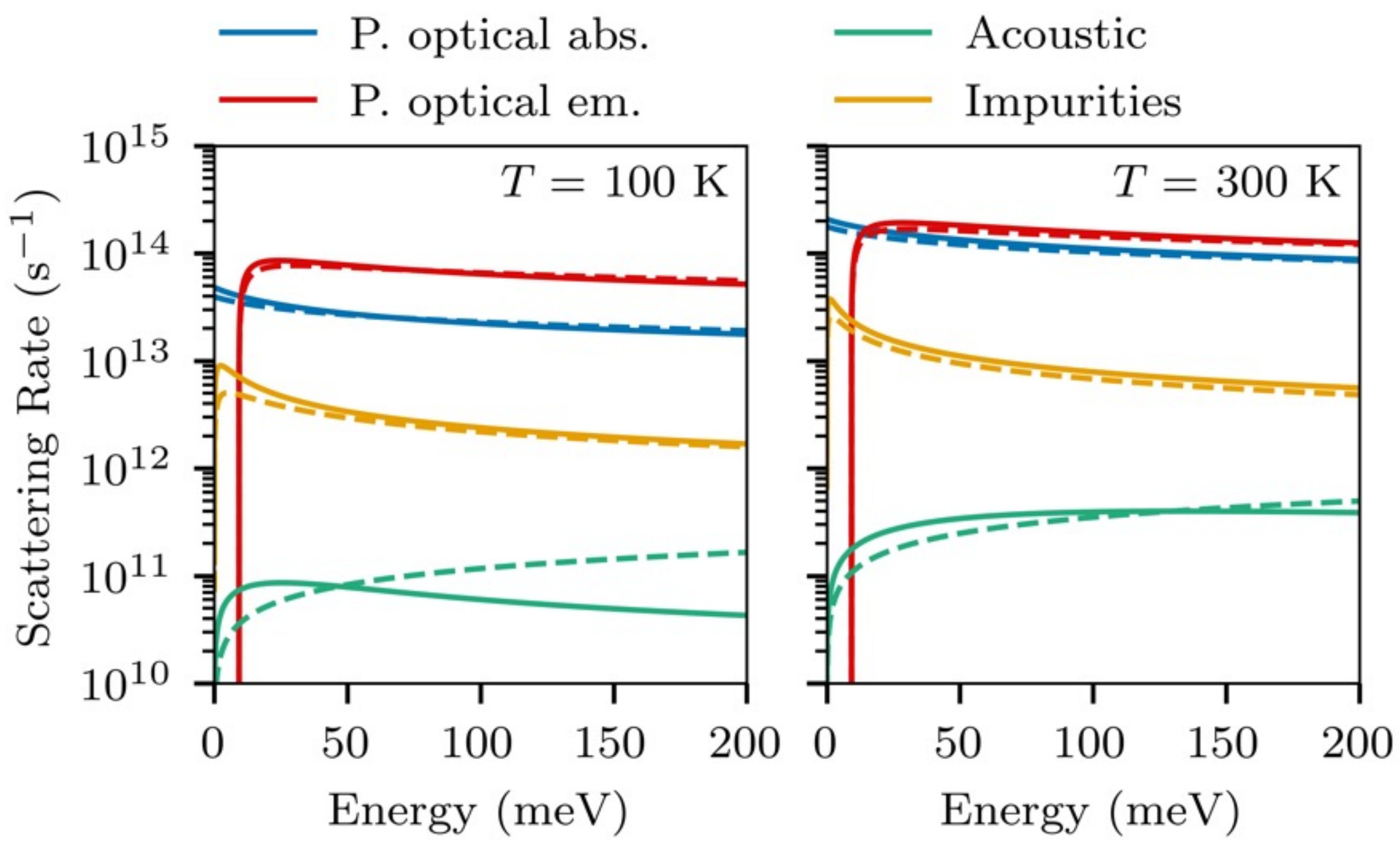


- Polaron radius >> lattice constant
- Itinerant state
- Shallow electronic state
- Observed that carrier mobility decreases with temperature

To what extent are charge carriers protected from interactions with phonons, defects and other charge carriers by their incorporation into polarons?

C Francini et al *Nature Rev Mat* (2021)

Scattering rates for polarons and band electrons in $MAPbI_3$

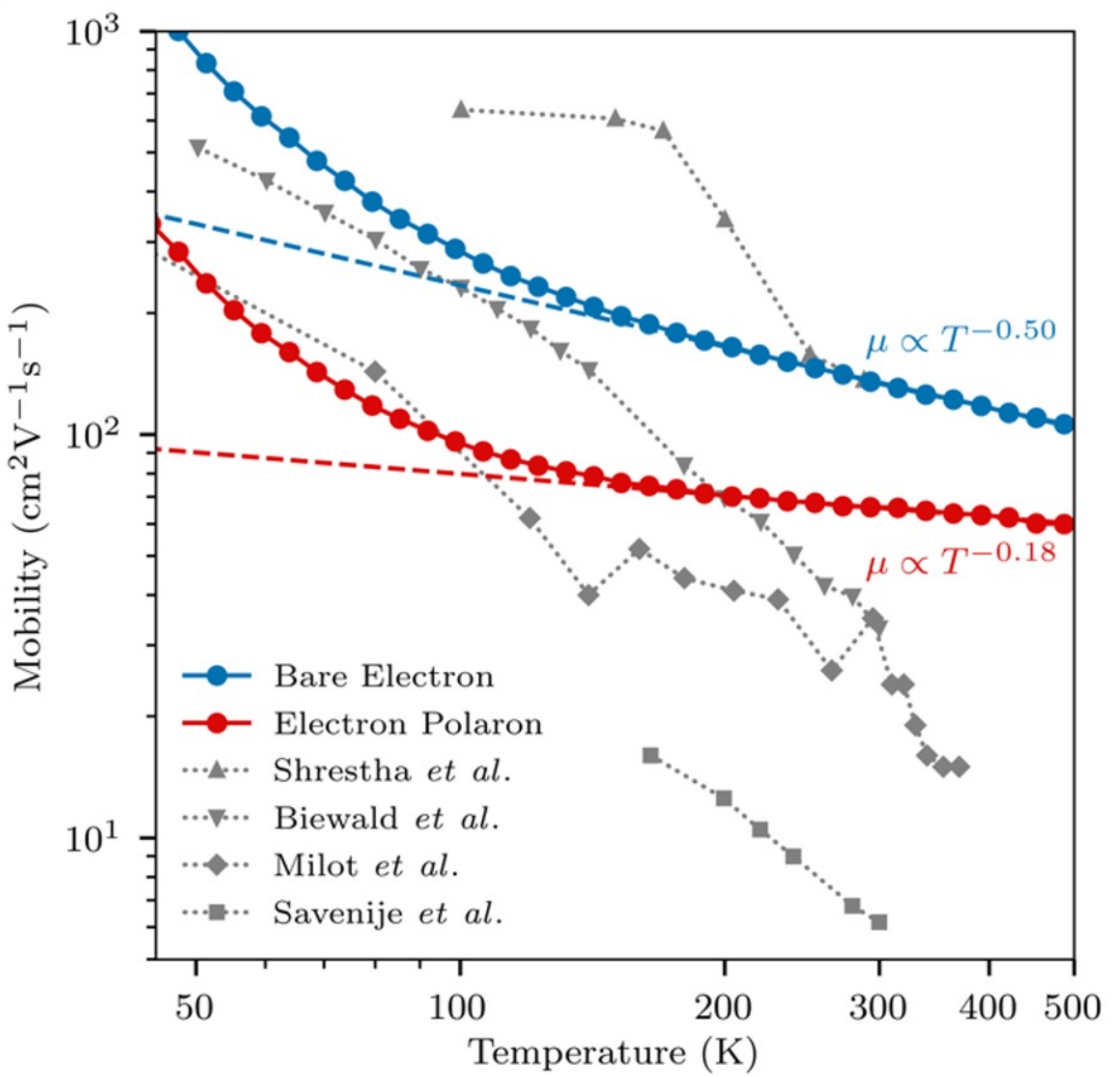


Solid lines: large electron polarons
Dashed lines: band electrons

- Acoustic phonon scattering rate most affected by polaron formation
- Negligible polaron influence on polar optical and impurity scattering rates
- Difference in scattering rates between bare band electrons and polarons reduced at 300 K cf 100 K.
- Polar optical phonon scattering dominates for band electrons and polarons.

L A D Irvine et al, *Phys Rev B* **103**, L220305 (2021)

Predicted mobility vs temperature



- For bare band electrons at T > 200 K $T^{-0.50}$ dependence (optical phonons)
- Polaron mobility at T > 200 K ~ $T^{-0.18}$
- $MAPbI_3$ experimental mobility ~$T^{-1.5}$

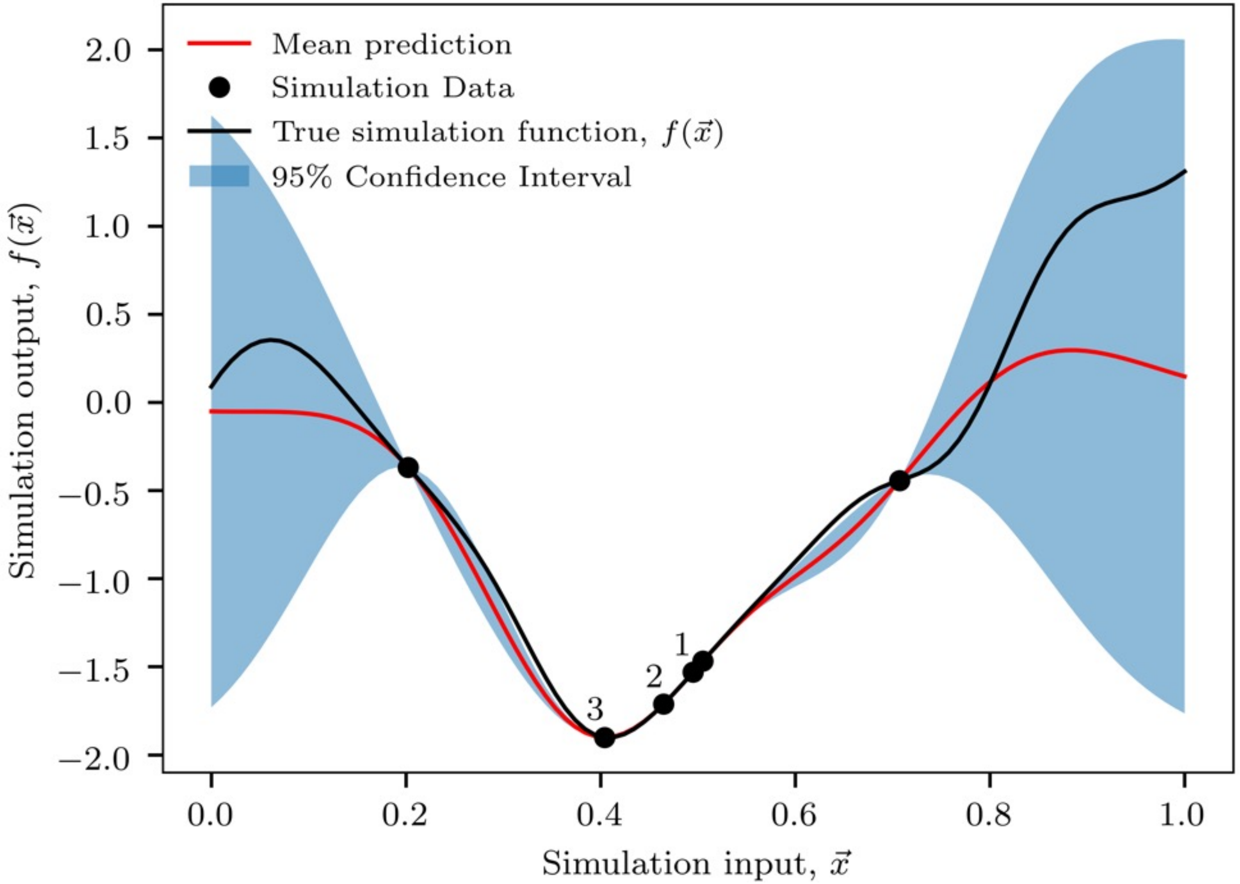
- Polaron formation does not have a large impact on carrier transport
- $MAPbI_3$ unusual amongst perovskites in showing a large discrepancy with our model.

L A D Irvine et al, *Phys Rev B* **103**, L220305 (2021)

Machine Learning


Sam McCallum


Jamie Lerpiniere



Example for single simulation input showing how minimum of true simulation function can be found

- Bayesian optimization, BO: strategy for minimizing BoltMC predictions for the mobility temperature exponent
- Input parameters vary within ±20% of best estimates
- BO uses Gaussian Processes to predict mean mobility temperature exponent from BoltMC and variance of GP prediction
- Minimum predicted exponent is – 0.69
- Probability of measured exponent (– 1.5) is 10^{-24}

Key parameters for estimating mobilities

- The Inverse length scale is a measure of the sensitivity of the mobility with respect to input parameter
- Acoustic and optical phonon scattering rates depend on dielectric constants at infinite and zero frequency, effective mass, elastic constant, polar optical phonon frequency, deformation potential
- ML tells us to which parameters the band electron mobility and electron polaron mobility are most sensitive

Summary

- Multiscale models are essential for understanding cell performance
- Kinetic models and Monte Carlo simulations bridge atomistic and macroscopic length scales.
- We can build up the complexity of the model systematically
- Machine Learning, ML, is a new tool to determine critical mechanisms responsible for experimental observations
- ML can be applied to models at all length scales with varying computer resource requirements

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