Efficient ab initio electronic transport methods

Z. Li, P. Graziosi*, and N. Neophytou
School of Engineering, University of Warwick, Coventry, CV4 7AL, UK
*CNR-Bologna, Italy
e-mail: n.neophytou@warwick.ac.uk

INTRODUCTION

Recently there has been an increase in efforts to compute the electronic properties of complex electronic structure materials for many applications, e.g. novel devices and energy materials. The typical process is to use the Boltzmann Transport equation (BTE) in the relaxation time approximation, while scattering rates are extracted at various degree of complexity and accuracy. The most accurate fully ab initio methods, however, are computationally extremely expensive and rarely used. Here we present a method to compute electronic transport from first principles in materials of arbitrary bandstructure.

METHODS

Typical ab initio calculations use Density Functional Theory (DFT) to extract the electronic structure, and then Density Functional Perturbation theory (DFPT) to extract the phonon attributes and the electron-phonon matrix elements. Dense electronic and phonon meshes are needed, which often are a result of interpolations, and up to billions of matrix element calculations are required. This makes the calculation very expensive. Scattering rates are then extracted and are used within the BTE. Instead of computing billions of matrix elements, we follow a practical approach, where we identify and compute a small relevant sample of required matrix elements, and use those to compute deformation potentials involving all relevant phonon modes. We then form the usual deformation potential scattering expressions to use within a BTE which considers numerically the full band electronic structure.

RESULTS

We show this for Si as an example in Figs 1-3 [1]. Only a narrow energy region around the electronic band extrema contributes to transport (red regions in Fig. 1), and only a small region in the phonon spectrum meets energy/momentum conservation to facilitate transitions (red regions in Fig. 2). The corresponding matrix elements are shown in Fig. 3 for some valence (LA-blue and LO-red modes) and conduction (LO-green mode) band transitions. The slope of the LA mode will give the deformation potential for acoustic modes, while the value of the matrix element itself, that for optical modes. The green line clearly captures the fact that LO transitions are only inter-valley in Si (it is close to zero at Γ and finite at *g*). We use the deformation potentials to form scattering rates, and then use then within *ElecTra*, a full band BTE that we have developed [2]. The code takes into account all relevant transitions (intra/inter-band), for all relevant scattering mechanisms (elastic, inelastic, isotropic, anisotropic), as illustrated in Fig. 4. The computed Si hole mobility, entirely from first principles, is shown in Fig. 5, with excellent match to experiment (blue-squared line).

The method can be applied to materials with arbitrary band complexity. As an example we show in Fig. 6 the electronic and phononic bands for Mg_3Sb_2 (6a, b), and by identifying all relevant transition types between CBM energy surfaces (6c), we compute matrix elements for all phonon modes, indicting the dominant ones (box in 6d).

CONCLUSION

We presented an efficient and accurate ab initio method to extract electronic transport properties in materials with arbitrary band complexity.

ACKNOWLEDGMENT

This work received funding from the European Research Council (ERC) under the EU's H2020 research and innovation programme (GA 678763).

REFERENCES

[1] Z. Li, P. Graziosi, and N. Neophytou, *Deformation potential extraction and computationally efficient mobility calculations in silicon from first principles*, Phys. Rev. B **104**, 19, 195201 (2021).

[2] P. Graziosi, Z. Li, and N. Neophytou, *ElecTra code: Full band electronic transport properties of materials*, Computer Physics Communications **287**, 108670 (2023).

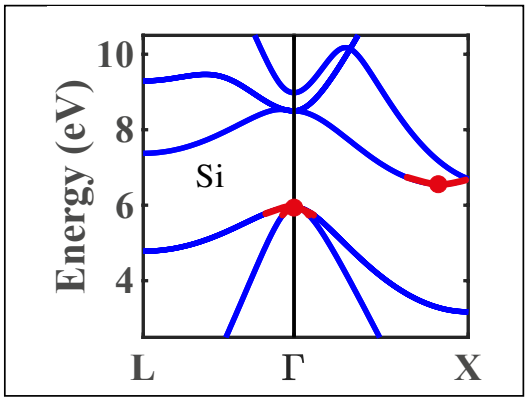

Fig. **1**. Electronic structure of Si with relevant transport regions indicated in red [1].

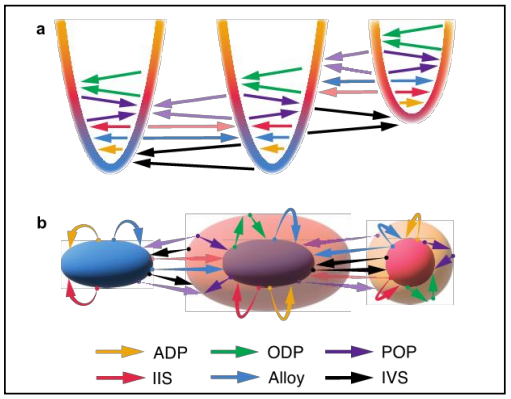

Fig. 4. Illustration of scattering transitions in *ElecTra* code: (a) intra/inter-valley and (b) elastic/inelastic transitions [2].

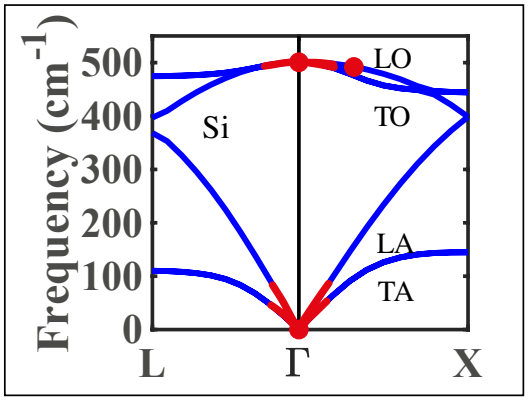

Fig. 2. Phonon spectrum of Si with scattering relevant regions indicated in red [1].

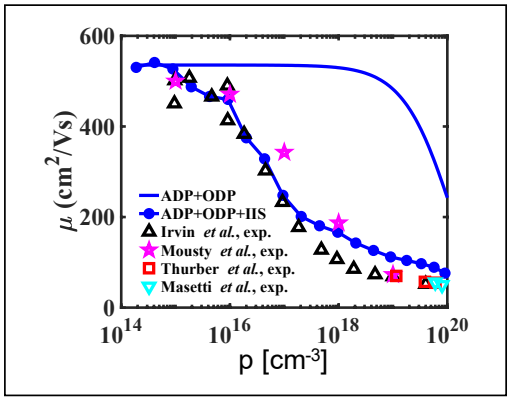

Fig. 5. Fully ab initio calculated Si hole mobility (blue lines) and comparison to experimental data [1].

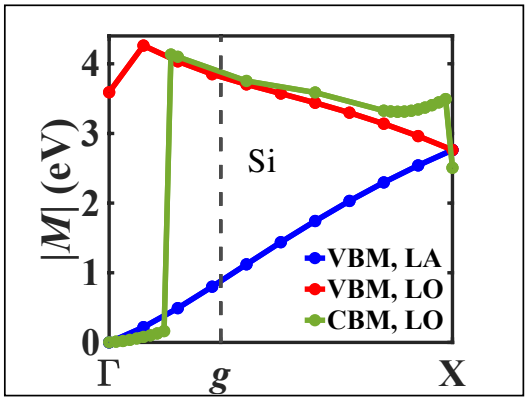

Fig. 3. Matrix elements involving LA and LO modes for the valence and conduction bands in Si [1]. For the CBM and LO mode, clearly the inter-valley nature of the *g*-process is evident (green line is zero close to Γ, and finite close to *g*)

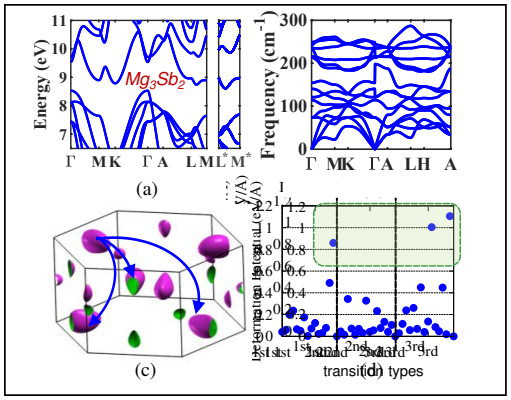

Fig. 6. Matrix element extraction in a complex band material Mg_3Sb_2. (a-b) Electronic and phononic structure. (c) Three dominant transition types. (d) Matrix elements for each transition type involving all phonon branches.