# Can the vibrational modes of silicon nanostructures be modelled with machine learning?

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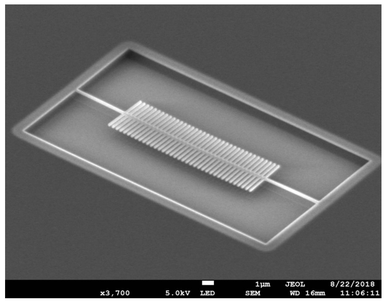



Figure 1: Scanning electron microscopy image (captured by Chris Reardon) of a silicon nanostructure which may display the thermoelectric effect strongly.

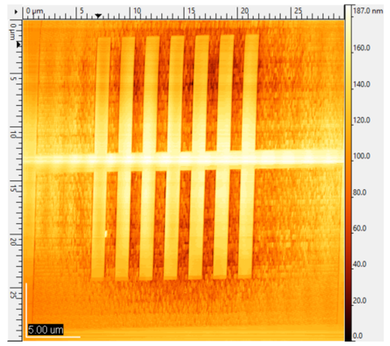



Figure 2: Atomic force microscopy image (by James Lees): silicon substrate consisting of a central shaft and multiple protrusions, which act as local resonators.

## Introduction

- **Thermoelectric materials** are able to convert temperature differences into electric voltage, but they need to be more cost-effective before they are adopted widely.
- Some **silicon-based, nanostructured materials** - shown in figures 1 and 2 - may be cost-effective solutions [1], but often have features that are too large to model using first-principles computational methods, while atomistic "force-field" approaches often fail to accurately simulate the thermal properties of nanostructures.
- To be effective an thermoelectric material, silicon must have its thermal conductivity significantly lowered.
- **Novel machine learning techniques** might provide a solution to modelling and designing large structures, while maintaining both the accuracy and speed of first-principles approaches.
- While avoiding assumptions based on empirical observations, machine learning techniques attempt to compress first-principles methods into accurate, predictive models that can simulate large systems efficiently.
- Here, we use a general-purpose interatomic potential for silicon - a **Gaussian approximation potential** (GAP) - to model silicon-based nanostructures.

## Further information

- **Email**: finn.hambly@york.ac.uk
- **Poster download & software source code**: github.com/finnhambly/fastphononics/poster.pdf

## Method

The GAP model was generated by Bartók *et al.* [2], and finds the energy of an atomic arrangement with equation 1:

$$E = \sum_{i<j} V^{(2)}(r_{ij}) + \sum_{i} \sum_{s}^{M} \alpha_s K(\mathcal{R}_i, \mathcal{R}_s), \quad (1)$$

where $i$ and $j$ cover the range of atoms in the system, $V^{(2)}$ describes the repulsion between a pair of atoms, $r_{ij}$ is the interatomic distance term, $\mathcal{R}_i$ describes the environment surrounding atom $i$, and $K$ is a smoothly-varying kernel function, which returns the similarity between $\mathcal{R}_i$ and atomic environments $\mathcal{R}_s$ in its database. $K$ returns a description of $\mathcal{R}_i$ with a linear combination of $\mathcal{R}_s$, with coefficients $\alpha_s$.

This GAP model was implemented within the Atomic Simulation Environment with quippy [3], and the structure phonon properties were calculated with Phonopy [4].

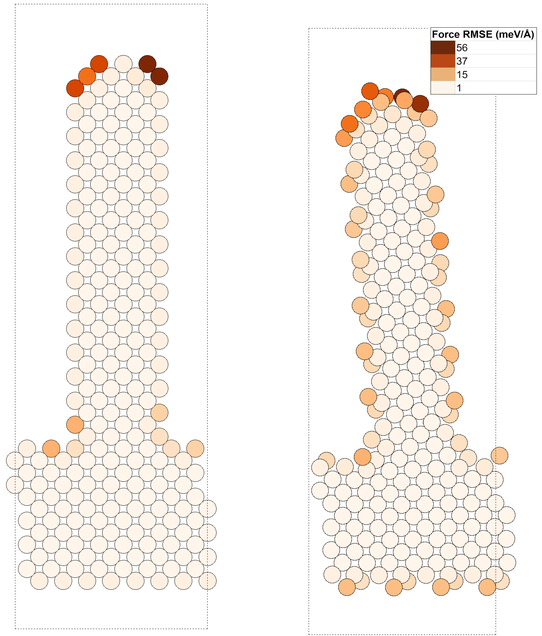
To verify the confidence of the GAP model in simulating a proposed structure, the predicted root mean square error (RMSE) of the atomic forces within the lattice is measured and plotted. The obtained predicted errors are consistent with the errors associated with first-principles-based calculations.

## References

1. Davis & Hussein, *Phys. Rev. Lett.* **112**,055505 (2014)
2. Bartók *et al.*, *Phys. Rev. X* **8**, 041048 (2018)
3. Csányi *et all. IoP Comput. Phys. Newsletter, Spring* (2007)
4. Togo & Tanaka, *Scr. Mater.*, **108**, 1-5 (2015)

## Structure optimisation

GAP-modelled walled silicon nanostructures, after structure optimisation, were found to reconstruct their surface atoms.




(a) Initial structure (b) After optimisation

Figure 3: Visualisation of a walled nanostructure, before and after a structure optimisation process under the GAP model. The root-mean-square error values for the force of each atom are illustrated by the intensity of an atom's colour.

These structures display imaginary phonon frequencies, which may be the result of neighbouring nanostructures nanostructures. This indicates they may be structurally unstable in the presence of certain vibrational modes, which is not found by Stillinger-Weber-modelled calculations (due to the lack of surface reconstruction in these models).

## Phonon band structure

Comparing the nature of the walled nanostructure's dispersion curves with those of a bare membrane, as shown in figure 4, reveals the influence of the wall on the structure's thermal conductivity.

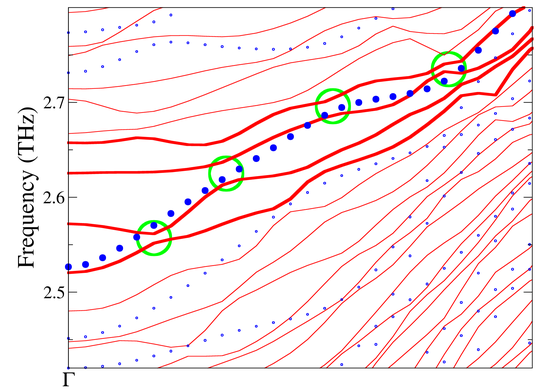



Figure 4: Portion of the silicon nanowall's phonon band structure superimposed onto the phonon band structure of the silicon membrane. The modes of a bare membrane are shown with blue circles and the nanowall's modes are in red. The four emboldened nanowall modes track the mode of the original membrane, but have hybridised (in green) in a manner that leads to a decreased mean group velocity, resulting in a lower thermal conductivity.

## Conclusions

- The GAP model finds that walled silicon nanostructures have a lower thermal conductivity than bare membranes.
- The GAP model's structure optimisation process was dominated by the energetic favourability of reconstructing surfaces.
- This led to nanostructures relaxing into geometries that were unstable, where simpler interatomic potential do not.