Quantum Electron Dynamics Simulation of Materials on High-Performance Computers

André Schleife, Department of Materials Science and Engineering, UIUC

Erik Draeger, Lawrence Livermore National Laboratory

Victor Anisimov, National Centre for Supercomputing Applications

Alfredo Correa, Lawrence Livermore National Laboratory

Yosuke Kanai, Department of Chemistry, Univ. of North Carolina, Chapel Hill

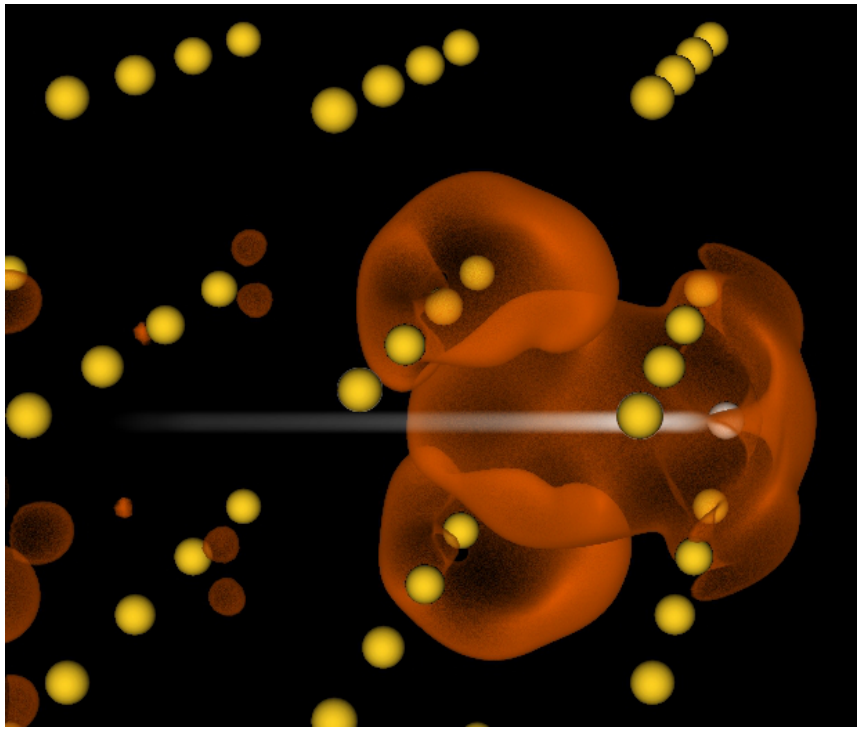


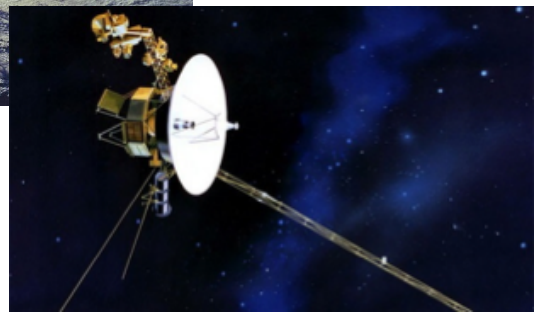
Image credit: David Semerano

Blue Waters Symposium, May 2014

Beyond Born-Oppenheimer: Non-adiabatic dynamics

Examples:

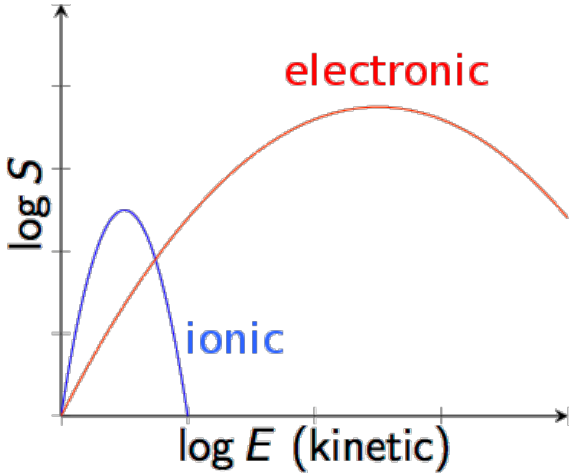
- Solar cells on satellites
- Nuclear reactors
- Magnetic confinement/inertial confinement fusion
- Surface adsorption



→ Excited-electron dynamics

Radiation damage:

- Slow projectiles: ionic stopping dominant
- Fast projectiles: interaction with electronic system important stopping mechanism



Goal:

- Parameter-free framework to model stopping

Andre Schleife @ MatSE @ UIUC · Email: schleife@illinois.edu · Web: http://schleife.matse.illinois.edu · @aschleife

Beyond Born-Oppenheimer: Background

beyond Born-Oppenheimer is necessary:

Time-dependent Kohn-Sham equations:

$$\mathrm{i}\hbar\frac{\partial\varphi_i(t)}{\partial t} = \hat{H}[n]\varphi_i(t) = \left[\hat{T} + v_{\mathrm{ext}}(\mathbf{r},t) + v_{\mathrm{H}}[n](\mathbf{r}) + v_{\mathrm{XC}}[n](\mathbf{r})\right]\varphi_i(t)$$

- Popular for excitation spectra: linear-response TDDFT
- Here: Integrating the equations of motion: “Real-time” TDDFT

- Challenging: Highly parallel implementation
- Runge-Kutta explicit integration scheme

- Compute forces at each time step and update positions of the atoms

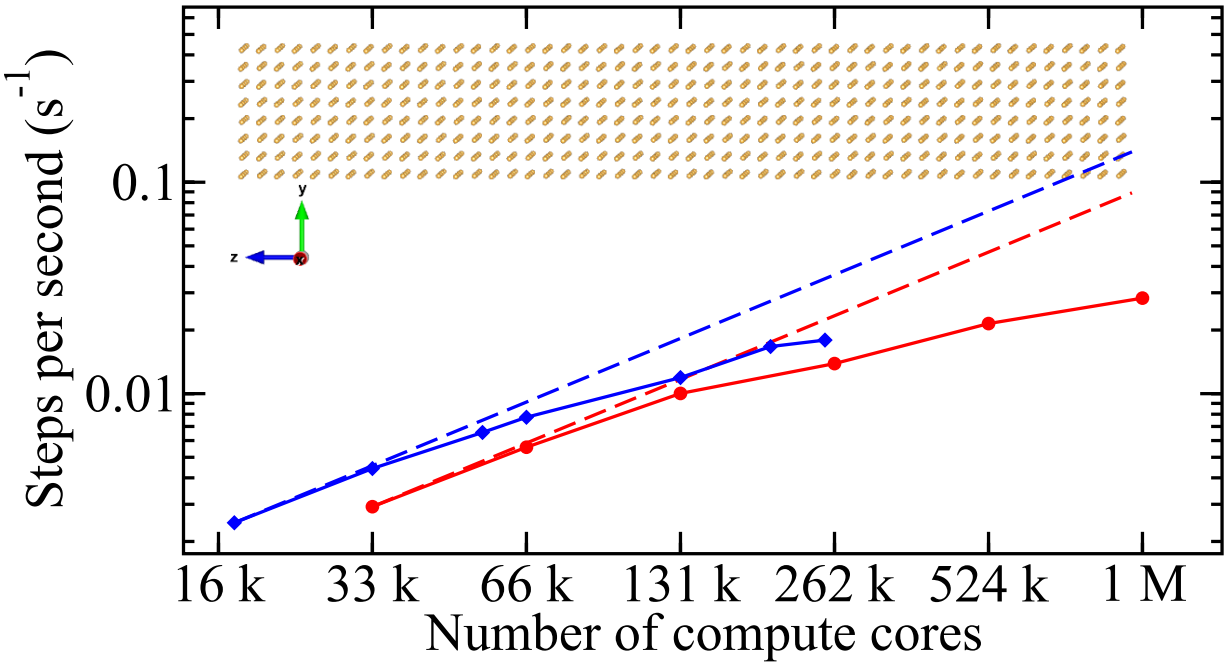
Ehrenfest molecular dynamics

A. Schleife *et al.*; J. Chem. Phys. **137**, 22A546 (2012)

Beyond Born-Oppenheimer: Strong Scaling

Running on modern super computers:

- Solid red: Sequoia (20 PFlop/s IBM Blue Gene/Q @ LLNL)
- Solid blue: Blue Waters (13.3 PFlop/s Cray XE/XK @ UIUC)



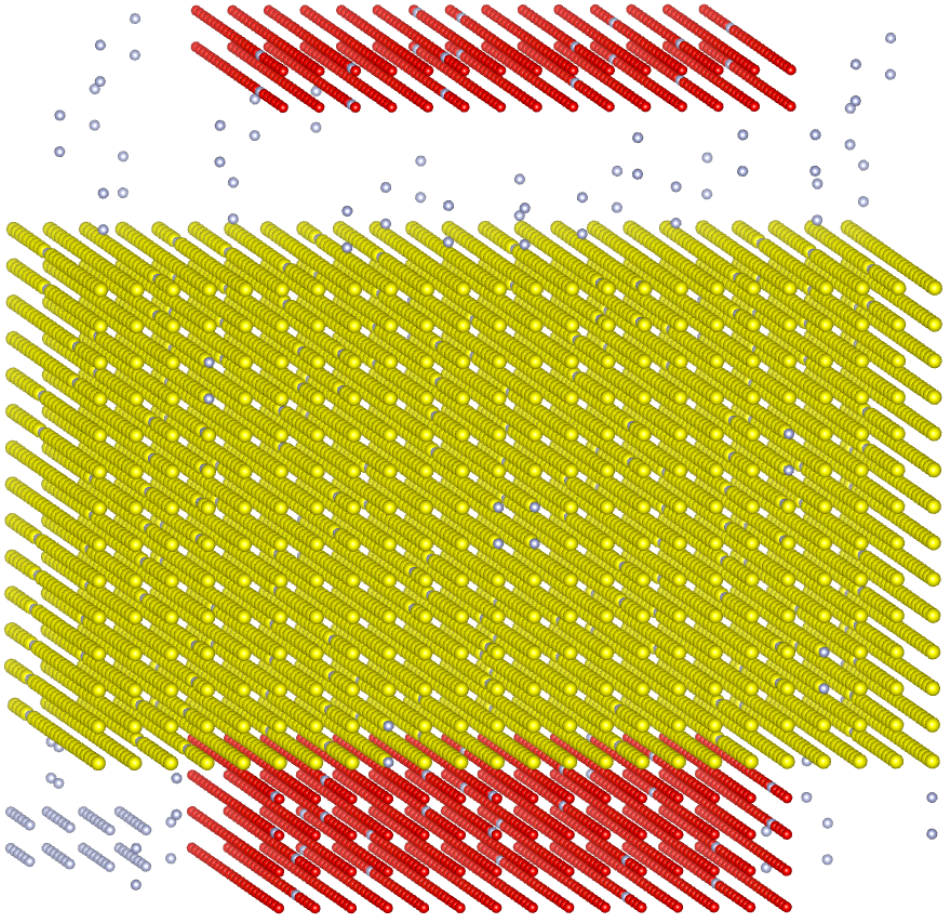
Hydrogen in Gold

- 27,200 electrons
- 1,500 atoms

Explicit integrators ideal choice for highly parallel implementation

Explicit Integration: Strong Scaling

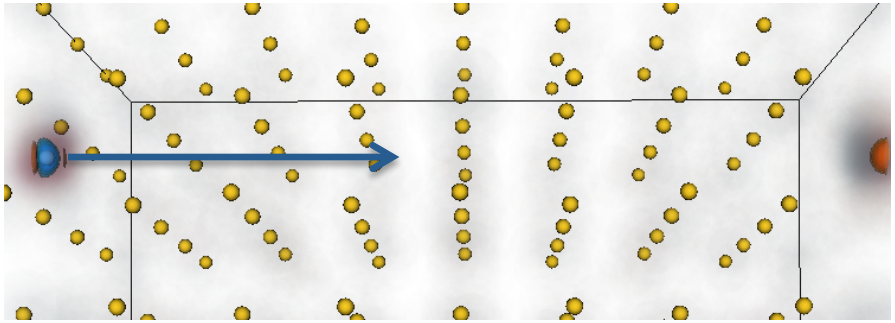
Layout on Blue Waters

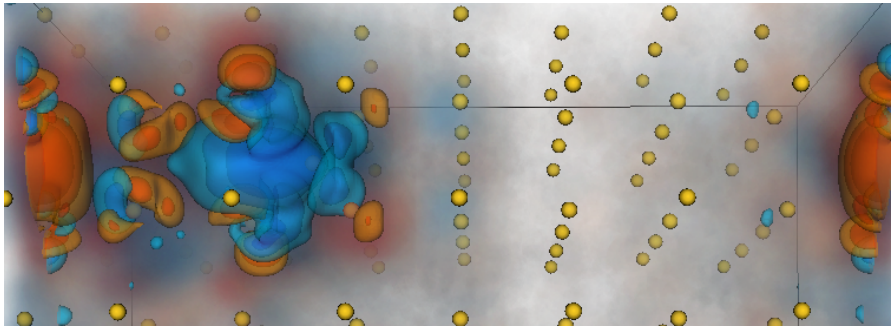


- Empty spaces: unused nodes
- Red: XK (GPU) section
- Yellow: our job
- Blue: service nodes

Beyond Born-Oppenheimer: Hydrogen in Gold

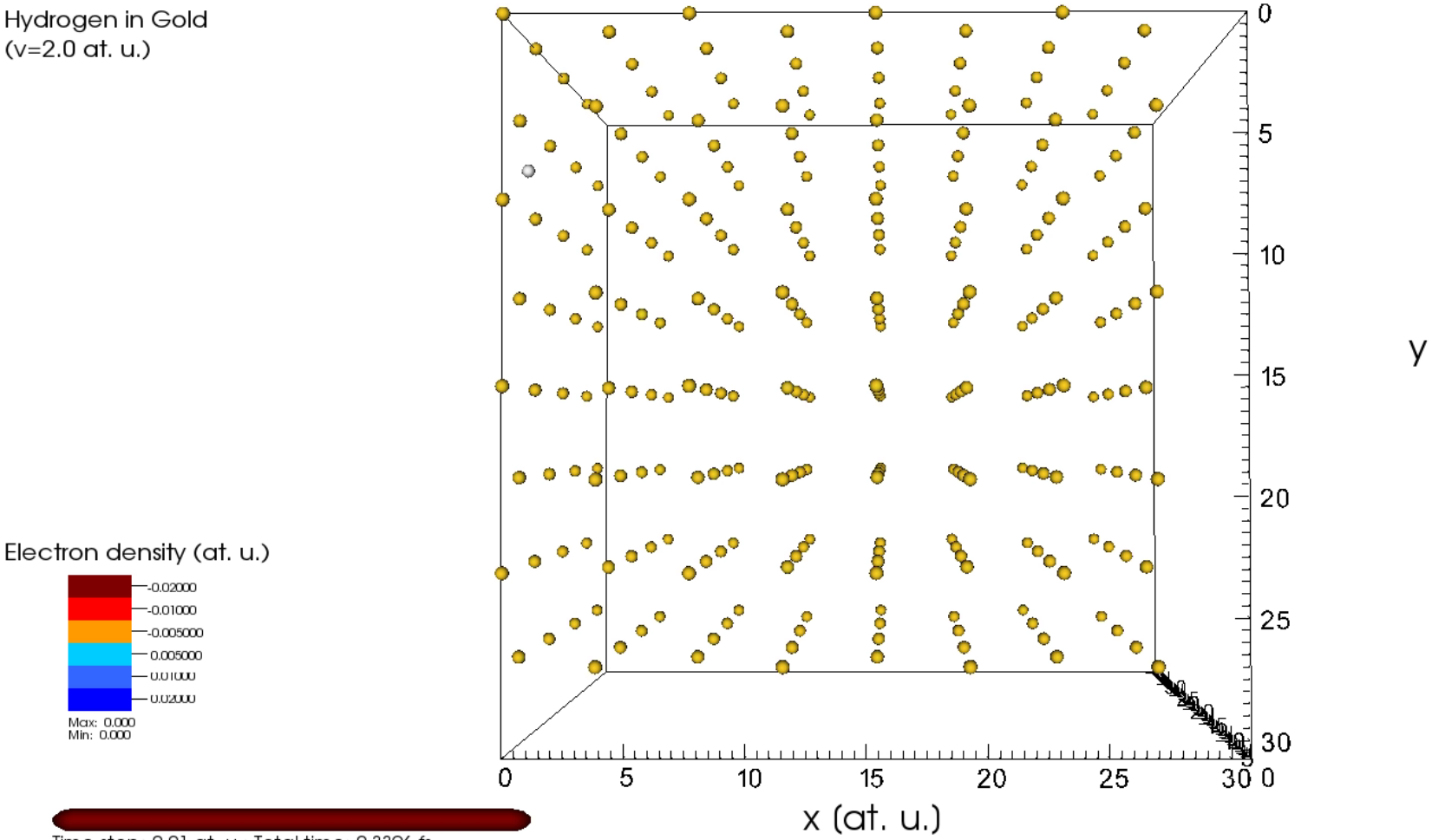
- Capture electron dynamics on atto-second time scale





- Energy transferred to electrons, stopping power: $S = -\frac{\mathrm{d}E}{\mathrm{d}x}$

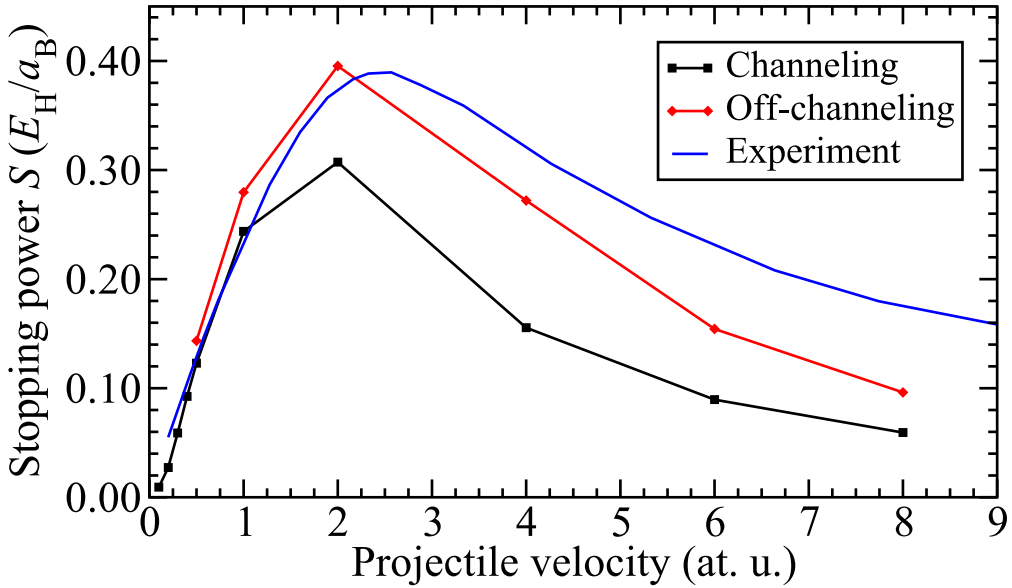
Beyond Born-Oppenheimer: Hydrogen in Gold



A. Schleife, E. Draeger, V. Anisimov, A. Correa, Y. Kanai (2013)

Beyond Born-Oppenheimer: Hydrogen in Gold

Comparison to database with experiments (TRIM):



hydrogen: v=1.0 a.u.≈25 keV

- Systematic convergence to ensure high accuracy
- Excellent agreement for slow projectiles
- Deviations at higher velocities: off channeling

- Investigate initial-state dependence

→ Predictive!

A. Schleife *et al.* (submitted)

Summary and Outlook

Future Perspective:

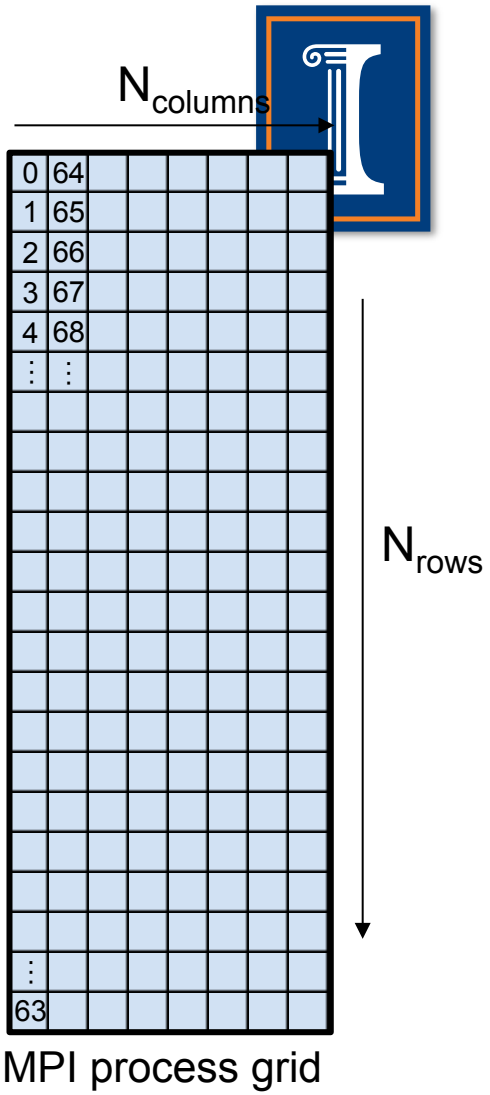
- Essential: Excited electronic states and their dynamics
- Accurate parameter-free description achievable
- Highly parallel implementation in place for leadership-class computers

Towards Predictive Materials Design:

- Umportant to understand for the degradation of nuclear reactor materials
- How is energy deposited into the material
- How do defects evolve?

Parallel Implementation in Qbox code

- LLNL version (qb@ll) is used here
- C++/MPI+OpenMP
- norm-conserving nonlocal pseudopotentials, plane waves
- orbitals distributed on 2D process grid
- each process column owns subset of orbitals and the charge density (typically $N_{rows} >> N_{columns}$)
- parallel dense linear algebra (double or complex) handled using ScaLAPACK (machine-optimized libraries)
- parallel 3D FFTs (complex to complex) carried out within process columns

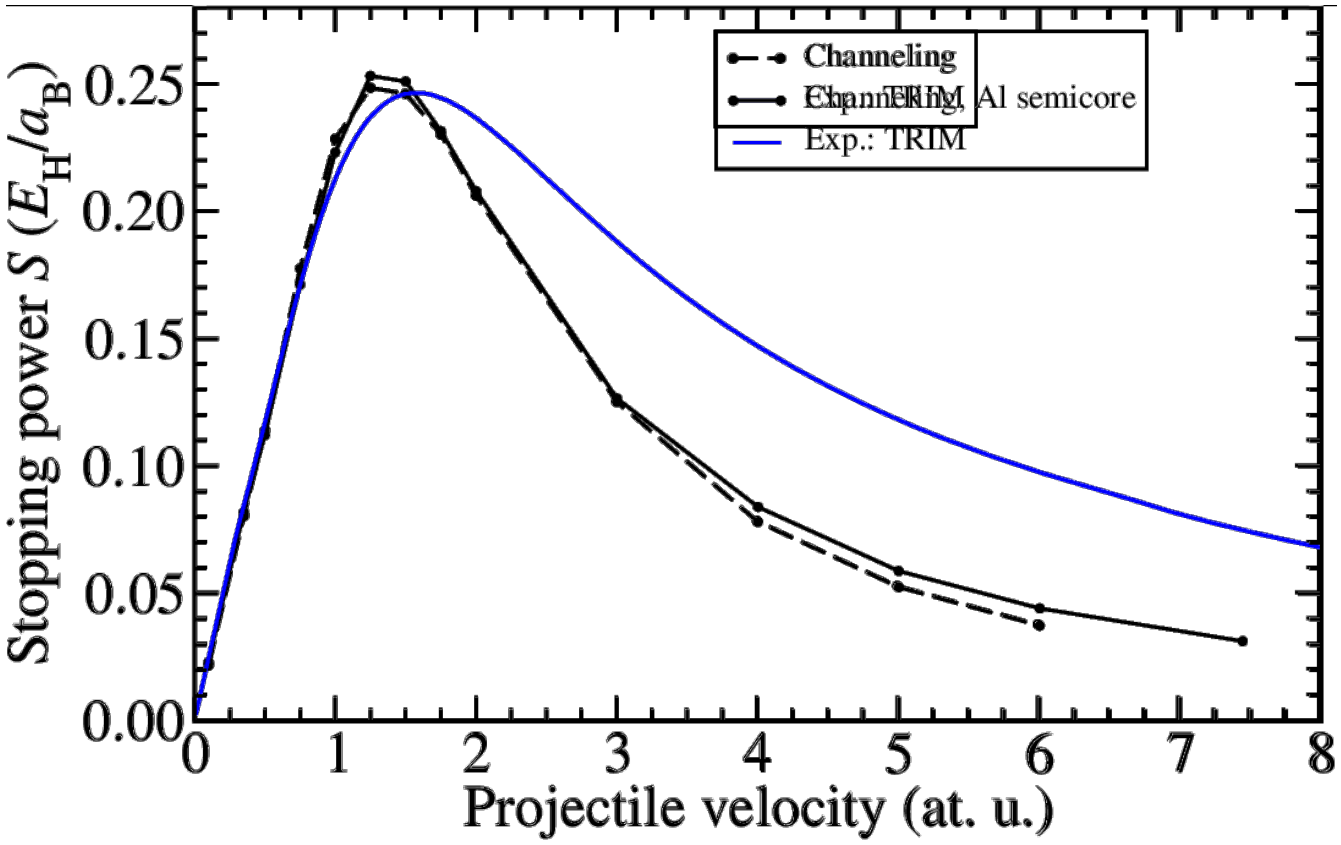


this data distribution allows the majority of communication to occur within non-intersecting subcommunicators

Hydrogen in Aluminum

Comparison to Experiment (TRIM):

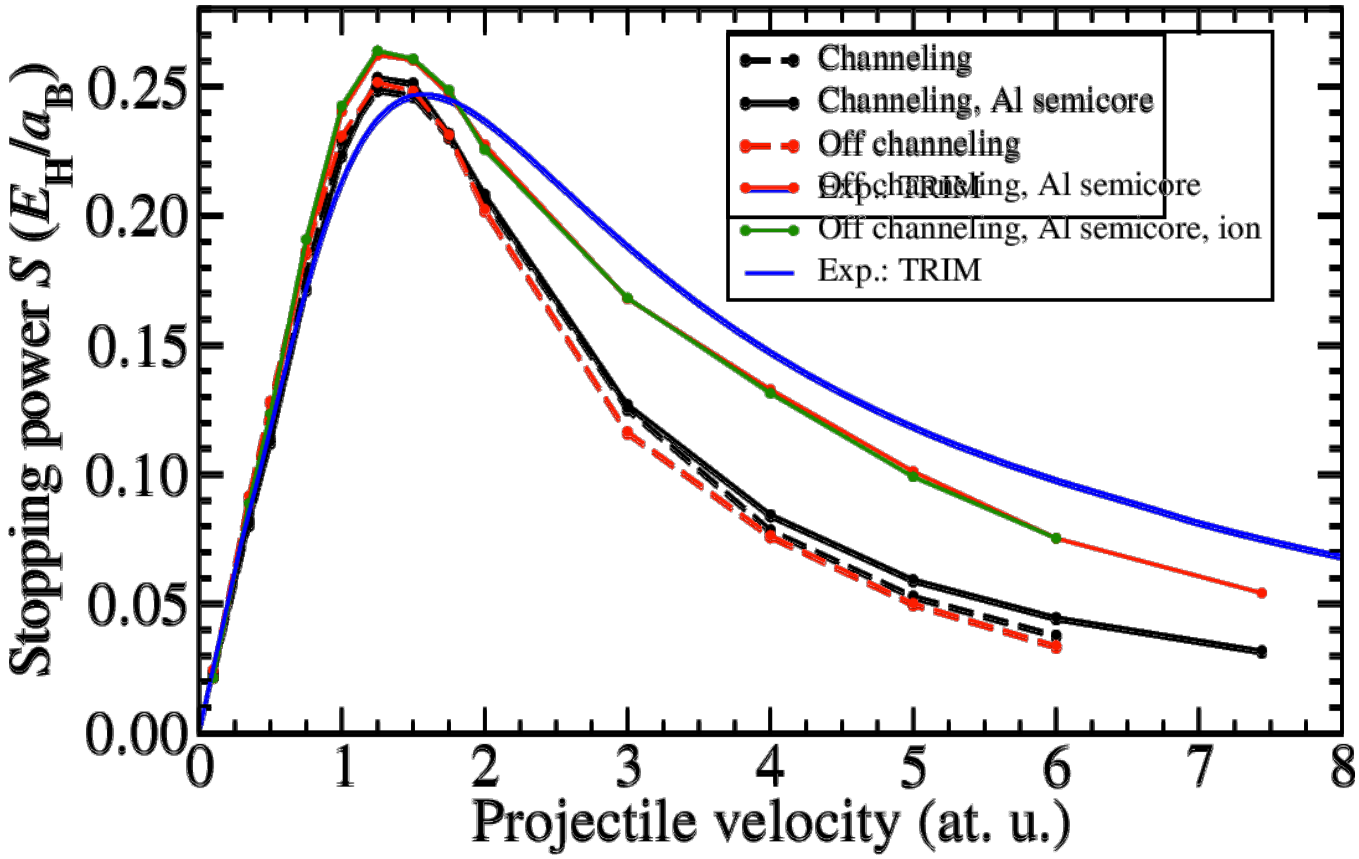
- Excellent agreement for slow projectiles
- Deviations at higher velocities?



Hydrogen in Aluminum

Comparison to Experiment (TRIM):

- Excellent agreement for slow projectiles
- Deviations at higher velocities: Off-channeling!



Helium in Aluminum

- Good overall agreement with experiment
- Practically no influence of initial condition (charge state)