Multi-scale modelling of carrier capture and recombination in halide perovskite solar cells

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Collaborator: Sam Stranks (Physics, University of Cambridge)

Abstract
Metal halide perovskites are at the forefront of a new class of “soft crystalline semiconductors” with applications in optoelectronic devices. The vibrational and defect behaviour of these materials defy established conventions for inorganic semiconductors with large thermal displacements, strong phonon-phonon interactions, and light-activated defect formation and redistribution. Recent reports include anomalous electrostrictive and photostrictive effects [1]. New models are required to provide insights into the underlying physics and identify avenues to engineer this behavior into useful functionality.

We have maintained an international lead in the theory of halide perovskites over the past 6 years. Up to now, we have attacked opposite ends of the relevant length scales, ranging from Monte Carlo models of polar domain formation to the many-body electronic structure [1]. This project will focus on bridging first-principles calculations and device-level simulations of defect-mediated electron-hole recombination. This requires a description of electron-phonon coupling, the statistics of carrier recombination, and the dynamics of charge transport and collection in solar cells. The ultimate goal is to develop a model that enables rapid screening of “defect tolerance” in novel semiconducting materials.

1. Recent review presentation on halide perovskites: https://speakerdeck.com/aronwalsh/unsolved-mysteries-of-halide-perovskites

What is the multi-scale nature of the project?
This project is inherently multiscale. We will link macroscopic device behaviour to the nature of point defects at an atomistic level. Relaxation of photo-excited charges occurs on a femtosecond timescale, the response of the lattice and the formation of polaron on a picosecond timescale, and then the device behaviour is on a second timescale, with a micron-scale feature size. The large parameter space and problem complexity makes supervised machine learning models attractive, which will be tested as a way to accelerate bridging theory and experiment.

How do the expertises of the supervisors complement each other?
Walsh has a strong background in computational materials chemistry and semiconductor physics, while Barnes has an expertise in device measurements and simulations (e.g. the drift diffusion package https://github.com/barnesgroupICL/Driftfusion). The two ICL supervisors have a demonstrated track record of successful projects. The external partner Stranks is developing a novel approach for photoluminescence tomography that will allow the identification of trap centres in photovoltaic devices and will provide valuable data for training our recombination models.

Literature Review
Title: Electron-hole recombination in perovskite solar cells

Metal halide perovskites are the breakthrough solar energy materials of the 21st century. They efficiently convert sunlight to electricity, and can be deposited at low cost using scalable processing.
The main function of a photovoltaic device is to separate the electrons and holes generated in a semiconductor upon illumination. The maximum current and voltage that can be extracted depends on the rates of electron and hole recombination. The kinetics of recombination are usually fitted to a rate equation that includes one- (defect), two- (radiative) and three-electron (Auger) components. This literature review will cover the understanding of the relative rates in the latest generation of perovskite solar cells and how they can be influenced by engineering defects and doping.

2. Instilling defect tolerance in new compounds, Nat. Materials 16, 964 (2017)

**MSc Project**

The MSc component will consist of three parts: (i) building a literature database on carrier capture cross-sections and recombination rates in semiconducting materials; (ii) parameterising our in-house code CarrierCapture based on density functional theory calculations; (iii) validating the method and assessing statistical models (e.g. gradient boosted regression) for linking bulk materials properties and predicted recombination rates.