Modelling Defect Energy Surfaces with DFT and Machine Learning

First-principles simulations of atomic and electronic structure in solids offer a powerful route to predict and understand material properties.[1,2] This is particularly relevant in the case of point defects which dramatically affect material properties yet present many challenges for experimental characterisation.

Recent years have seen renewed appreciation for the potential complexity of defect energy surfaces in solids, and the significant impact that metastabilities can have on predicted and observed behaviour.[3–5] I will discuss my collaborative efforts in these areas, including the development of global optimisation strategies,[6,7] case studies where defect metastabilities are crucial to macroscopic properties (such as charge compensation and carrier recombination),[8–10] and remaining challenges in this area. Lastly, I will discuss recent work on extending these approaches using machine-learning foundation models,[11–13] demonstrating exciting potential for these methods in the field of defect modelling, but with important caveats regarding their accuracy and reliability at present.

1. Freysoldt, C. *et al.* First-principles calculations for point defects in solids. *Rev. Mod. Phys.* **86**, 253–305 (2014).
2. Squires, A. G., Kavanagh, S., Walsh, A. & Scanlon, D. Guidelines for robust and reproducible point defect simulations in crystals. Preprint at https://doi.org/10.26434/chemrxiv-2025-3lb5k (2025).
3. Mosquera-Lois, I. & Kavanagh, S. R. In search of hidden defects. *Matter* **4**, 2602–2605 (2021).
4. Kononov, A., Lee, C.-W., Shapera, E. P. & Schleife, A. Identifying native point defect configurations in α-alumina. *J. Phys.: Condens. Matter* **35**, 334002 (2023).
5. Guo, X., Huang, M. & Chen, S. ${\mathrm{Si}/\mathrm{Si}\mathrm{O}}_{2}$ MOSFET reliability physics: From four-state model to all-state model. *Phys. Rev. Appl.* **24**, 044040 (2025).
6. Mosquera-Lois, I., Kavanagh, S. R., Walsh, A. & Scanlon, D. O. ShakeNBreak: Navigating the defect configurational landscape. *Journal of Open Source Software* **7**, 4817 (2022).
7. Mosquera-Lois, I., Kavanagh, S. R., Walsh, A. & Scanlon, D. O. Identifying the ground state structures of point defects in solids. *npj Comput Mater* **9**, 1–11 (2023).
8. Kavanagh, S. R., Scanlon, D. O., Walsh, A. & Freysoldt, C. Impact of metastable defect structures on carrier recombination in solar cells. *Faraday Discuss.* **239**, 339–356 (2022).
9. Kavanagh, S. R., Walsh, A. & Scanlon, D. O. Rapid Recombination by Cadmium Vacancies in CdTe. *ACS Energy Lett.* **6**, 1392–1398 (2021).
10. Kavanagh, S. R. *et al.* Intrinsic point defect tolerance in selenium for indoor and tandem photovoltaics. *Energy Environ. Sci.* **18**, 4431–4446 (2025).
11. Kavanagh, S. R. Identifying split vacancy defects with machine-learned foundation models and electrostatics. *J. Phys. Energy* **7**, 045002 (2025).
12. Mosquera-Lois, I., Kavanagh, S. R., Ganose, A. M. & Walsh, A. Machine-learning structural reconstructions for accelerated point defect calculations. *npj Comput Mater* **10**, 1–9 (2024).
13. Tan, C. W. *et al.* High-performance training and inference for deep equivariant interatomic potentials. Preprint at https://doi.org/10.48550/arXiv.2504.16068 (2025).