Recursive open boundary and interfaces method for material property predictions

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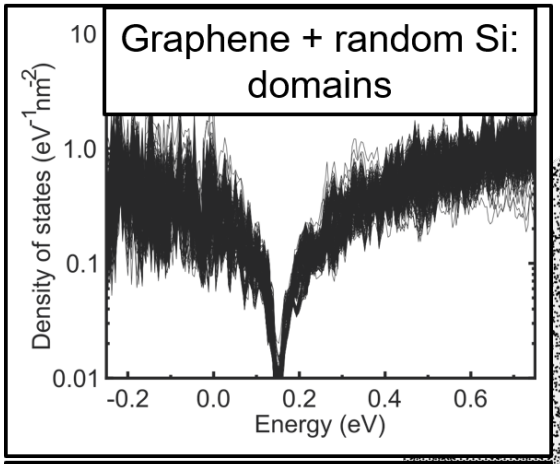
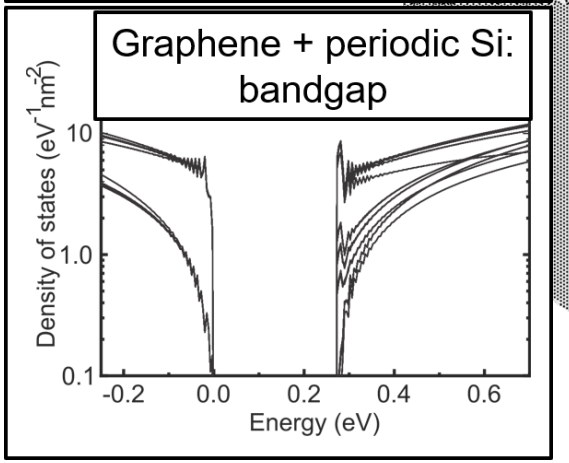
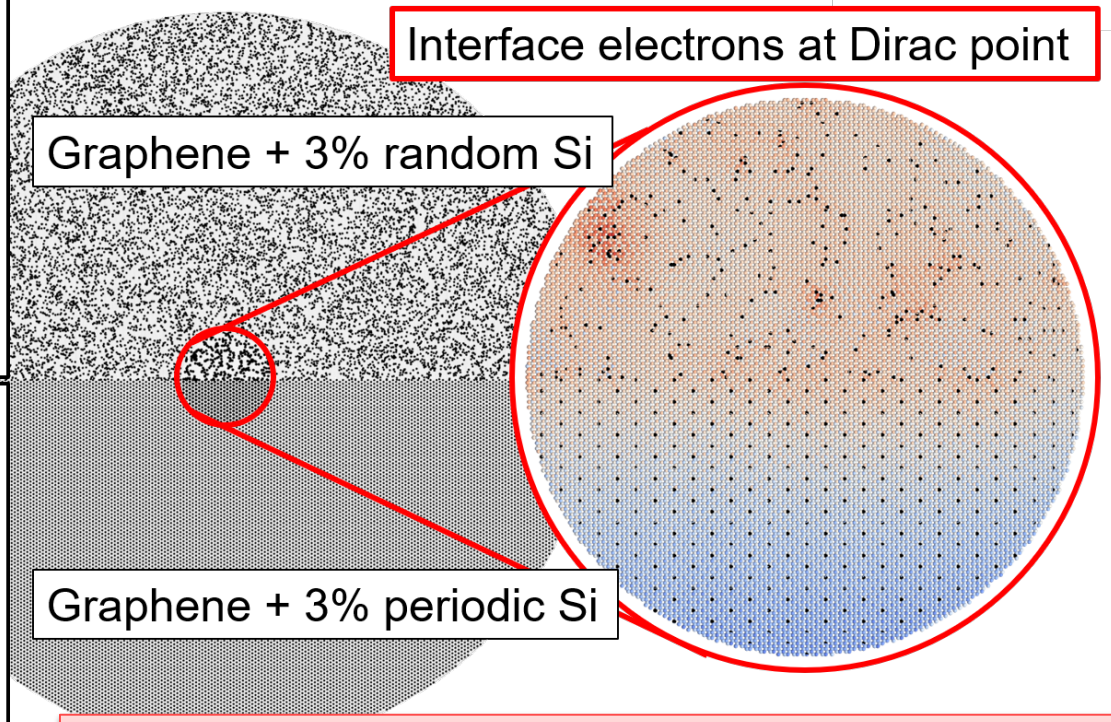
Highlight: Impact of periodicity assumption

Motivation: Periodic boundary conditions omnipresent in (material) modeling







Periodic boundary conditions boost small perturbations to systematic material change

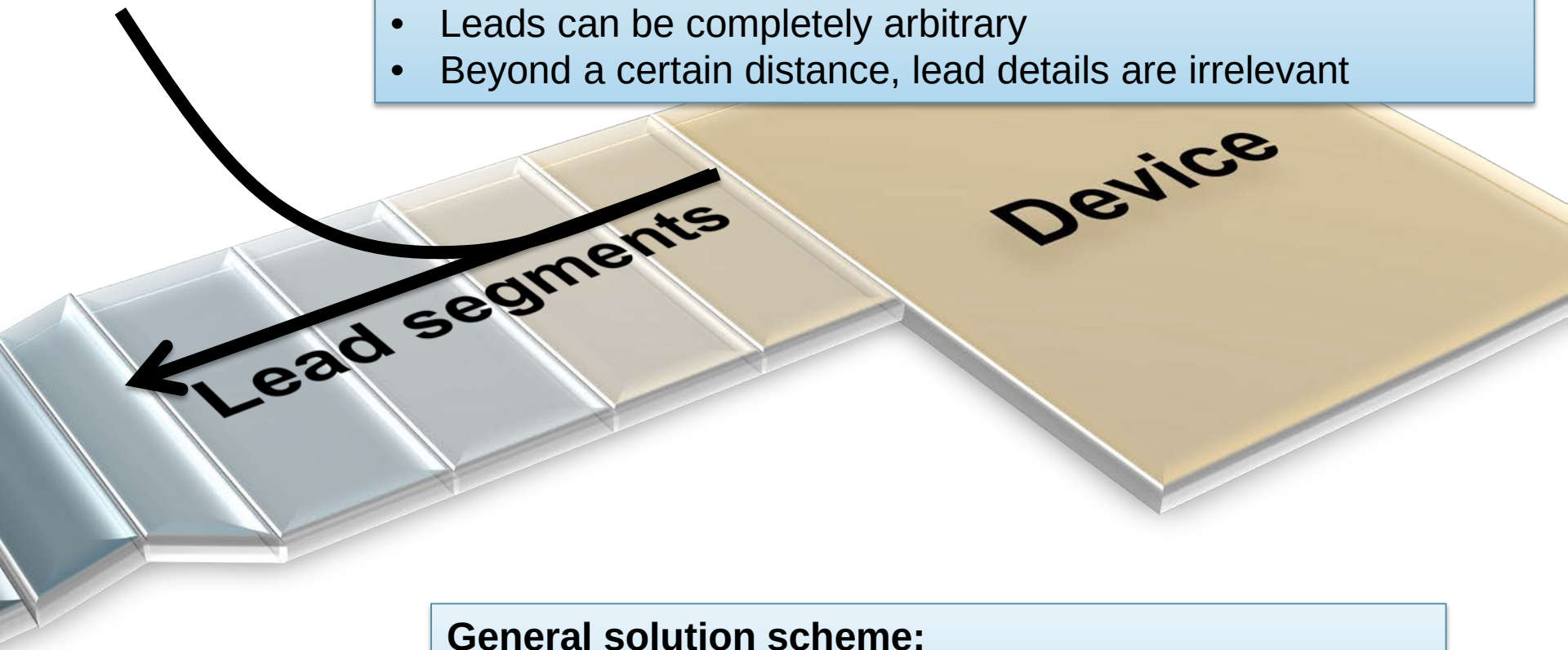
ROBIN – recursive open boundary and interfaces method

General: regular and irregular systems; Fast: handles millions of atoms

General iterative lead contact self-energy

This method:

- Adaption of complex absorbing potential (CAP) method (e.g. J. Driscoll et al, Phys. Rev. B. Vol. 78, pp. 245118, 2008)
- Leads can be completely arbitrary
- Beyond a certain distance, lead details are irrelevant



Utilizing uncertainty

General solution scheme:

Divide lead into segments

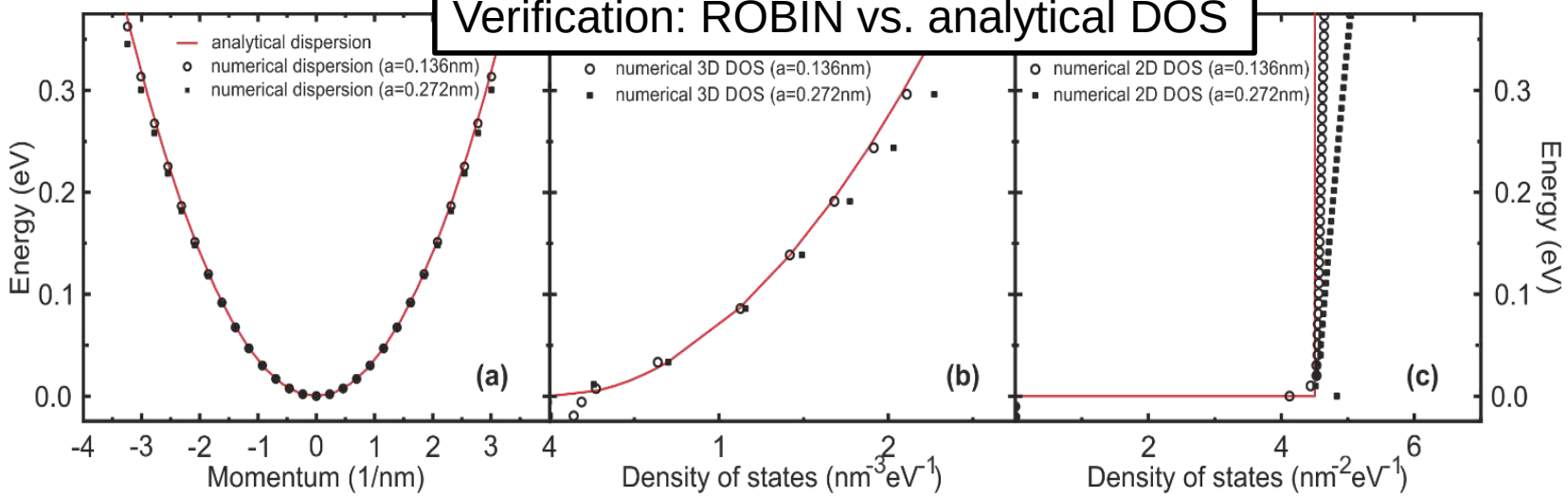
Add smooth damping potential , e.g. $V(R) = \exp(R\lambda)\, i\, V_0$

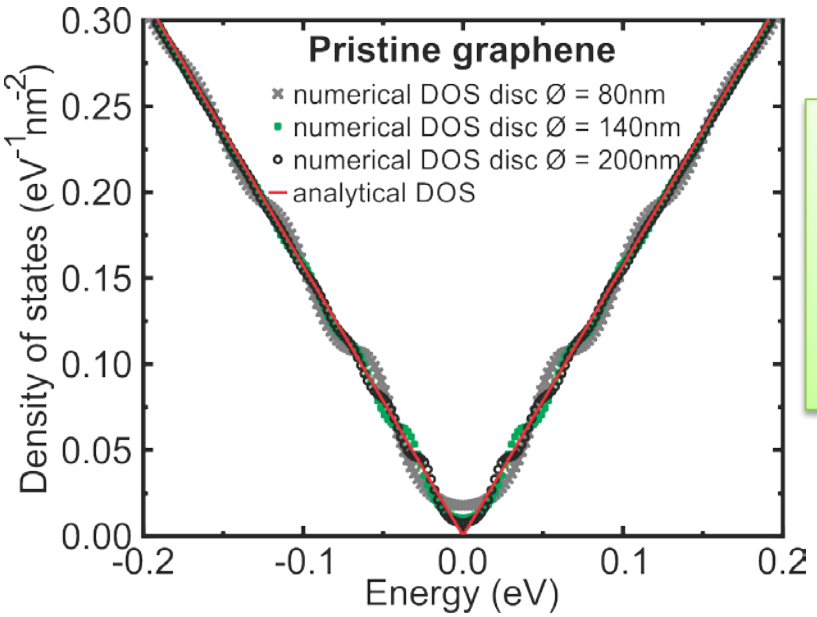
Apply RGF on lead surface Green's function

ROBIN method verification

ROBIN= Irregular lead method extended to 2D and 3D geometries, i.e. materials

Verification: ROBIN vs. analytical DOS



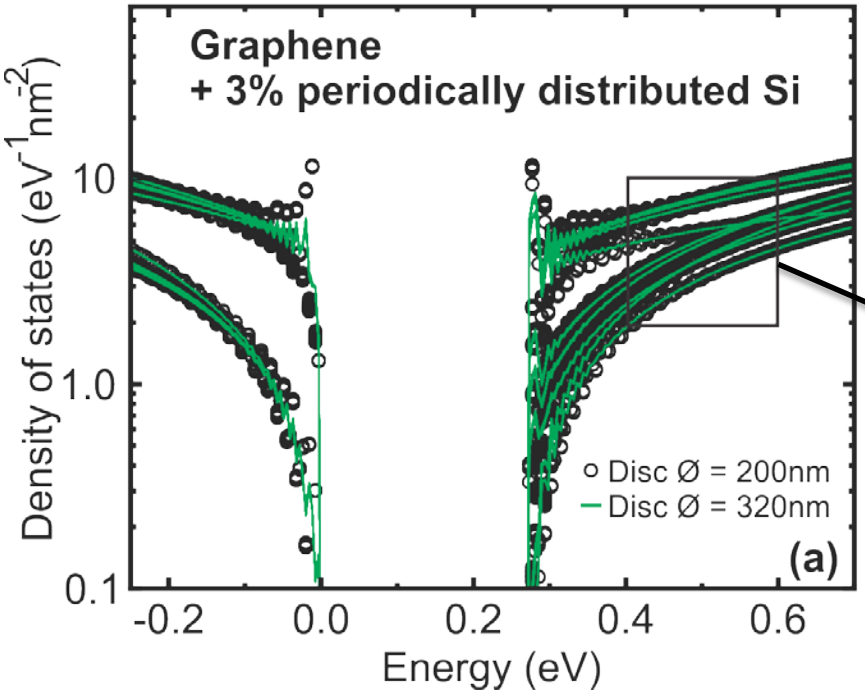


Continuum and atomistic models:

- ✓ Reproduction of analytical DOS
- ✓ Quality around band edges/Dirac point convergence dependent…
  …similar to irregular lead model

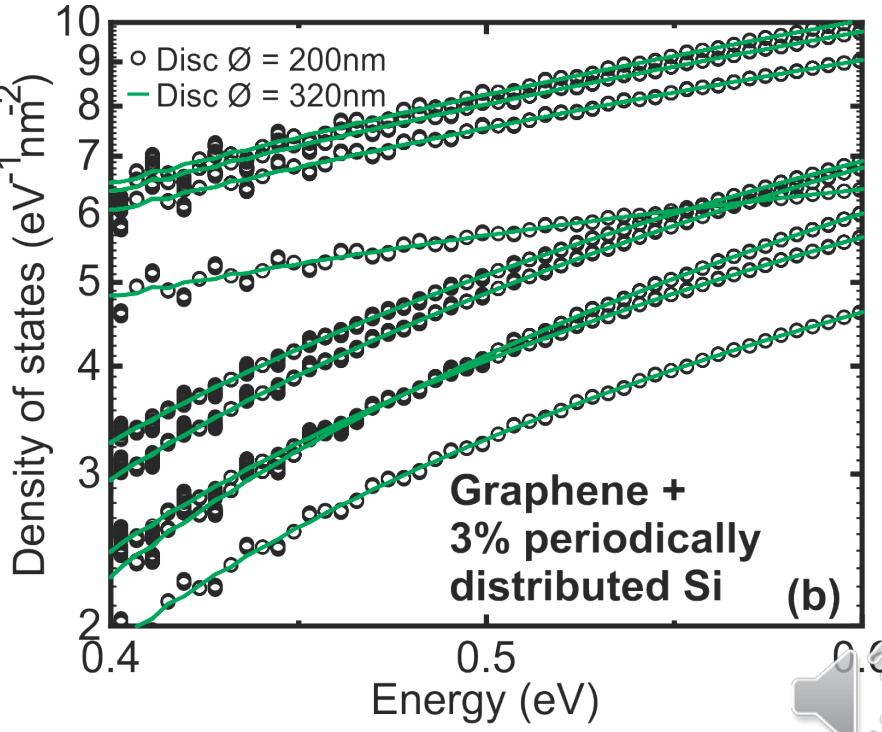
ROBIN paper:
ACS Materials Lett. 2020, 2, 247

Results: graphene + periodic Si doping



Graphene + 3% periodic Si substitution:

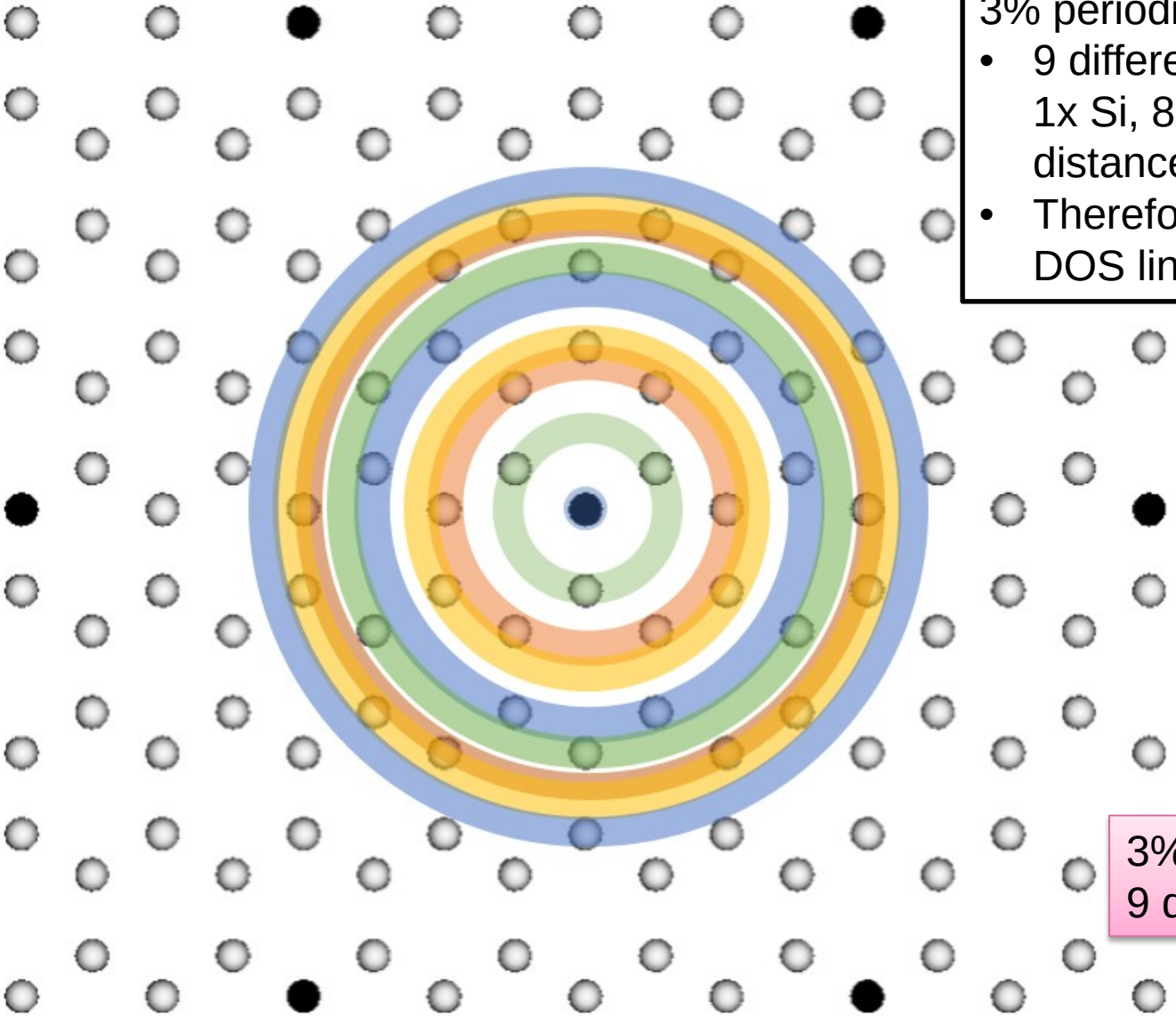
- Reported to have 247meV band gap (S. J. Zhang et al. Nanoscale, 8 (2015) 226)
- Reproduced with ROBIN



This ROBIN simulation:

- 282 atoms considered in central device region (~3 million atoms in total)
- 282 DOS-lines shown fall into 9 different groups of lines

Results: graphene + periodic Si doping

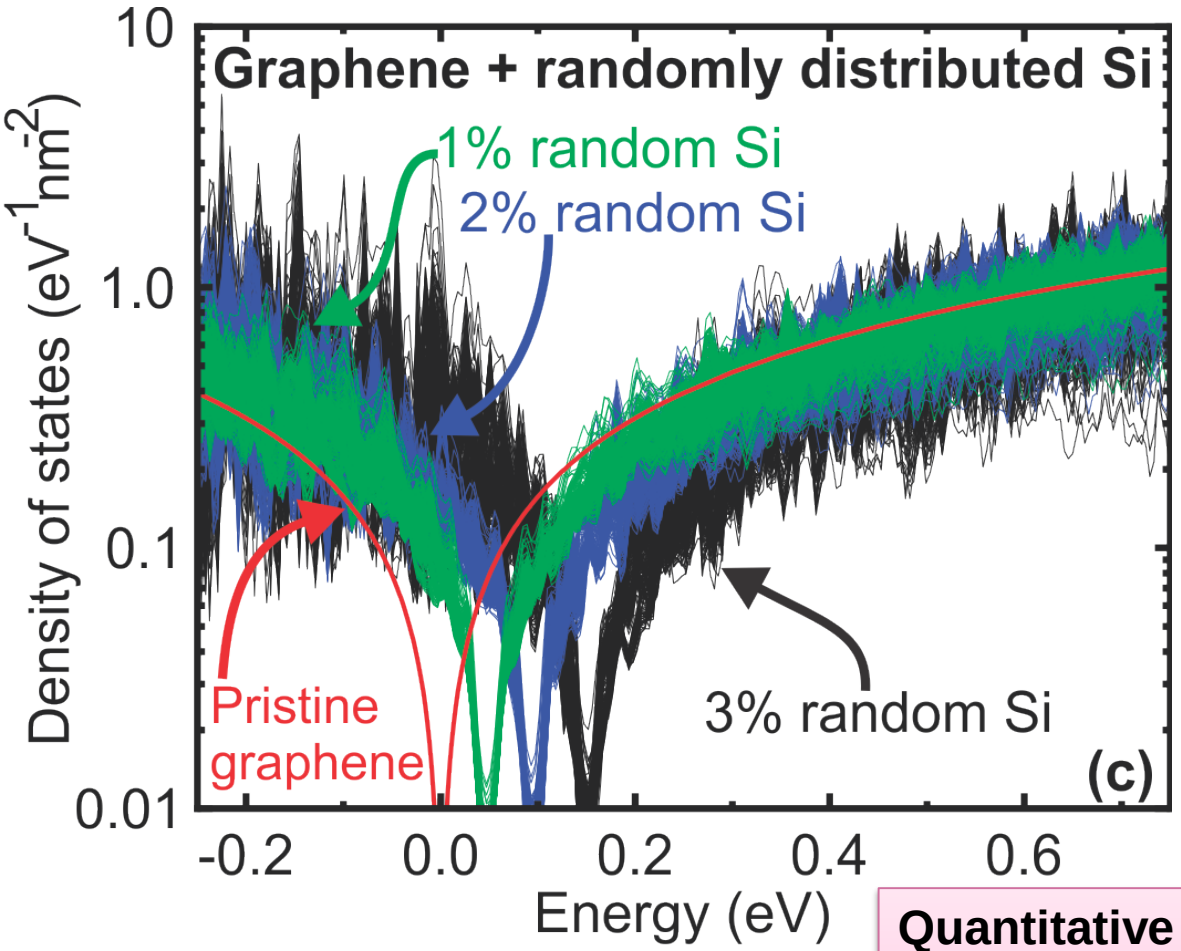


3% periodic Si substitution:

- 9 different atom types:
  1x Si, 8x C in 8 different distances to Si
- Therefore, 9 different atomic DOS lines

3% periodic Si graphene:
9 different atom types

Results: graphene + random Si doping



3% random Si substitution:
- 282 atoms explicitly solved
- 282 different DOS results

No band gap,
but linear shift of Dirac cone

3% random Si graphene:
All atoms with different chemical surrounding ("noise")

Quantitative agreement with experiment:
- DOS shift with ROBIN:
  0.147eV for 3% Si (~47meV per Si-%)
- Work function shift in experiment:
  0.13eV for 2.7 - 4.5% Si

(S. J. Zhang et al. Nanoscale, 8 (2015) 226)

Avoid periodicity assumption for nonperiodic systems

Conclusion

This talk:

- ✓ Introduction to the recursive open boundary and interfaces (ROBIN) method
- ✓ Quantitative agreement with experiments
- ✓ ROBIN applied on Si substitutional doping of graphene:
  - ✓ **Reproduced bandgap opening of periodic Si doping**
  - ➢ **No band gap, but linear shift of Dirac cone for random Si doping**

Take home message:

- Open boundaries are applicable on any material and interface situation
- Periodic boundaries enhance small perturbations to systematic changes of material properties
- **Avoid periodicity assumption for non-periodic systems!**

Marda-alliance.org: working to connect materials data infrastructure

ROBIN paper:
ACS Materials Lett. 2, 247 (2020)

Thank you!