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CarrierCapture.jl: Anharmonic Carrier Capture

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The modern theory of defects in semiconducting materials allows for the accurate prediction of equilibrium point defect concentrations and transitions levels within the band gap from first-principles quantum mechanical simulations (Park, Kim, Xie, & Walsh, 2018). The procedures for such calculations are now well established (Freysoldt et al., 2014).

Beyond an equilibrium description, the operation and performance of optoelectronic devices, including solar cells and light-emitting diodes, relies on the interaction of points defects with non-equilibrium populations of electrons and holes. The non-radiative capture of charge carriers by lattice defects results in efficiency loss, which can range from suppressing luminescence to irreversible chemical degradation (Stoneham, 1981).

`CarrierCapture.jl` is designed to calculate the rates of carrier capture by point defects from first-principles data. It builds on a large body of well established theory (Stoneham, 1981), which was recently adapted to be compatible with quantities accessible from density functional theory (DFT) calculations (Alkauskas, Yan, & Van de Walle, 2014). In our implementation, we remove the harmonic approximation for the potential energy surfaces, which can be strongly asymmetric (Kim et al., 2019a).

A standard workflow involves building potentials and computing carrier capture coefficients. `CarrierCapture.jl` provides handy tools:

- Finding a best fit to the first-principles data
- Solving the one-dimensional Schrödinger equation for the potential energy surfaces
- Computation of the overlap between nuclear wave functions
- Computation of the capture coefficients as a function of temperature

We also provide auxiliary scripts to process the first-principles data. To our best knowledge, `carrierCapture.jl` is currently the only open-source package to provide such functionality. Common computational workflows that reproduce published examples (Kim et al., 2019a, 2019b) are available on Github pages. The API documentation including the guide to the installation is also up-to-date on GitHub pages.

Interfacing to other codes: A range of input parameters are required to describe the bulk and defective properties of the material. Scripts are provided to extract these from DFT calculations using `VASP`, but these can easily be modified to read data from other packages. Caution should to be taken in checking convergence with respect to calculation settings, e.g. basis sets and k-point sampling, as small errors in relative energies can change the resulting carrier capture cross-sections by orders of magnitude.

Author contributions

Sunghyun Kim wrote the majority of the code base with contributions from Samanth N. Hood. Lucy D. Whalley and Puck van Gerwen performed detailed code testing and contributed to the example and test suite. All authors along with Aron Walsh made decisions about code design and feature implementation. This manuscript was written with input from all co-authors.

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References

Alkauskas, A., Yan, Q., & Van de Walle, C. G. (2014). First-principles theory of nonradiative carrier capture via multiphonon emission. *Phys. Rev. B*, *90*(7), 075202. doi:10.1103/PhysRevB.90.075202

Freysoldt, C., Grabowski, B., Hickel, T., Neugebauer, J., Kresse, G., Janotti, A., & Van de Walle, C. G. (2014). First-principles calculations for point defects in solids. *Rev. Mod. Phys.*, *86*(1), 253. doi:10.1103/RevModPhys.86.253

Kim, S., Hood, S. N., & Walsh, A. (2019a). Anharmonic lattice relaxation during nonradiative carrier capture. *Phys. Rev. B*, *100*(4), 041202. doi:10.1103/PhysRevB.100.041202

Kim, S., Park, J.-S., Hood, S. N., & Walsh, A. (2019b). Lone-pair effect on carrier capture in $\mathrm{Cu_2ZnSnS_4}$ solar cells. *J. Mater. Chem. A*, *7*(6), 2686. doi:10.1039/C8TA10130B

Park, J. S., Kim, S., Xie, Z., & Walsh, A. (2018). Point defect engineering in thin-film solar cells. *Nat. Rev. Mater.*, *3*(7), 194–210. doi:10.1038/s41578-018-0026-7

Stoneham, A. (1981). Non-radiative transitions in semiconductors. *Rep. Prog. Phys.*, *44*(12), 1251. doi:10.1088/0034-4885/44/12/001