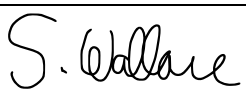
Appendix B: Statement of Authorship

<table>
<tr><th colspan="10">This declaration concerns the article entitled:</th></tr>
<tr><td colspan="10">Atomistic insights into the order-disorder transition in Cu2ZnSnS4 solar cells from Monte Carlo simulations

Pages 88 - 98 of thesis</td></tr>
<tr><td colspan="10">Publication status (tick one)</td></tr>
<tr><td>draft manuscript</td><td></td><td>Submitted</td><td></td><td>In review</td><td></td><td>Accepted</td><td></td><td>Published</td><td>X</td></tr>
<tr><td>Publication details (reference)</td><td colspan="9">Atomistic insights into the order-disorder transition in Cu2ZnSnS4 solar cells from Monte Carlo simulations
SK Wallace, JM Frost, A Walsh
Journal of Materials Chemistry A, 2019, DOI: 10.1039/C8TA04812F</td></tr>
<tr><td>Candidate's contribution to the paper (detailed, and also given as a percentage).</td><td colspan="9">Formulation of ideas (50%): The original code used in this work was provided by J. Frost and the idea to investigate Cu-Zn disorder in Cu2ZnSnS4 (CZTS) was provided by A. Walsh. Ideas for subsequent developments of the code and analysis of the data were mostly formulated and implemented by myself (S. Wallace) with support and discussions provided by J. Frost and A. Walsh throughout this study.

Design of methodology (50%): The Bespoke Monte Carlo model, Eris, (doi: 10.5281/zenodo.1248445) developed for this publication is based on the Starry Night code (doi: 10.5281/zenodo.10543) written by J. Frost to simulate dipole-dipole interactions and ferroelectric domains in a hybrid organic-inorganic perovskite solar cell. Eris was originally adapted by J. Frost from Starry Night to simulate thermodynamic Cu-Zn disorder in CZTS. Developments of the Eris code by S. Wallace include writing routines for:
• Lattice initialisation methods to ensure a stoichiometric CZTS crystal
• Testing the convergence in the electrostatic summations for calculating the change in lattice energy when performing a Monte Carlo move
• Developing and testing methods to check that the equilibrium disordered configuration was achieved for each simulation temperature
• Additional outputs from the code including: data to compute an order parameter, tests for convergence when computing on-site electrostatic potentials and outputting lattice on-site electrostatic potentials in various forms for further analysis
• Extending the model to include Cu/Zn disorder between the planes, i.e. allowing Zn from Cu-Zn layers to substitute onto Cu sites in the Cu-Sn planes

Experimental work (100%): All data generated from the Eris code that was used in the publication was produced by S. Wallace. Contributions from implementation of the code include running simulations to test for finite size effects in the model, generating data and developing post-processing tools to quantify and visualise Cu-Zn disorder in the system.

Presentation of data in journal format (70%): The first draft of the manuscript was prepared by S. Wallace and support was provided by the co-authors when finalising the manuscript for submission.</td></tr>
<tr><td>Statement from Candidate</td><td colspan="9">This paper reports on original research I conducted during the period of my Higher Degree by Research candidature.</td></tr>
<tr><td>Signed</td><td colspan="5">  </td><td>Date</td><td colspan="3">05/12/18</td></tr>
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