Usage of HPC in simulations of charge transport in semiconducting materials

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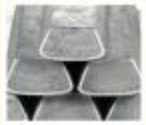
EuroCC4SEE Workshop
Belgrade, 20 May 2025

Motivation

- Ohm's law

$$R = \rho L / S$$

- A lot of physics behind ρ

Проводници		
Материјали		
Бакар	Алуминијум	Гвожђе
		
Специфична отпорност [Ωm]		
$1{,}7 \cdot 10^{-8}$	$2{,}8 \cdot 10^{-8}$	$9{,}7 \cdot 10^{-8}$

Полупроводници		
Материјали		
Графит	Германијум	Силицијум
		
Специфична отпорност [Ωm]		
$3 \cdot 10^{-5}$ до $6 \cdot 10^{-4}$	0,001 до 0,5	0,1 до 60

Изолатори		
Материјали		
Дрво	Стакло	Пластика
		
Специфична отпорност [Ωm]		
10^{8} до 10^{14}	10^{10} до 10^{14}	10^{13}

Materials by design

TOPICAL REVIEW • OPEN ACCESS

The 2019 materials by design roadmap

Kirstin Alberi[1], Marco Buongiorno Nardelli[2], Andriy Zakutayev[1], Lubos Mitas[3], Stefano Curtarolo[4,5], Anubhav Jain[6], Marco Fornari[7], Nicola Marzari[8], Ichiro Takeuchi[9], Martin L Green[10] + Show full author list

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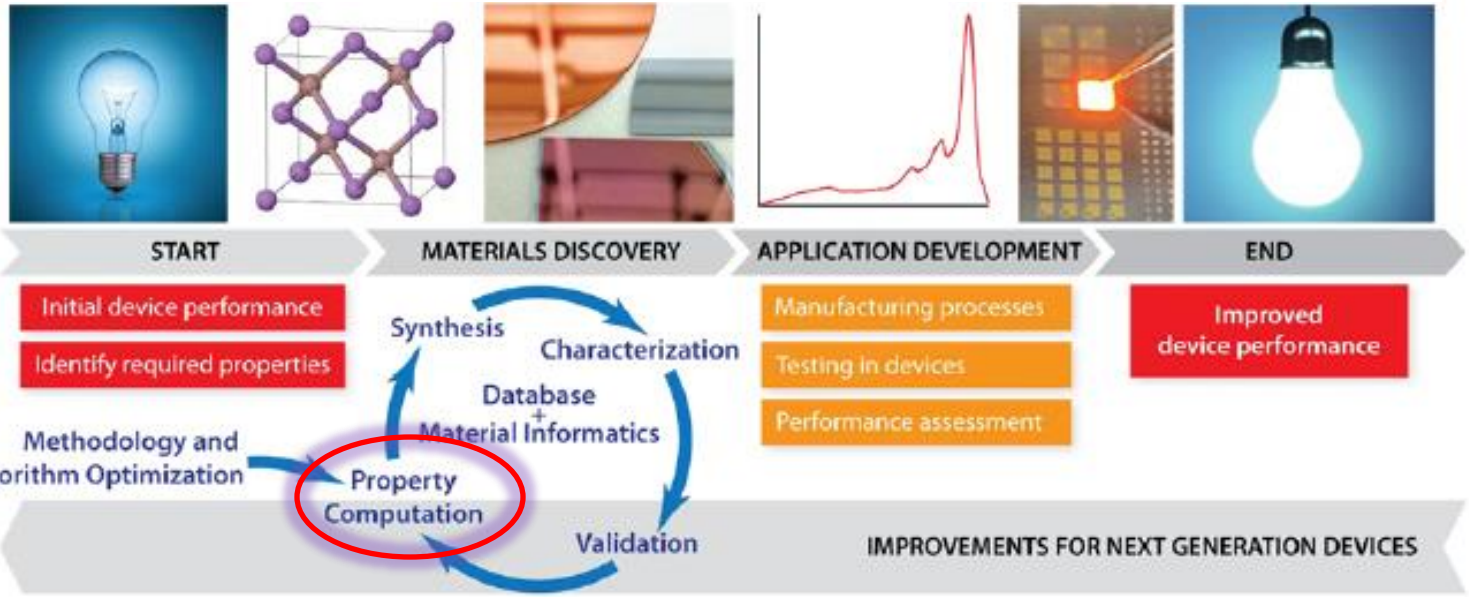
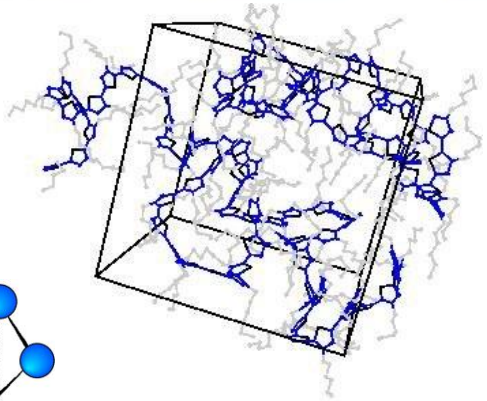
Citation Kirstin Alberi *et al* 2019 *J. Phys. D: Appl. Phys.* **52** 013001

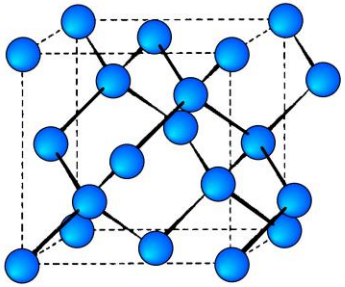


Different charge transport mechanisms

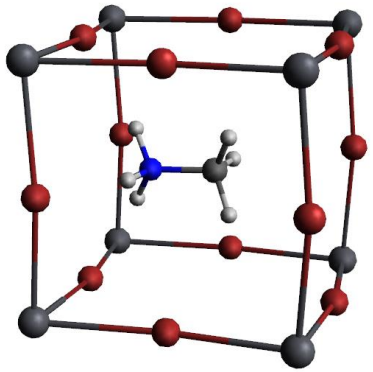
- Hopping transport (e.g. disordered organic semiconductors relevant for organic cells)



- Band transport (crystalline semiconductors, e.g. silicon)



- Intermediate transport (emerging semiconductors, e.g. halide perovskites)

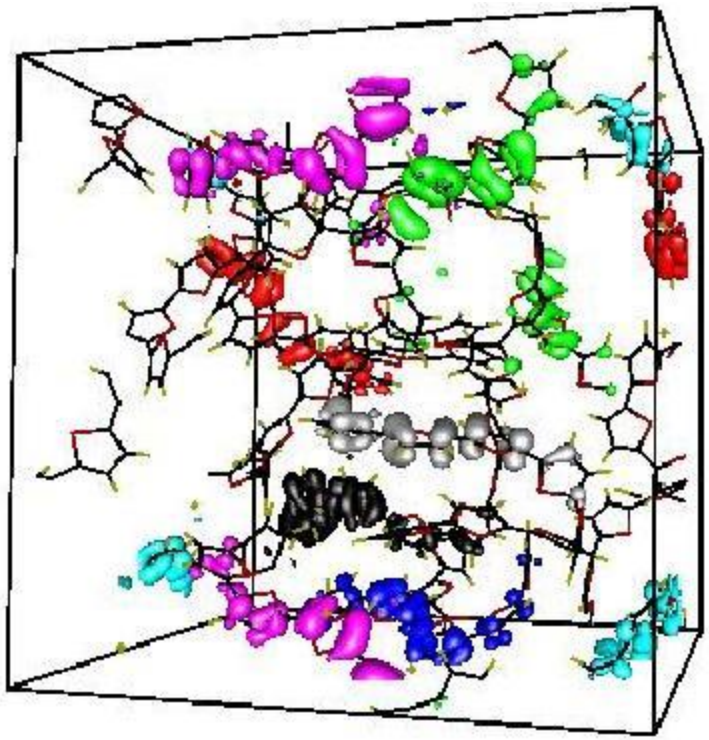


- Each of these mechanism calls for a different simulation / modeling approach

Hopping transport

Amorphous materials

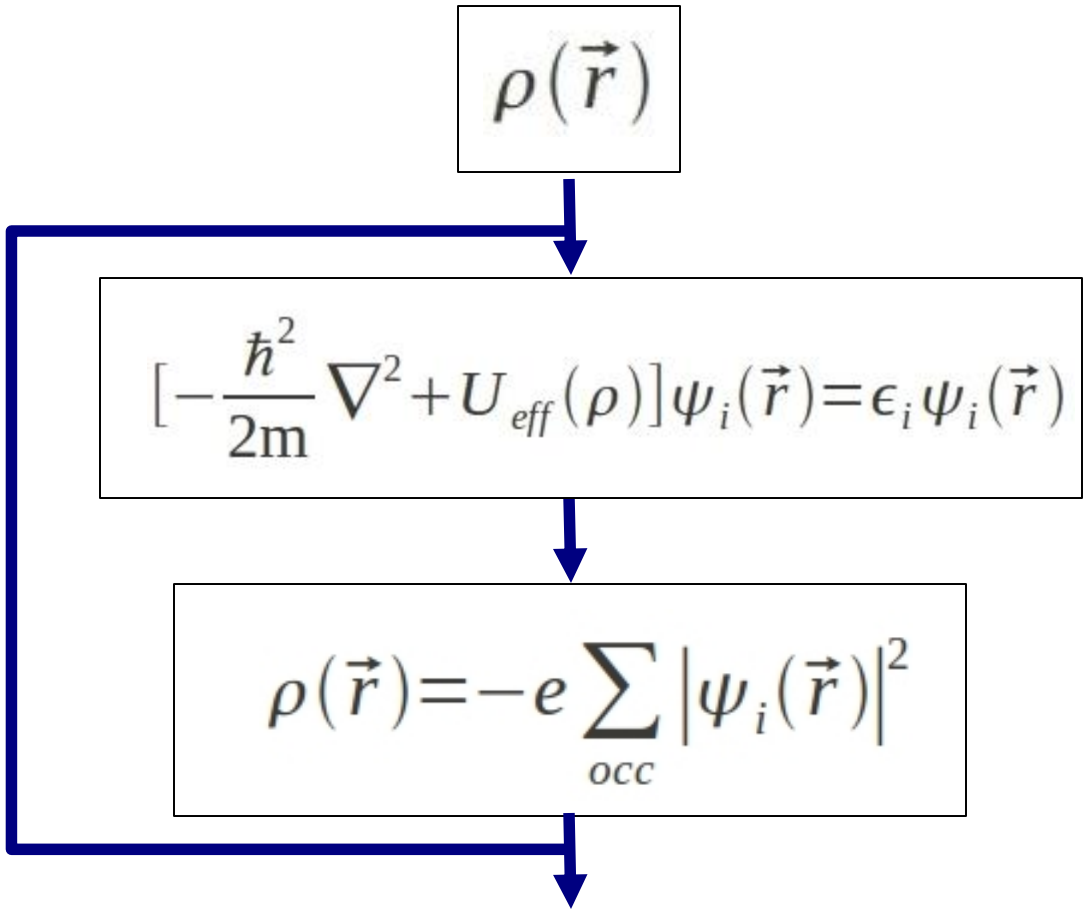
- Localized electrons, move slowly.
- Large system calculations needed to extract transport properties.

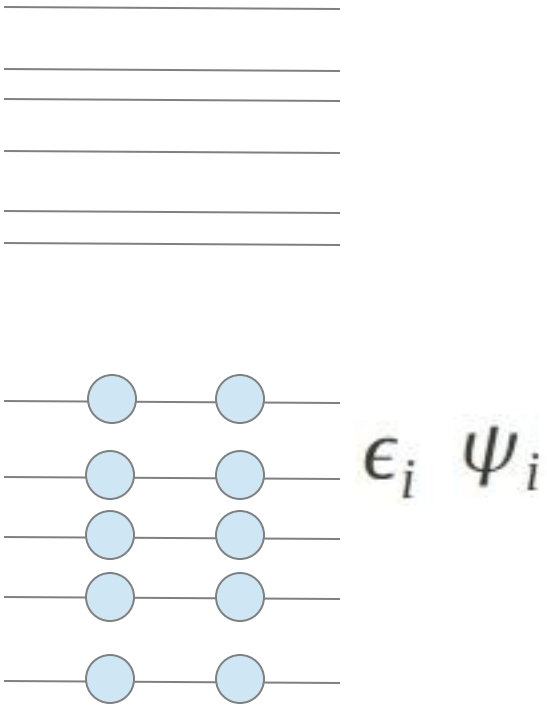


N. Vukmirović and L. W. Wang, J. Phys. Chem. B 113, 409 (2009).

Density Functional Theory

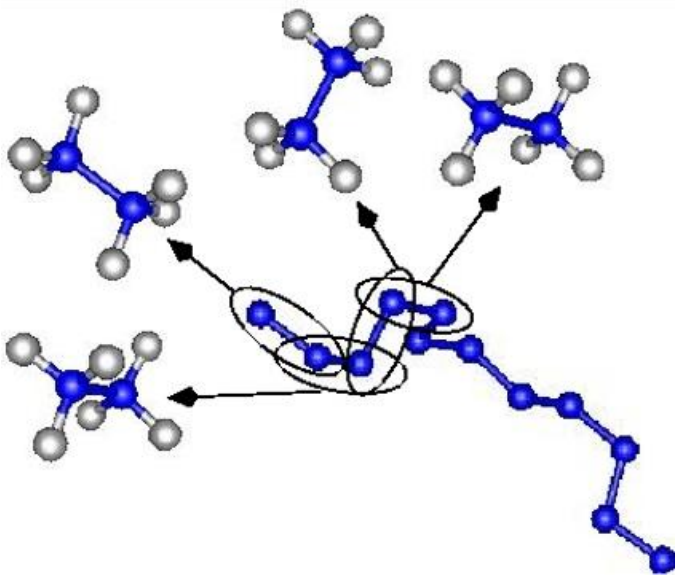
- Limitation to ~100-1000 atom systems.



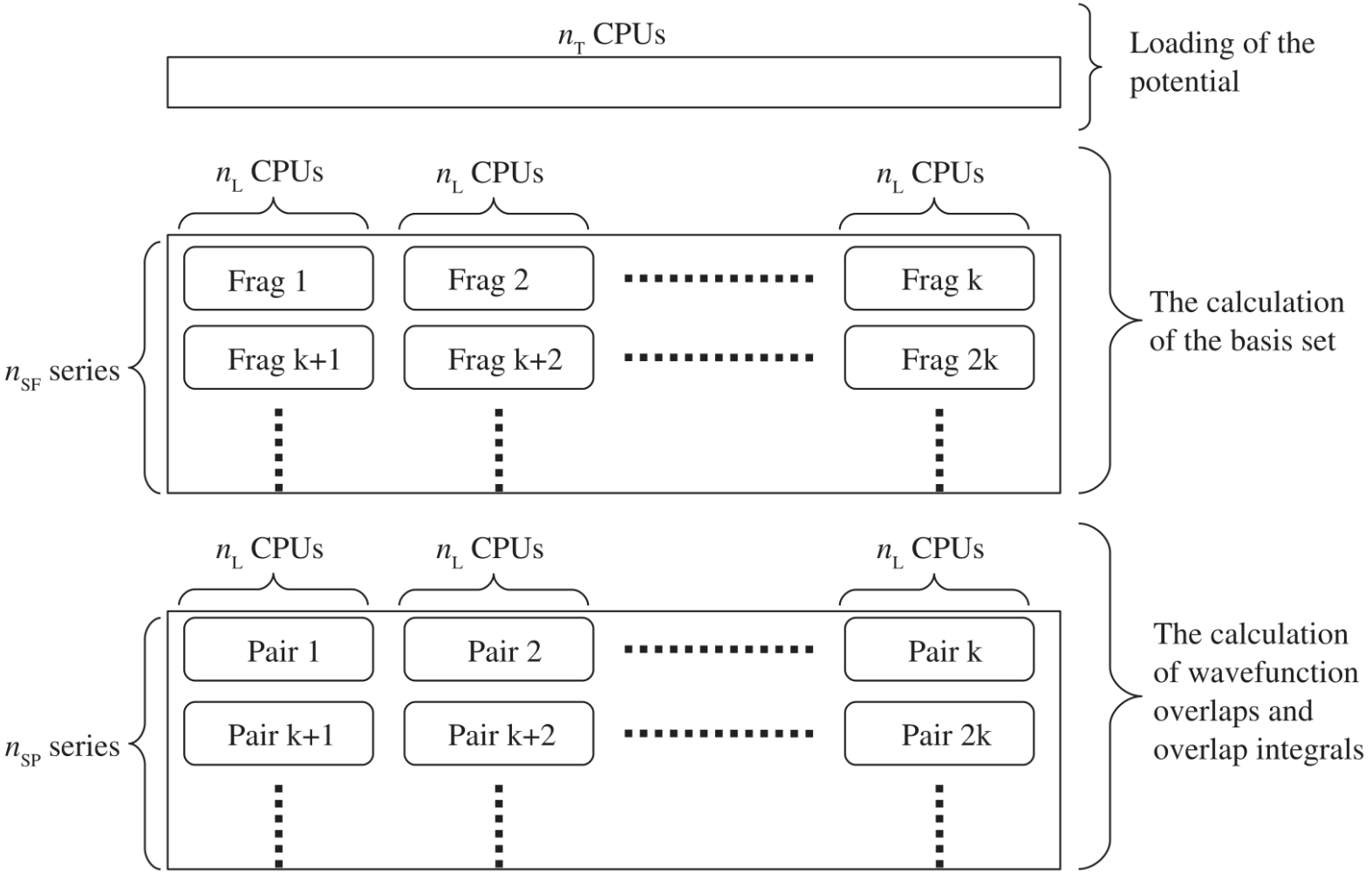


Overlapping fragments methods

- Division of the system into (mutually overlapping) fragments.
  - HOMOs (LUMOs) of the whole system linear combination of HOMOs (LUMOs) of fragments.
- Procedure:
  - Calculation of fragment orbitals ϕ_i
  - Calculation of $S_{ij}=\langle\phi_i|\phi_j\rangle$ and $H_{ij}=\langle\phi_i|H|\phi_j\rangle$
  - Solution of the generalized eigenvalue problem $\det(H_{ij}-ES_{ij})=0$

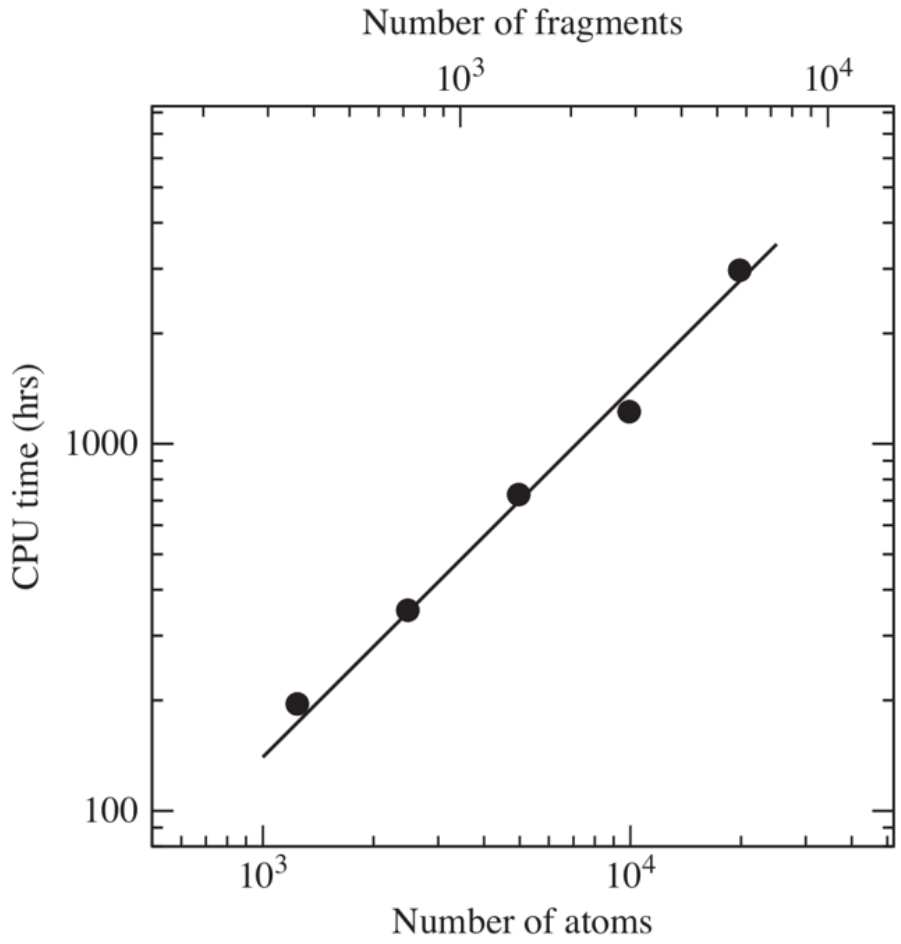


Overlapping fragments methods - implementation



N. Vukmirović and L.-W. Wang, J. Chem. Phys 134, 094119 (2011).

Overlapping fragments methods - scaling

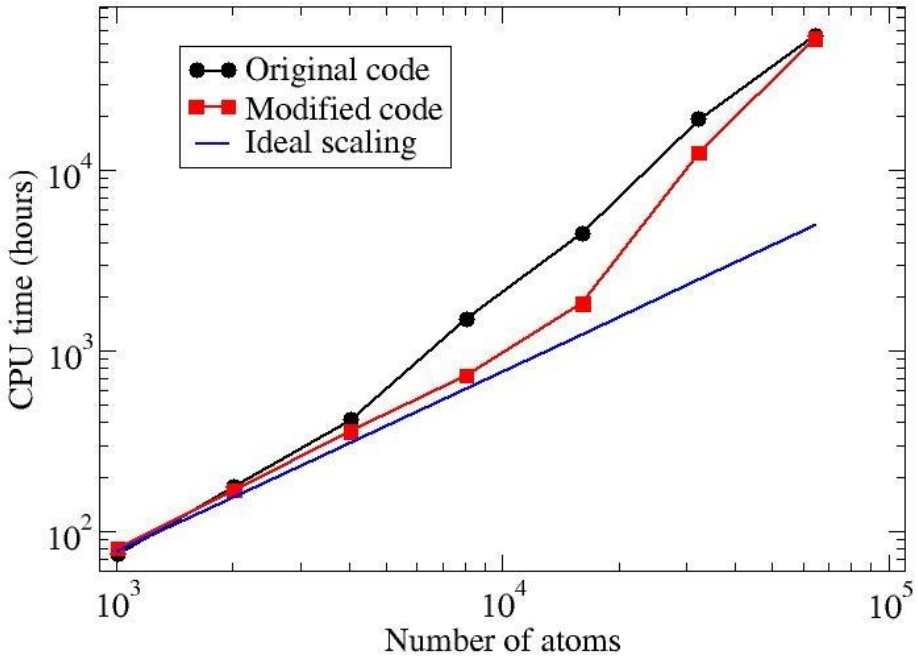


- Amorphous alkane system.
- Runs with typically 5000 CPU c ores.
- One orbital per fragment.

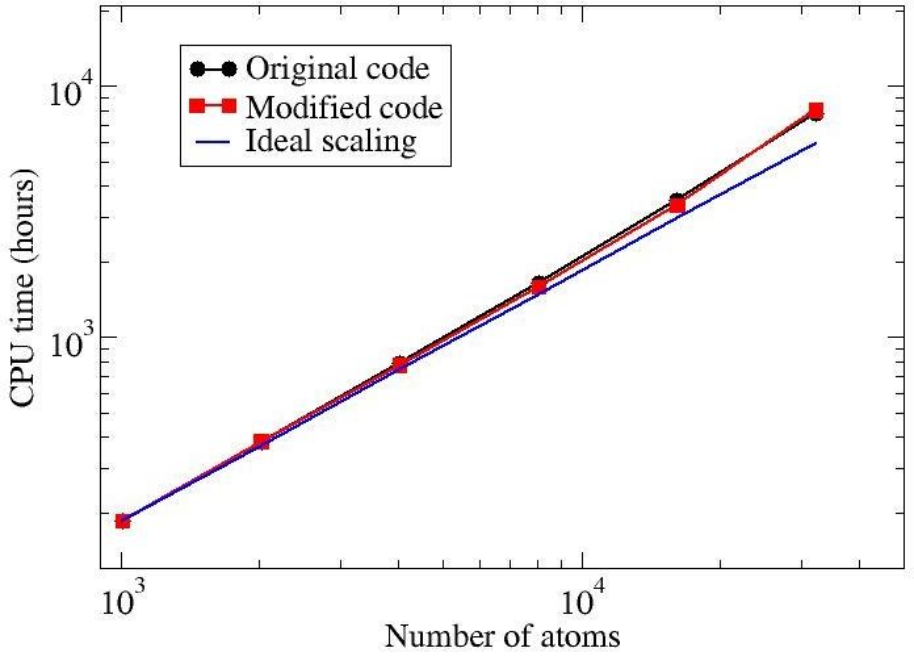
N. Vukmirović and L.-W. Wang, J. Chem. Phys 134, 094119 (2011).

Overlapping fragments methods – role of communication

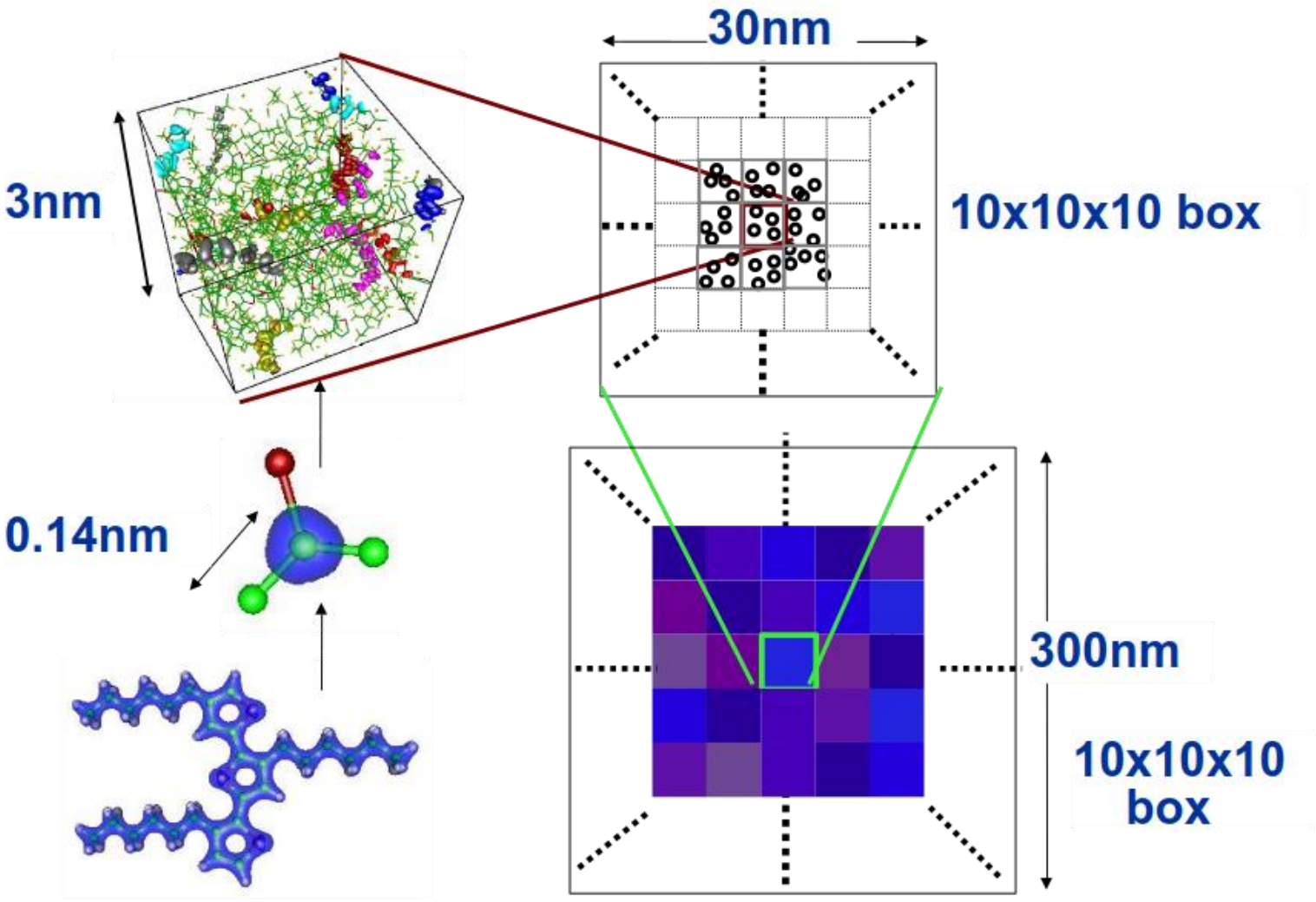
- Weak scaling test – Curie (Paris)
- Infiniband network.
- Tests using from 256 to 16384 cores.



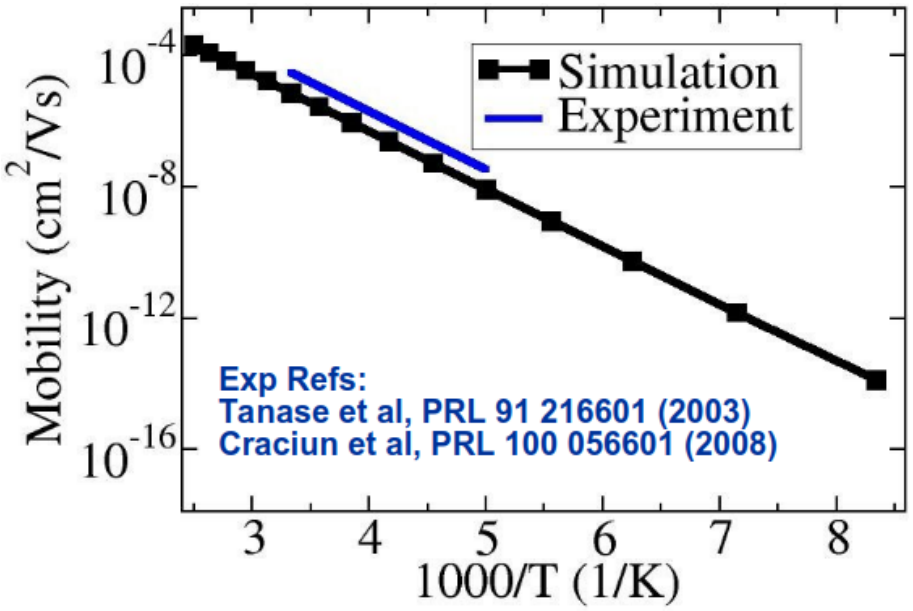
- Weak scaling test – HLRS (Stuttgart)
- Cray XE6 within Cray Gemini network.
- Tests using from 256 to 16384 cores.

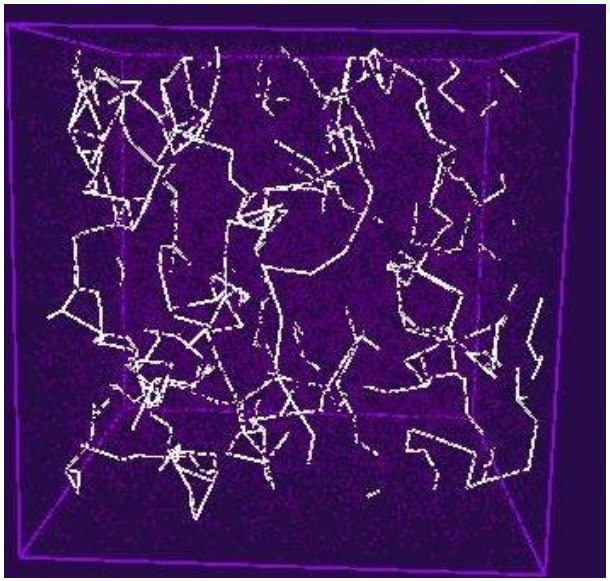


Multiscale method for carrier transport



Temperature dependence of mobility

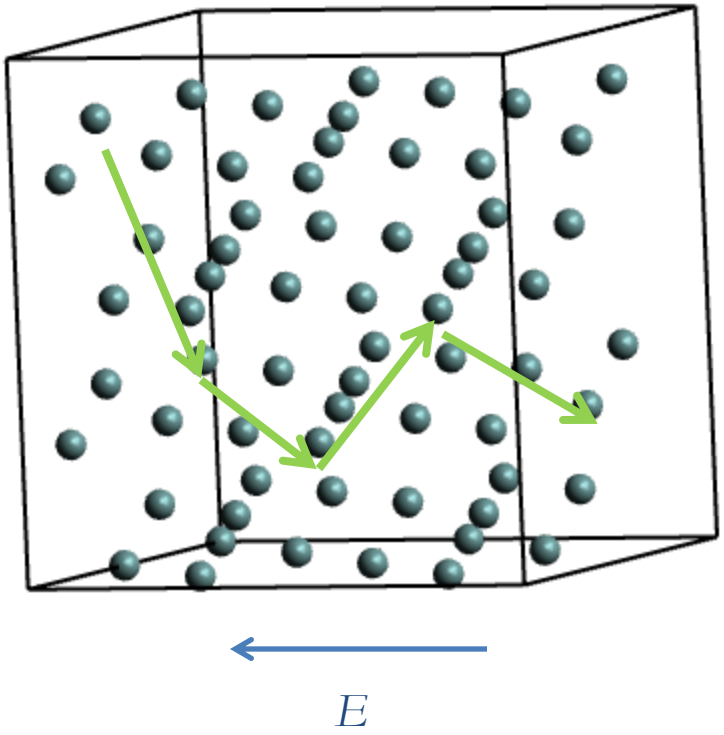


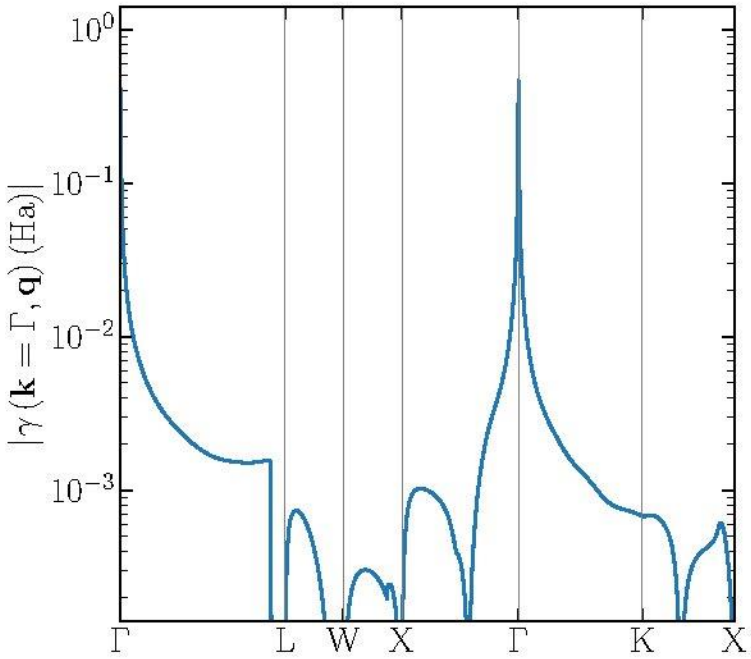


Band transport

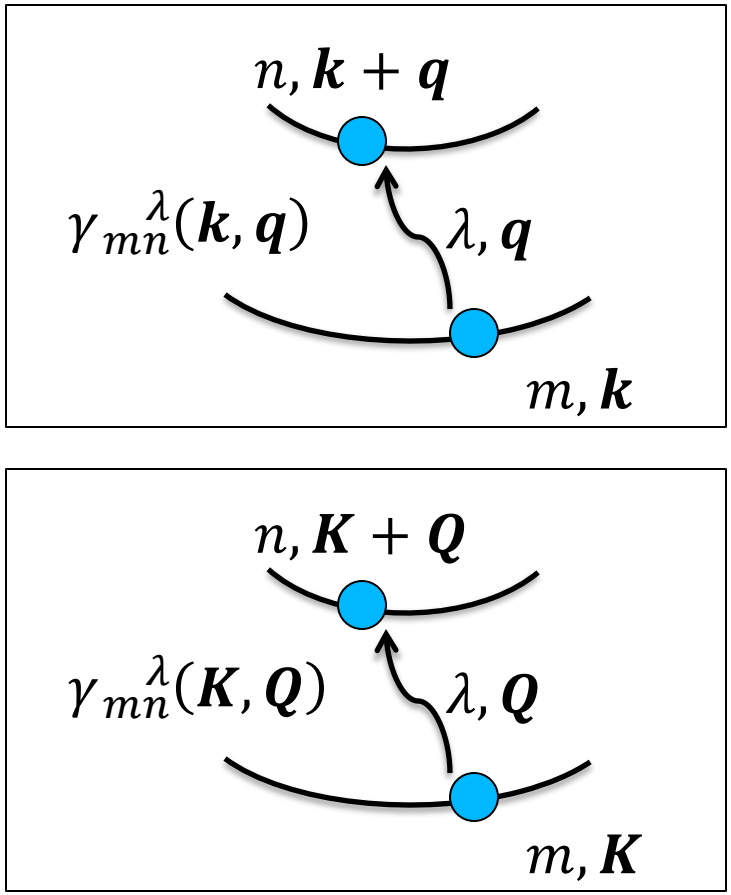
Crystalline semiconductors

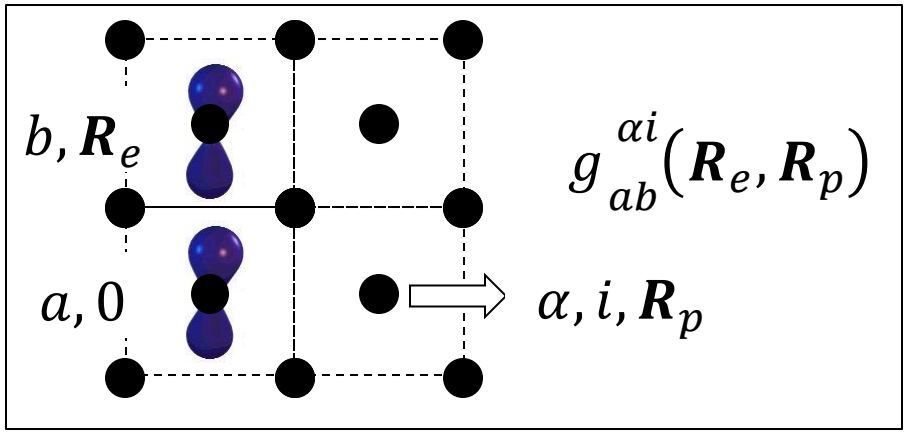
- Delocalized electron, moves easily.
- Lattice vibrations slow it down and limit its mobility.
- One needs to calculate electron-phonon coupling constants.
- These are strongly varying with q, so dense grids are needed.





Electron-phonon coupling constants interpolation



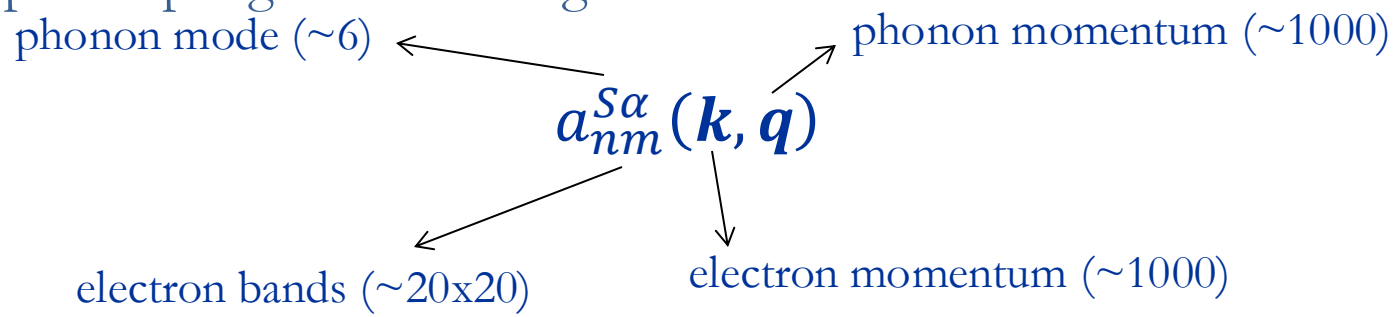


F. Giustino et al, PRB 76, 165108 (2007).

Need for large data structures

- Electronic states, phonon modes and e-ph coupling constants from ABINIT code. This is used as input to our parallel code.

- E-ph coupling elements in germanium:

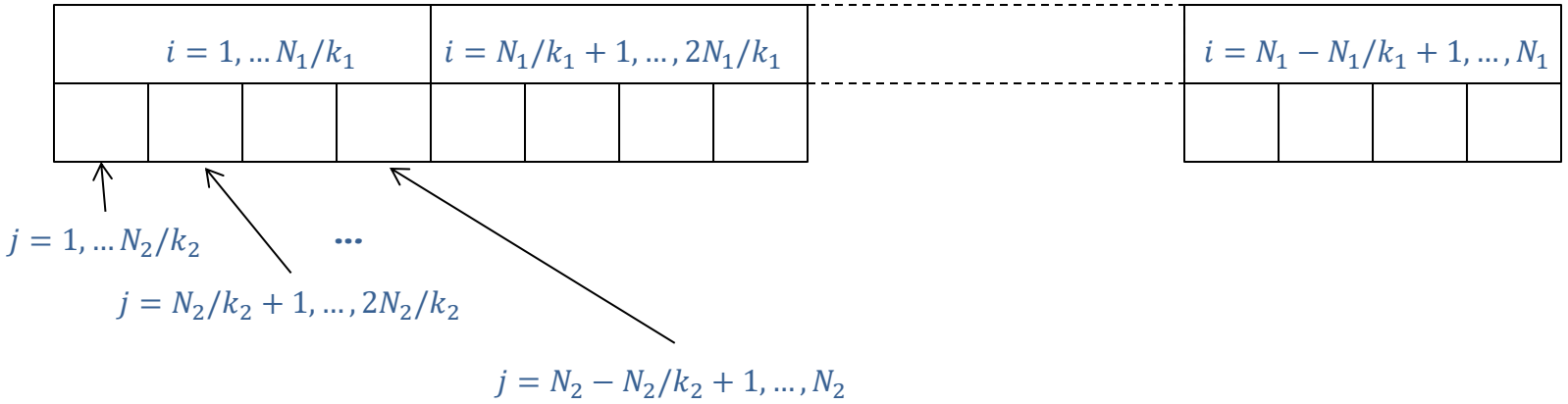


- One needs ~40GB to store e-ph coupling elements on the course grid for germanium (2 atoms per unit cell)
- Distribution of these elements in memory of different nodes therefore necessary.

Data distribution

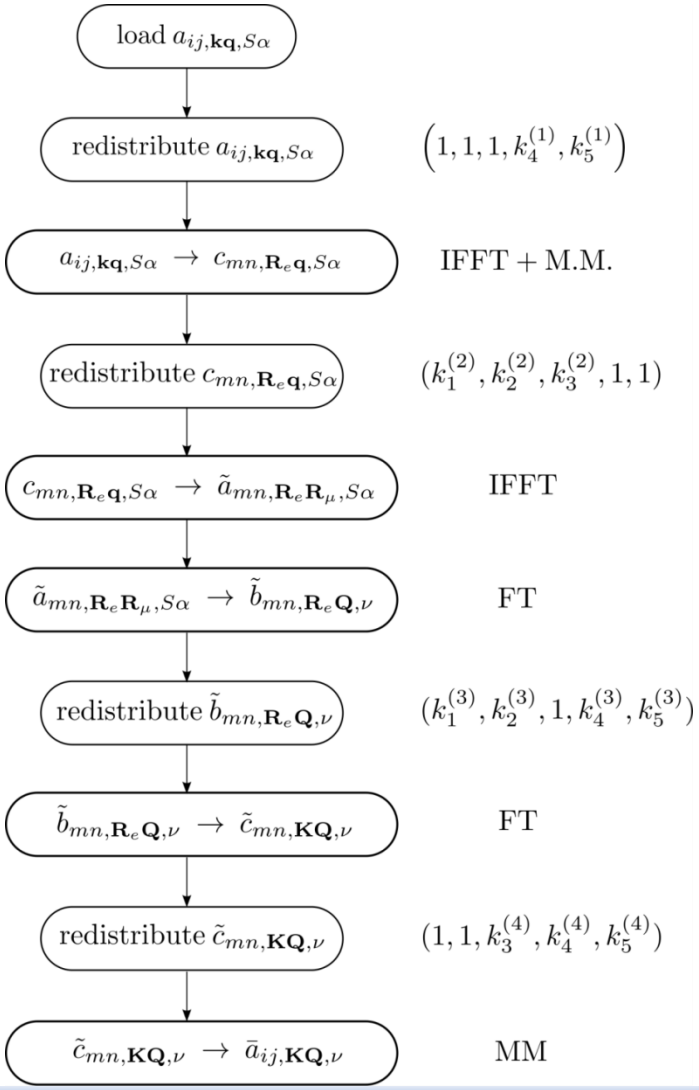
- Array a_i with N elements, machine with k cores.

- Array a_{ij} with $N_1 N_2$ elements, machine with $k_1 k_2$ cores.



- Same approach to distribute the array a_{ijklm} with $N_1 N_2 N_3 N_4 N_5$ elements, on a machine with $k_1 k_2 k_3 k_4 k_5$ cores.

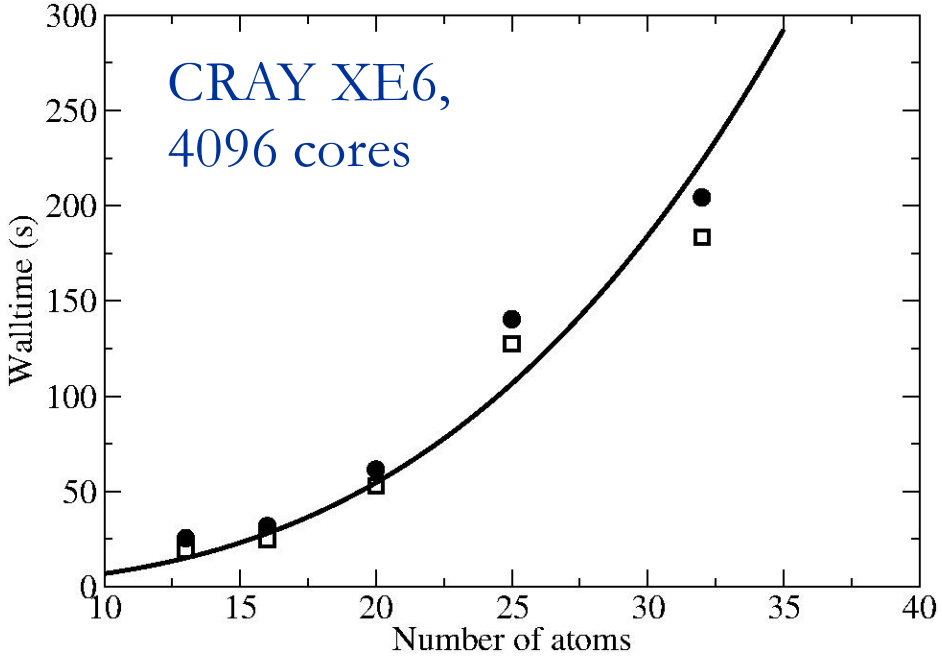
Parallelization



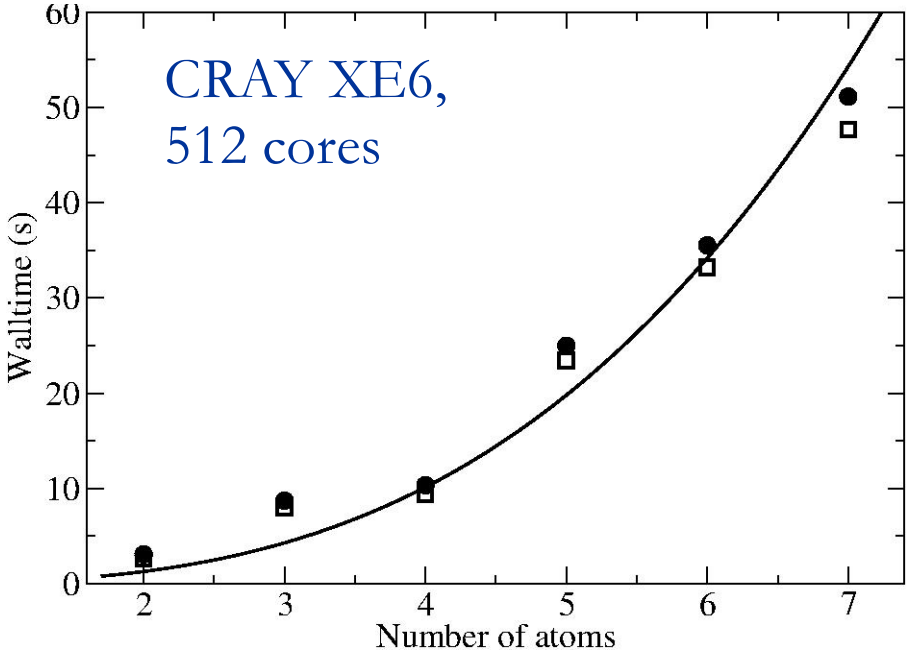
- The strategy is to have all array elements involved in the computation of FT or MM on the same core.
- To achieve this, we adopt redistribute – compute – redistribute approach.
- mpi_alltoallv used for redistribution.

Scaling with system size

- Parameters nb=2nat, nk=nq=216, nK=1, nQ=200



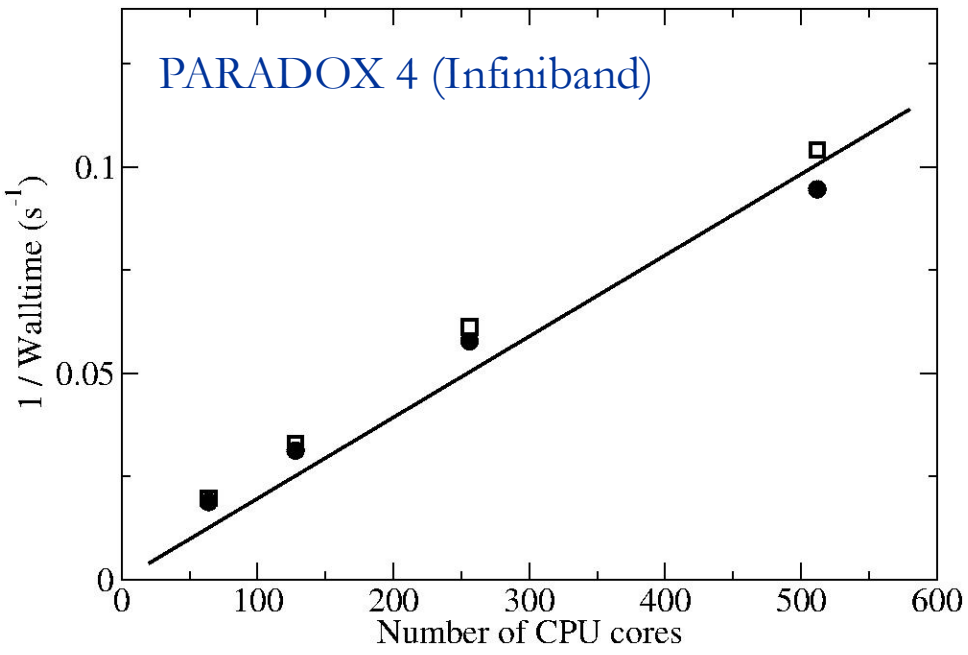
- Parameters nb=2nat, nk=nq=216, nK=1000, nQ=200

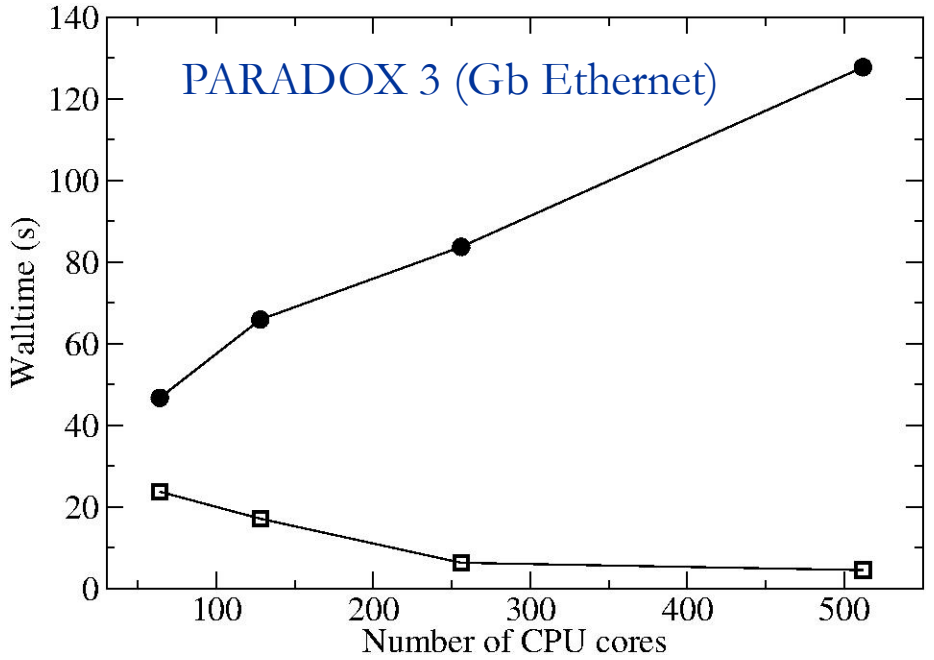


- $\sim n_{at}^3$ scaling with system size.
- communication negligible.

Scaling with number of CPU cores

- Parameters nat=4, nb=2nat, nk=nq=216, nK=1000, nQ=200

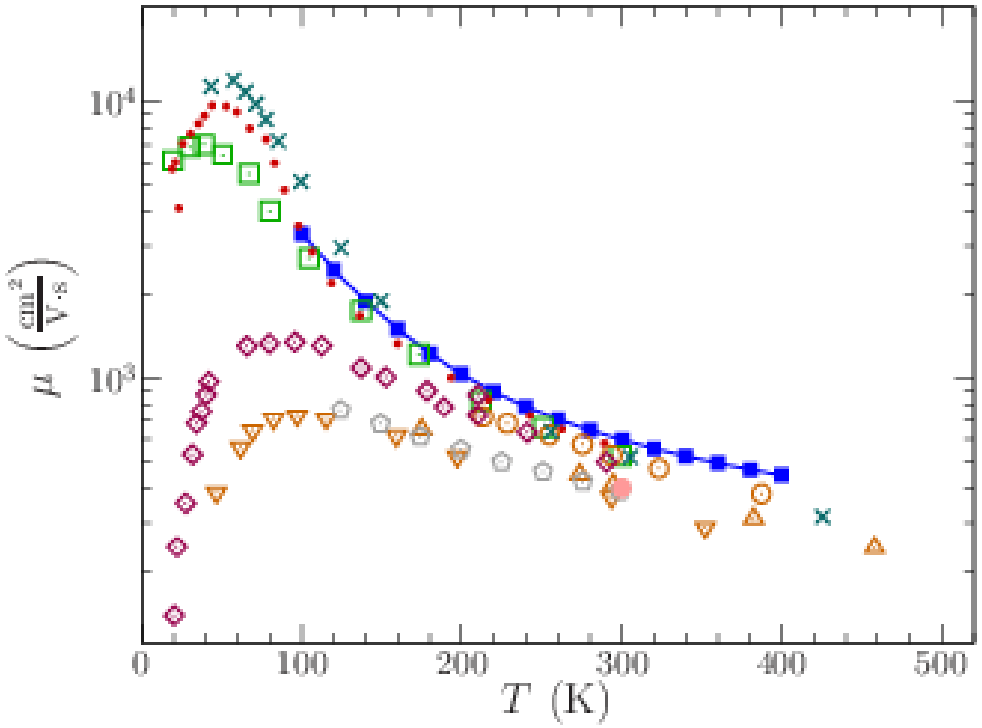




- linear scaling with system size on machine with infiniband network, poor scaling with Gb Ethernet.

Temperature dependence of mobility

- Mobility decreases with an increase in temperature.
- Example – ZnSe material, comparison with experiment.

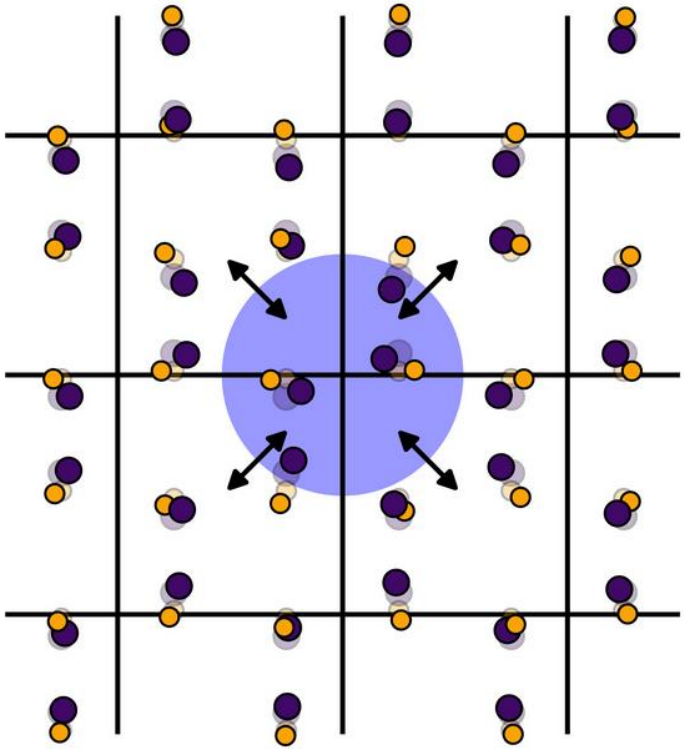


N. Vukmirović, Phys. Rev. B 104, 085203 (2021).

Intermediate regimes

Polarons in materials

- Polaron formation – mixed electron-phonon states.



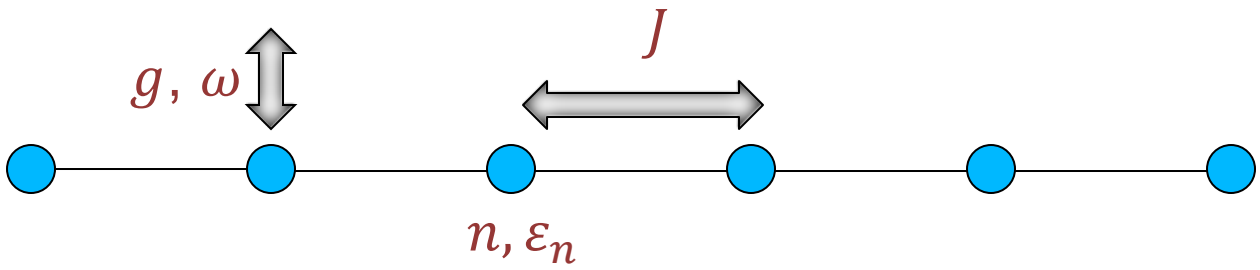
PNAS 119(3):e2113967119 (2022)

- On going project: Polaron Mobility in Model Systems and Real Materials (PolMoReMa – 5468) (2024-2026).
- http://polmorema.ipb.ac.rs

Science Fund of the Republic of Serbia

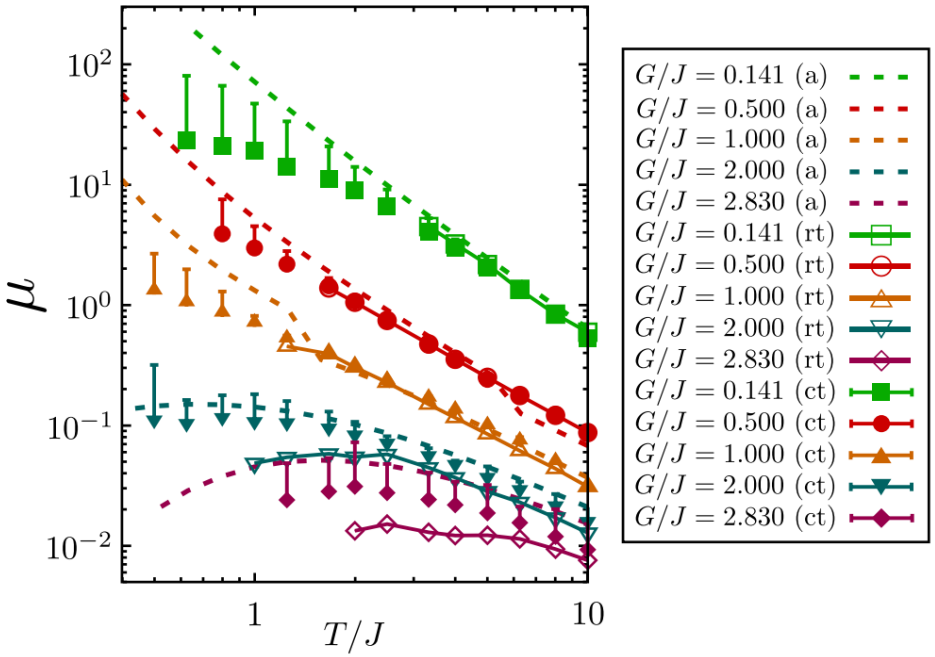
Numerically exact approaches

- Direct evaluation of current-current correlation function starting from the Hamiltonian of the system.
- Example: Holstein model
- Direct diagonalization - feasible up to only ~3 sites
- Other approaches:
  - Path-integral Quantum Monte Carlo
  - Hierarchical equations of motion
  - Time dependent density matrix renormalization group
  - ...



Quantum Monte Carlo for mobility of Holstein polaron

- Evaluation of integrals involving Gaussian functions using Monte Carlo sampling
- Essentially single processor code
- Straightforward parallelization: to obtain Monte Carlo statistics



S. Miladić, N. Vukmirović, Phys. Rev. B 107, 184315 (2023).

Summary

- Different physics requires different algorithms even for the same problem of charge transport in semiconducting material.
- HPC is indispensable in each case.
- New physical insights:
  - HPC
  - physical intuition
  - algorithm and code development

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