

Multiscale models for perovskite optimisation

Philippe Baranek^{1,2}, Antoine Gissler^{1,2}, Philip Schulz³,
Michel Rérat⁴ and Roberto Dovesi⁵, James P. Connolly⁶

¹EDF R&D, EDF Lab Paris-Saclay, Department SYSTEME, 7 boulevard Gaspard Monge, F-91120
Palaiseau, France

²IPVF, Institut Photovoltaïque d'Ile de France, CNRS, Ecole Polytechnique, 18 boulevard Thomas Gobert,
91120, Palaiseau, France

³Institut Photovoltaïque d'Ile-de-France (IPVF), UMR 9006, CNRS, Ecole Polytechnique, IP Paris, Chimie
Paristech, PSL, 18 boulevard Gaspard Monge, F-91120 Palaiseau, France

⁴Université de Pau et des Pays de l'Adour, E2S UPPA, CNRS, IPREM, 2 avenue du Président Pierre Angot,
F-64053 Pau, France

⁵Accademia delle Scienze di Torino, via Accademia delle Scienze, 6, I-10123 Torino, Italy

⁶GeePs, Group of Electrical Engineering Paris, CNRS, CentraleSupélec, Université Paris-Saclay, Sorbonne
Université, 3&11 rue Joliot-Curie, Plateau de Moulon, F-91192 Gif-sur-Yvette CEDEX, France

Abstract

Perovskite solar cells have progressed extremely rapidly from 3.8% efficiency¹ in standard test conditions in 2009 to 25.2% efficiency² in similar conditions in September 2024. Tandem efficiencies have furthermore breached the single-junction Shockley-Queisser efficiency limit, reaching a reported 34.6% efficiency³ in June 2024 awaiting certification. While this rapid increase is unmatched by any other technology, it remains crippled by stability issues, an obstacle for the industrial and societal application of these materials. This paper presents a multiscale approach to evaluate the solar cell performance using perovskite material properties determined at the atomistic scale from first-principles calculations, in device scale models. The atomistic models provide optical, electronic, phonon, and structural properties. These properties, including charge transport parameters, are coupled to a numerical drift-diffusion device model enabling evaluation of the performance of corresponding single junction devices. Concerning materials, we present results on both organic and inorganic perovskite materials. For the organic perovskites, a key element is stability implications of the effect of phase transitions on the nonlocal lattice distribution of organic moieties through the lattice. The materials phase transitions and chemical composition impacts on the corresponding materials properties are investigated. The first-principles simulation applies a hybrid exchange-correlation functional adapted to the studied family of compounds. Validation by available experimental data is presented from materials properties to device performance, justifying the use of the approach for predictive evaluation of existing and novel perovskites. The coupling between atomistic and device models is described in detail by presenting a framework for exchange of optical, vibrational, and electronic parameters between the two scales. While the main focus of this work is perovskite materials and therefore of single-junction perovskite solar cells, the extension to tandem solar cells is included given the importance of multijunction device exceeding single junction Shockley-Queisser efficiency limits. The result of this theoretical investigation is a methodology for designing and optimising perovskite materials for both cell performance and stability, the key obstacle in the societal implementation of these record-breaking new materials.

TOPIC 3 : Nouveaux matériaux, nanostructures, photonique

¹ A. Kojima, K. Teshima, Y. Shirai, T. Miyasaka, Organometal Halide Perovskites as Visible-Light Sensitizers for Photovoltaic Cells, *Journal of the American Chemical Society*. 131 (2009) 6050–6051. <https://doi.org/10.1021/ja809598r>

² Martin A. Green, Keith Emery, Yoshihiro Hishikawa and Wilhelm Warta, Ewan D. Dunlop, *Prog. Photovolt: Res. Appl.* 2024; 23:1–9 <https://doi.org/10.1002/pip.3831>

³ PV Magazine : <https://www.pv-magazine.com/2024/06/14/longi-claims-34-6-efficiency-for-perovskite-silicon-tandem-solar-cell/>