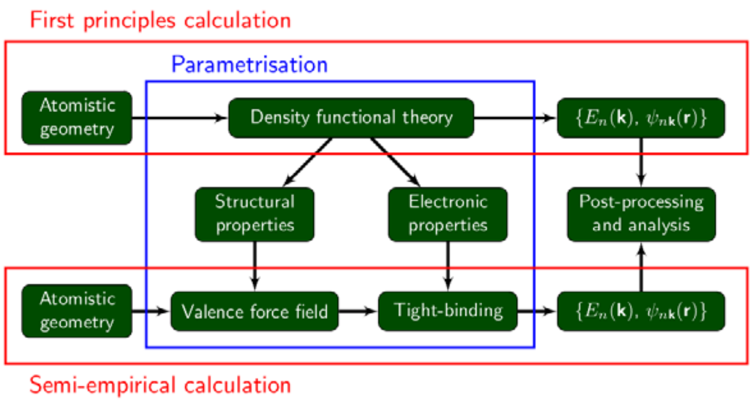
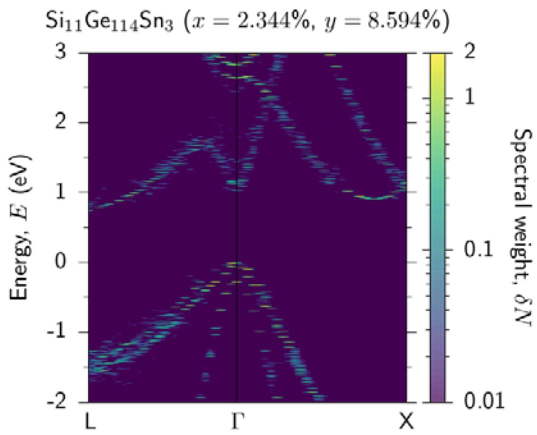
Acadamh Ríoga na hÉireann
Royal Irish Academy

Charlemont Grant Report

Recipient Name:	Dr Christopher Broderick
Discipline:	Sciences
Amount and year awarded:	€1,652 in 2019
Title of Project:	Theory and design of emerging semiconductor alloys for applications in high efficiency solar cells





Summary of findings:	1. Silicon-germanium-tin (SiGeSn) alloys: SiGeSn alloys, which can be grown lattice-matched – i.e. with the same lattice constant as – germanium (Ge) substrates while having band gaps close to 1 eV, represent a potentially suitable material system for applications in highly-efficient multi-junction solar cells. Growth of this emerging class of semiconductor alloys has only been established recently, and to date there has been little detailed analysis of the evolution of the alloy electronic structure. The research visits supported by this grant have helped to establish this understanding, by facilitating close interactions between theory and experiment. The key findings of this research are related to direct comparison between theoretical calculations carried out by the Awardee and experimental measurements carried out by collaborators at Imperial College London (ICL), U.K. for SiGeSn alloys. During two research visits to ICL the Awardee established a combination of first principles and semi-empirical calculations – based respectively on density functional theory (DFT), and on a combination of a valence force field (VFF) potential and atomistic tight-binding (TB) Hamiltonian – of the properties of realistic, disordered SiGeSn alloys. These calculations have been validated via comparison to experimental measurements, have revealed the rich nature of the alloy electronic structure, and have indicated material properties which are likely to have significant implications for device applications. Firstly, calculations for existing SiGeSn material samples – grown by industrial partner IQE PLC and characterised at ICL via photoluminescence, spectroscopic ellipsometry and photo-modulated reflectance measurements – have been used to elucidate the origins of the alloy electronic structure based on features observed in the optical characterisation experiments. A key finding has been the identification of strong alloy band mixing (hybridisation) effects, leading to a strongly perturbed alloy band structure close in energy to the conduction band minimum. Band mixing effects of this type, which are minimal in "conventional" semiconductor alloys, have significant implications for the alloy properties. In SiGeSn we find that such effects are prominent close in energy to the conduction band edge, and can hence be expected to have strong impact on

	several material parameters relevant to solar cell applications. Specifically, in electronic structure calculations for disordered alloy supercells we observed strong inhomogeneous energy broadening of the Bloch character associated with zone-centre states. This will have significant impact for device applications, leading to spectrally broad optical absorption in addition to electron mobility limited by strong alloy scattering. Having identified this key characteristic of the alloy electronic structure, collaborative research is ongoing to quantify the associated implications for the alloy optical and transport properties. In response to initial analysis undertaken during the two research visits to ICL, new SiGeSn material samples have been grown by IQE PLC and provided to ICL for experimental characterisation. This experimental data has been passed to the Awardee, who is performing further theoretical calculations with the aim of identifying alloy composition ranges providing optimum material properties for solar cell applications. Plans regarding this ongoing work are outlined below. Secondly, beyond the range of Si and Sn compositions currently achievable via epitaxial growth – respectively < 10% and 5% - the Awardee has established first principles and semiempirical calculations of the structural and electronic properties of SiGeSn alloys across the entire composition range. This has enabled the precise ratio of Si to Sn composition to achieve lattice matching to Ge – or, alternatively, a desired level of lattice-mismatch in strained epitaxial layers – to be determined quantitatively. Beyond the composition ranges of interest for solar cells, the calculations established and benchmarked via this collaboration will inform ongoing analysis of SiGeSn alloys in the context of the development of Sicompatible semiconductor lasers (a key technological requirement to facilitate the development of Si photonics). Given the strong recent growth of interest in group-IV semiconductor alloys – driven by their potential for applications in several classes of semiconductor devices – it is expected that the fundamental understanding being developed by the theory experiment collaboration initiated through this award will be influential in informing research on SiGeSn alloys in the coming years. 2. Software development: An additional goal of the research collaboration with ICL was to advance work by the Awardee towards the development of a new materials science module for the open-source Solcore solar cell simulation library (http://solcore.solar). This new module is based upon software developed by the Awardee over the past several years, which provides a reciprocal space (plane wave) implementation of a multi-band k.p envelope function solver for arbitrary multiple quantum well heterostructures based on conventional III-V semiconductor alloys, as well as highly mismatched semiconductor alloys containing nitrogen (N) and bismuth (Bi). This will develop several critical new capabilities to Solcore, which is currently not capable of performing such calculations or treating highly-mismatched semiconductor alloys (the latter being of interest for multi-junction solar cell applications). The visits facilitated by this award allowed for detailed planning of integration between the Awardee's software and the Solcore library, in terms of (i) porting the Awardee's material parameter database for highly-mismatched alloys, and (ii) determining strategies for including the Awardee's solver in Solcore. With these plans formalised, software development by the Awardee is ongoing.
Plans for continuing collaboration:	The collaborations initiated with the support of this award are ongoing. 1. Electronic and optical properties of SiGeSn alloys: The research visits supported by this award enabled the Awardee to establish and benchmark first principles and semi-empirical calculations of the electronic structure of SiGeSn alloys. The Awardee is continuing to work on theoretical calculations of the electronic and optical properties of SiGeSn alloys, while experimental collaborators at ICL are continuing to provide experimental data for new SiGeSn material samples grown and provided by industrial partner IQE PLC. This ongoing collaboration is helping to refine the Awardee's semiempirical

	simulation framework for SiGeSn alloys, and will therefore underpin future calculations related to applications of SiGeSn alloys not only in solar cells, but also in related devices such as semiconductor lasers. Since the two research visits by the Awardee to ICL remote collaboration has continued, with exchanges of information taking place via email, teleconferencing and electronic data-sharing. Both the Awardee and the key experimental collaborator from ICL, Ms. Phoebe Pearce, are attending the 2020 SPIE Photonics West conference in early February 2020. This will provide the opportunity to have further detailed technical discussions, as well as to finalise work on an in-progress collaborative paper (described below). 2. Development of open-source software: As described above, software development by the Awardee is ongoing. In addition to the software integration described above, this development also requires (i) the translation by the Awardee of key pieces of existing software from C to Python (Solcore being written in the latter) to minimise external dependencies in the distributed software, and (ii) post-processing of data produced by the Awardee's plane wave calculations of the electronic properties of alloyed III-V heterostructures, to allow for interoperability with Solcore's suite of optical and electrical characterisation routines. It is currently planned to complete a beta version of this new module by the end of Q1 2020. This beta version will be distributed to a small group of Solcore users in Q2 2020 for testing and feedback. Release of this module as part of the main Solcore library is planned in Q3 2020, and it is planned that an associated journal paper be prepared in Q3 2020 to describe the new capabilities of the library (described below).
Publication plans:	There are a total of four in-progress and planned publications associated with research supported by this award. 1. SiGeSn alloys: Sufficient data has been produced to facilitate the writing of a collaborative theory-experiment paper on the electronic and optical properties of SiGeSn alloys, and the implications of these properties of multi-junction solar cells incorporating a 1 eV SiGeSn junction. Work on this paper has begun, and it is planned that this will be completed and submitted in Q1 2020. Additionally, the Awardee plans to write a standalone theoretical paper describing the evolution of the SiGeSn alloy electronic structure across the full composition range, identifying the consequences of the alloy electronic structure for alternative applications – e.g. Si-compatible semiconductor lasers – where broader composition ranges are of interest. 2. Software development: Upon completion of the development and testing of the new Solcore materials science module, it is planned to write a paper (i) describing the capabilities of the new module, (ii) describing the underlying theoretical formalism, and (iii) providing several classes of examples relevant to practical use of Solcore in the design of prototype solar cells. This is a longer-term project: the theoretical formalism is already in place, as well as suitable reference results for example

	calculations (computed using existing software, which will be used in the first instance to benchmark the new Solcore module). However, completion of this paper will not occur until after distribution and validation of a beta version of the software in Q2 2020. 3. Book chapter: The Awardee is currently lead author on an invited book chapter on the theory of the electronic structure of semiconductor materials. This chapter forms part of a book, being edited by Dr. Diego Alonso-Álvarez at ICL (lead developer of Solcore), having the tentative title Modelling of Solar Cells and Semiconductor Materials. The aim of this book is to provide not only a contemporary review of theory pertinent to the design of novel solar cell technologies in a research context, but also to accompany this exposition with open-source software examples which can be downloaded and employed by readers as a practical learning tool. Discussions held during the Awardee's visits to ICL have contributed to the planning and (ongoing) writing of this book chapter, which is expected to be completed in Q1 2020 (with publication of the complete book expected in late 2020).
International dissemination:	The results of this ongoing research collaboration have not yet been presented at any international conferences. The first presentation of these results at an international conference will take place in February 2020 at the SPIE Photonics West conference in San Francisco, U.S.A. where the Awardee will be presenting a talk entitled "Multiscale modelling of group-IV semiconductor alloys: localisation, hybridisation and implications for device applications". The Awardee plans to attend three other prominent international conferences in 2020: the International Conference on Numerical Simulation of Optoelectronic Devices (NUSOD), UK Semiconductors (UKSC) and the 35th International Conference on the Physics of Semiconductors (ICPS). The Awardee plans to present the results of their ongoing programme of research on group-IV semiconductor alloys at each of these meetings, including key results arising from the collaboration initiated via this award.
National dissemination:	The results of this ongoing research collaboration have not yet been presented at any Irish conferences. The primary Irish conference relevant to the Awardee's research is Photonics Ireland, which is next scheduled to take place in late summer of 2020. The Awardee plans to attend this conference and present the results of their ongoing research on group-IV alloys, including key results arising from the collaboration initiated via this award.
Additional collaborations:	The primary experimental measurement and analysis related to this collaboration has been performed by Ph.D. student Ms. Phoebe Pearce at ICL. Ms. Pearce's research is supported by industry, via IQE PLC, who have been undertaking epitaxial growth of SiGeSn alloys and supplying material samples to faciltate experimental investigations. Recently, the senior collaborator at ICL, Dr. Nicholas J. Ekins-Daukes (doctoral supervisor to Ms. Pearce), has taken up the position of Associate Professor with the School of Photovoltaic and Renewable Energy Engineering (SPREE) at the University of New South Wales (UNSW), Australia. Dr. Michael Neilsen, a postdoctoral researcher working under the supervision of Dr. Ekins-Daukes at UNSW, has contributed to this research collaboration by undertaking optical

	characterisation (photo-modulated reflectance) measurements of SiGeSn samples supplied by IQE PLC. This data has provided valuable additional information on the alloy electronic and optical properties, complementing the photoluminescence and spectroscopic ellipsometry measurements and analysis undertaken at ICL by Ms. Pearce. Analysis of the data arising from these photomodulatedreflectance measurements is being undertaken on an ongoing basis at ICL by Ms. Pearce. Structural characterisation of the SiGeSn samples grown and provided by IQE PLC has been undertaken at the National University of Singapore, Singapore, via x-ray diffraction, transmission electron microscopy and Rutherford backscattering. These measurements have provided accurate information regarding alloy composition and microstructure, thereby providing valuable information to guide input to the Awardee's theoretical calculations.
Outreach:	The Awardee manages the Twitter account of the Photonics Theory Group (PTG) at Tyndall National Institute, University College Cork. This account is used to promote research activities and outputs from the PTG, as well as to promoting outreach undertaken by PTG members: http://twitter.com/TyndallPTG Upon receipt of this award, the Awardee coordinated with University College Cork to publicise both the award and associated research: https://www.ucc.ie/en/research/news-events/news/dr-christopher-broderick-awarded-charlemont-grant.html