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Electronic stopping in warm dense matter using Ehrenfest dynamics and TDDFT

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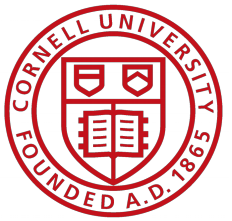

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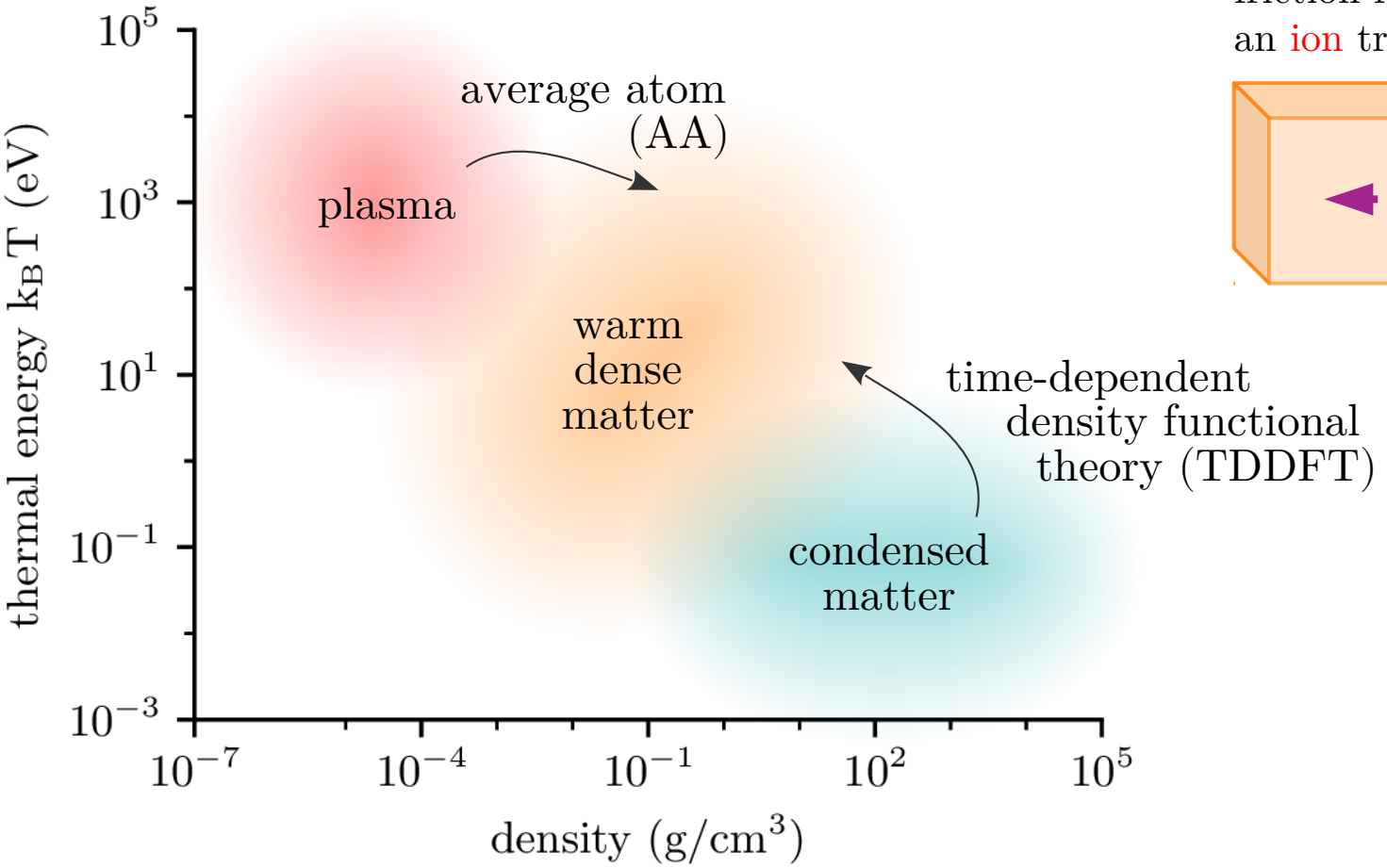
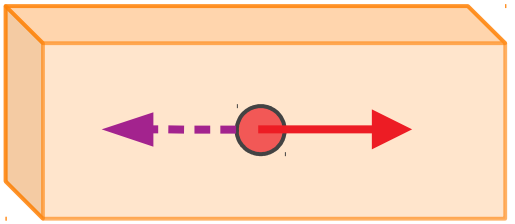
Bridging the Gap between Plasma and Condensed Matter

Goal: benchmark cheaper AA method against more accurate but expensive TDDFT calculations



stopping power:
friction force experienced by an ion traversing matter



Time-Dependent DFT (TDDFT)

Mean-field model of quantum electron dynamics

Initial condition: ground state from DFT

Evolve electron density $n(\mathbf{r}, t)$ over time

$$i\frac{\partial}{\partial t}\phi_j(\mathbf{r}, t) = \hat{H}[n(\mathbf{r}, t)]\phi_j(\mathbf{r}, t)$$

$$n(\mathbf{r}, t) = \sum_j f_j(T)|\phi_j(\mathbf{r}, t)|^2$$

$$\hat{H}[n(\mathbf{r}, t)](t) = -\frac{\nabla^2}{2} + V_{\text{ext}}(t) + \int \frac{n(\mathbf{r}', t)}{|\mathbf{r} - \mathbf{r}'|}\, d\mathbf{r}'^3 + V_{\text{xc}}[n(\mathbf{r}, t)]$$

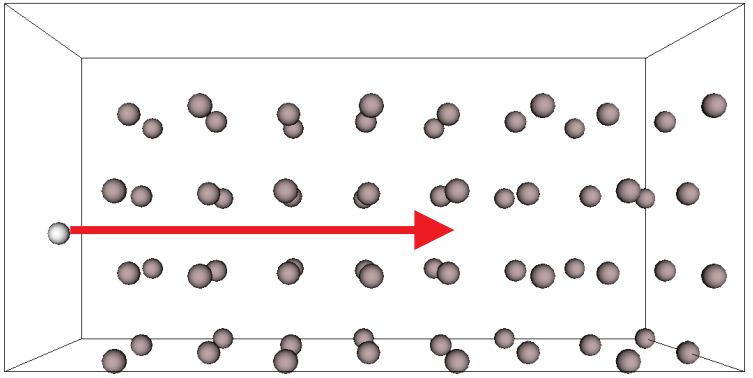
kinetic energy

external potential due to ions

Coulombic e-e interaction

exchange-correlation

explicit time-dependence from moving proton



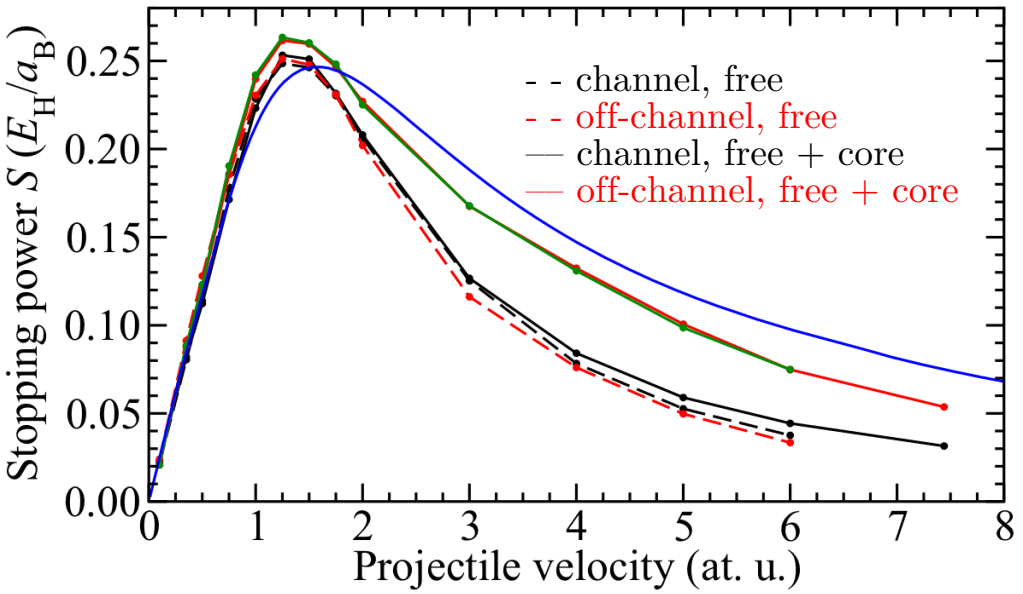
pseudopotential approximation

adiabatic local density approximation

Stopping power $\sim \mathbf{F}_\alpha[n](t) = -\left\langle \frac{\partial \hat{H}[n]}{\partial \mathbf{R}_\alpha} \right\rangle$

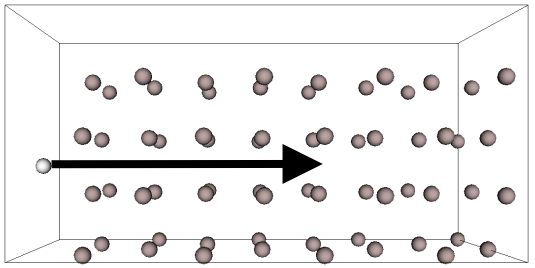
Background on Trajectories in Crystals

- Stopping in cold metals well-characterized
- Free electrons ~uniform
  - stopping ~independent of proton trajectory
- Core electrons highly localized
  - stopping sensitive to proton trajectory

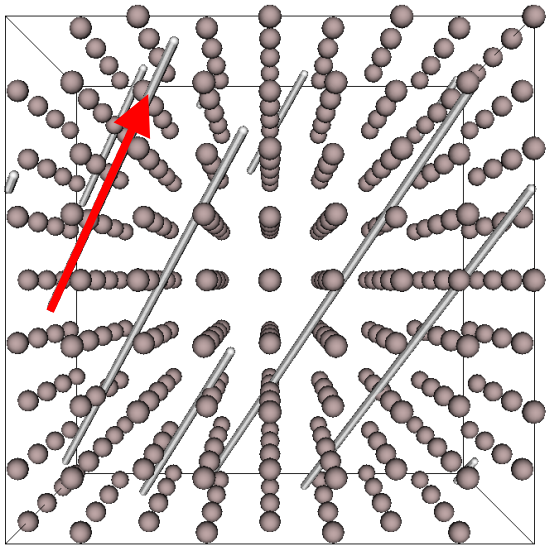


Schleife et al., PRB 91 (2015)

channeling



off-channeling



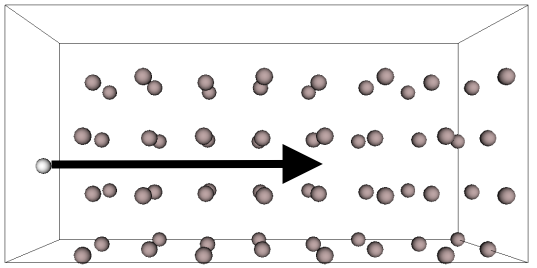
But how do we know if we've picked a "good" trajectory?

Quantitative Metric to Evaluate Trajectories

- Projectile should experience representative NN distances
- Ideal NN distribution: sample random points in cell
- Good trajectory achieves low Bhattacharyya distance

$$D_B = -\ln\left(\int \sqrt{P_{\text{traj}}(\delta_{NN}) P_{\text{ideal}}(\delta_{NN})}\, d\delta_{NN}\right)$$

channeling



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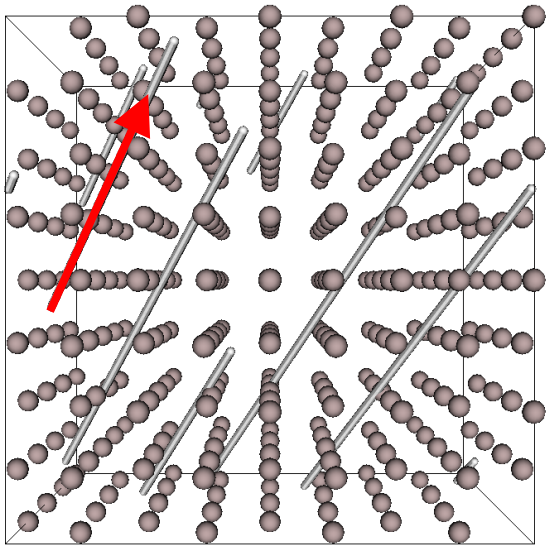
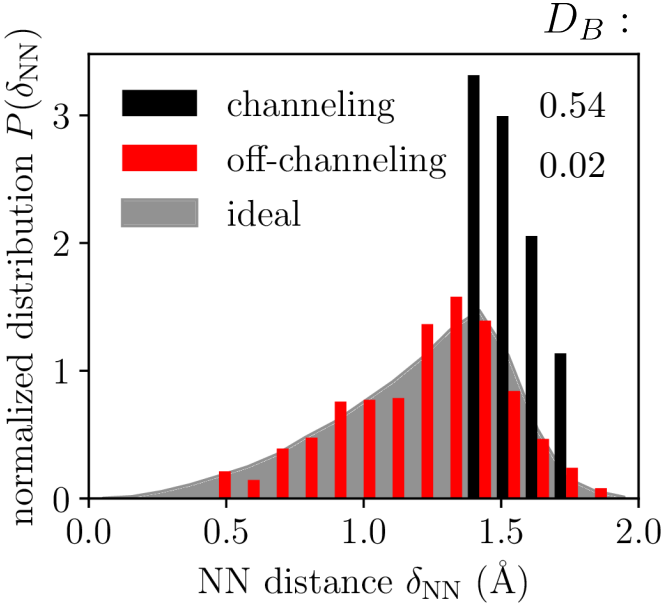
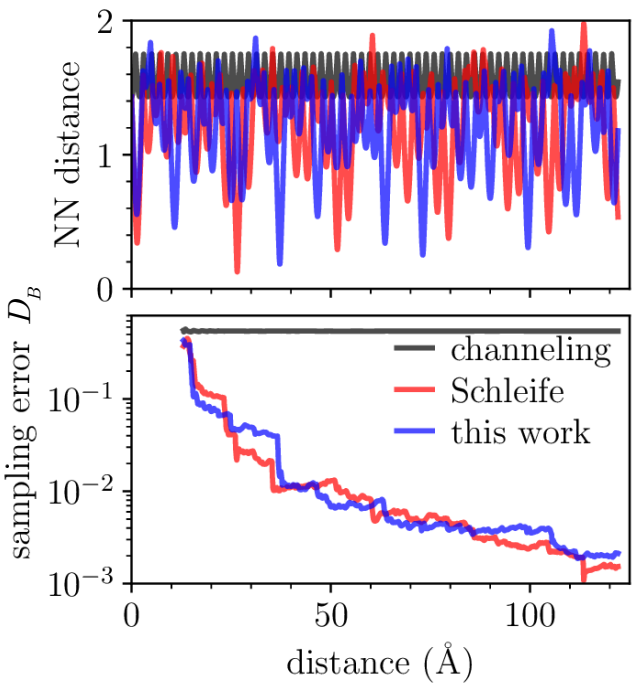
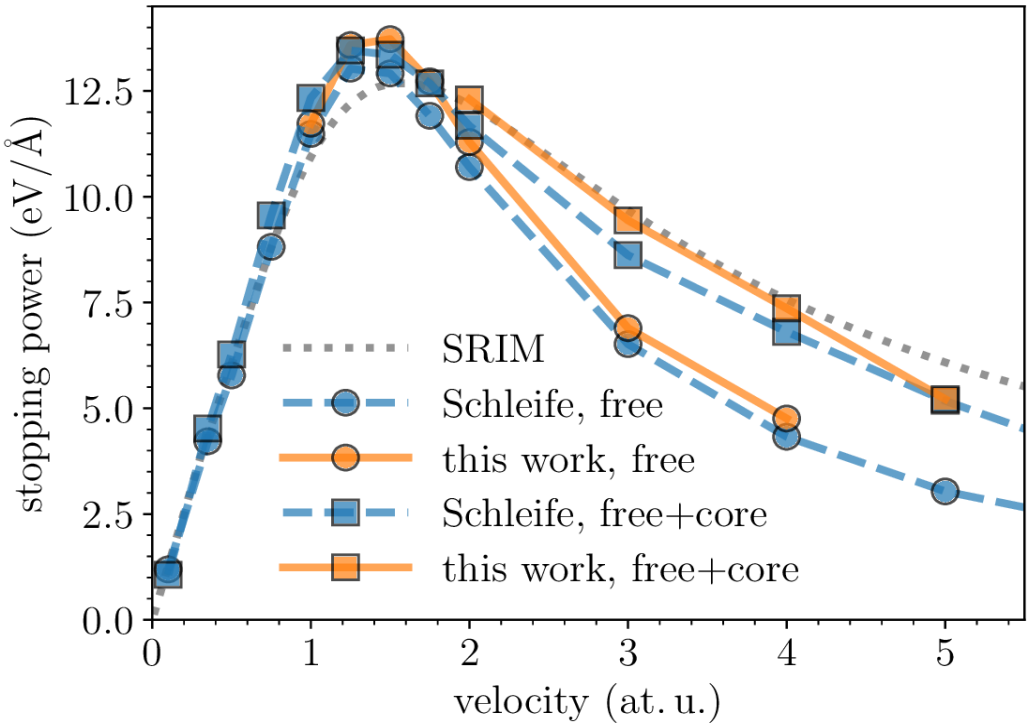






Proton Stopping in Aluminum: validation

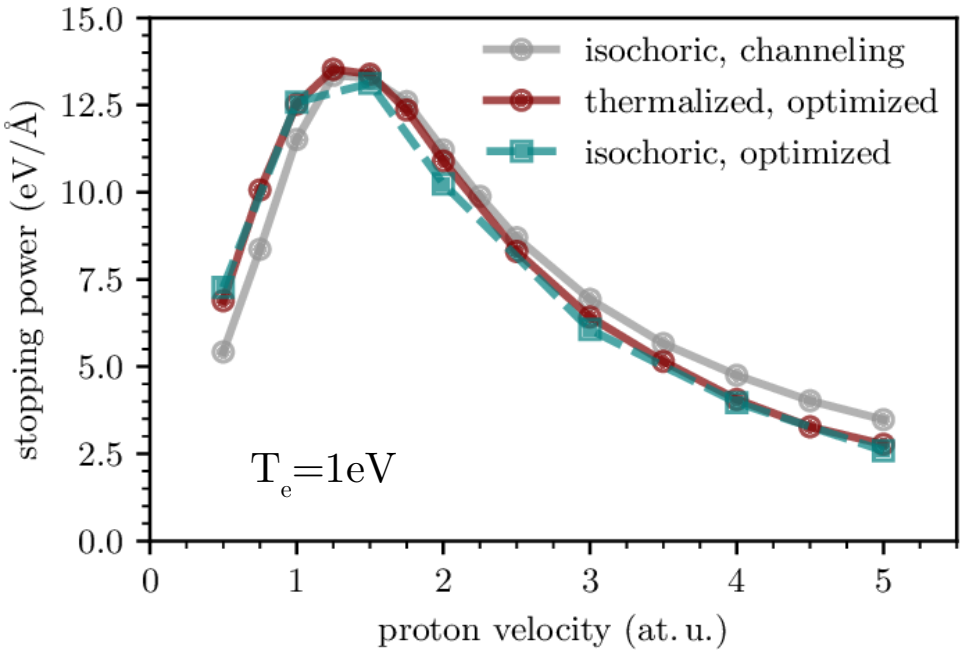
- Validated methodology for cold Al
  - good agreement with earlier TDDFT and empirical data
  - modest discrepancies from different trajectories, pseudopotentials



Schleife et al., PRB 91 (2015)
Ziegler et al., Nucl. Instrum. Methods B 268 (2010)

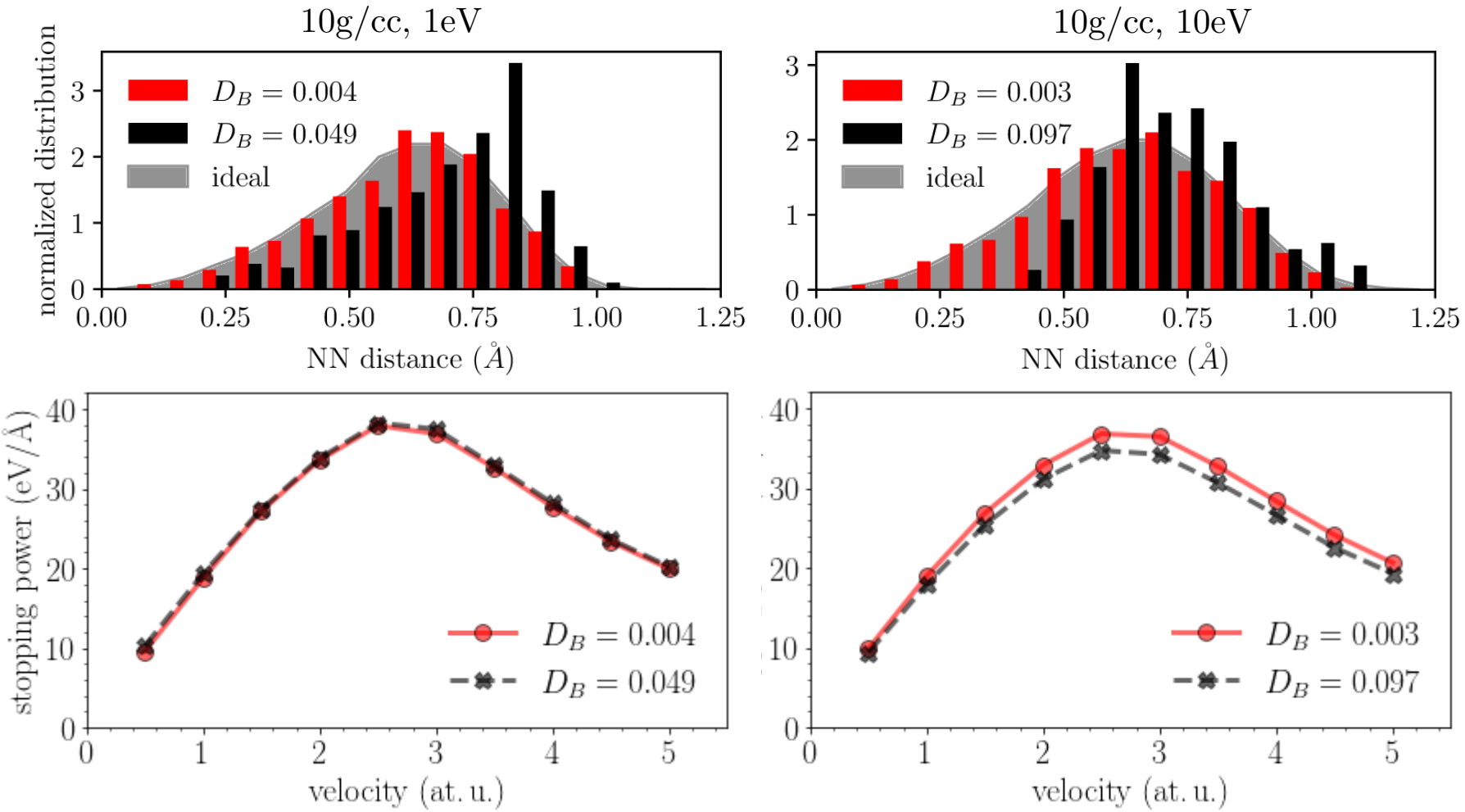
Proton Stopping in Aluminum: ion temperature

- Trajectory metric allows controlled comparisons in WDM
  - across different T
  - across different atomic configurations
  - thermalized vs. isochorically heated systems
- Free-electron stopping independent of ion temperature
  - slight variation with projectile trajectory
  - select similarly optimized trajectories
- To do: check independence of core contribution



Proton Stopping in Liquid Carbon: trajectory

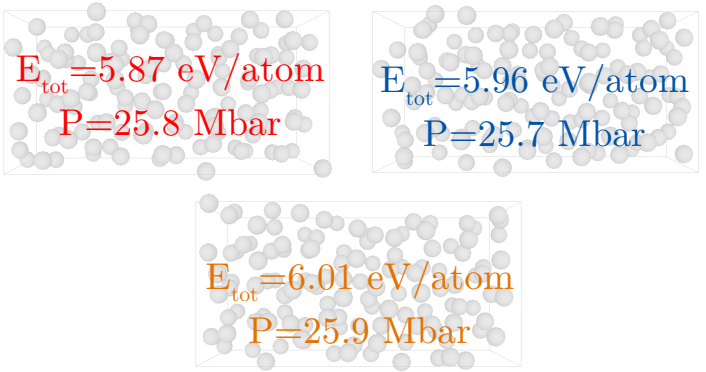
- Hard to find "bad" trajectory in disordered, low-Z system
- Fewer localized electrons in carbon than aluminum
- Little sensitivity to proton trajectory



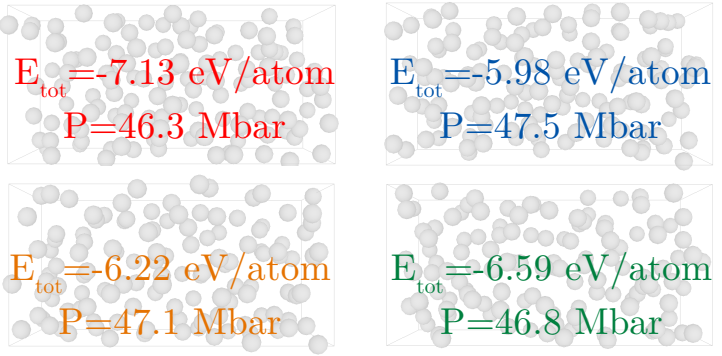
Proton Stopping in Liquid Carbon: atomic configurations

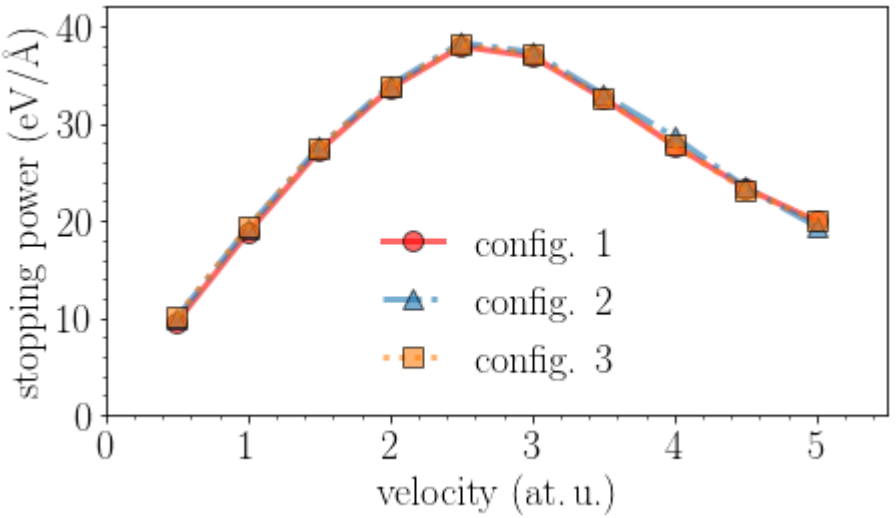
- Thermal fluctuations may affect stopping
- Separately optimized trajectory for several MD snapshots
- Little variation across atomic configurations
- Trajectory metric may eliminate need for configurational averaging

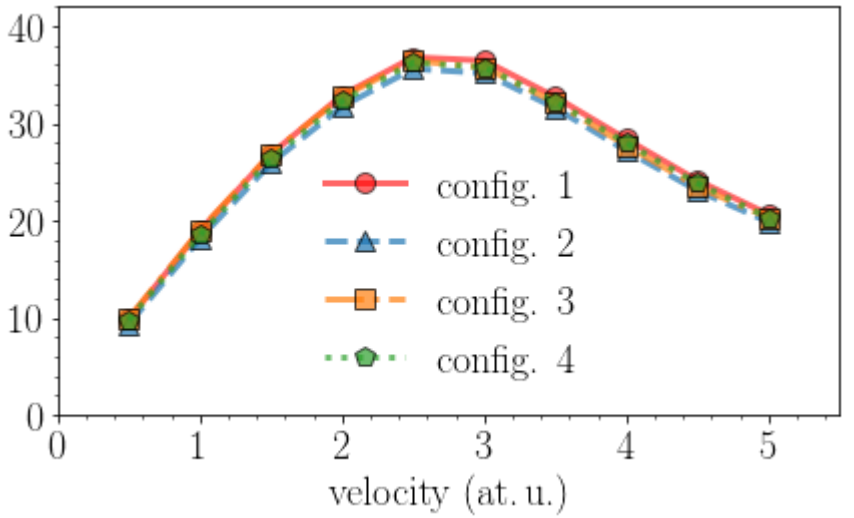
10g/cc, 1eV



10g/cc, 10eV

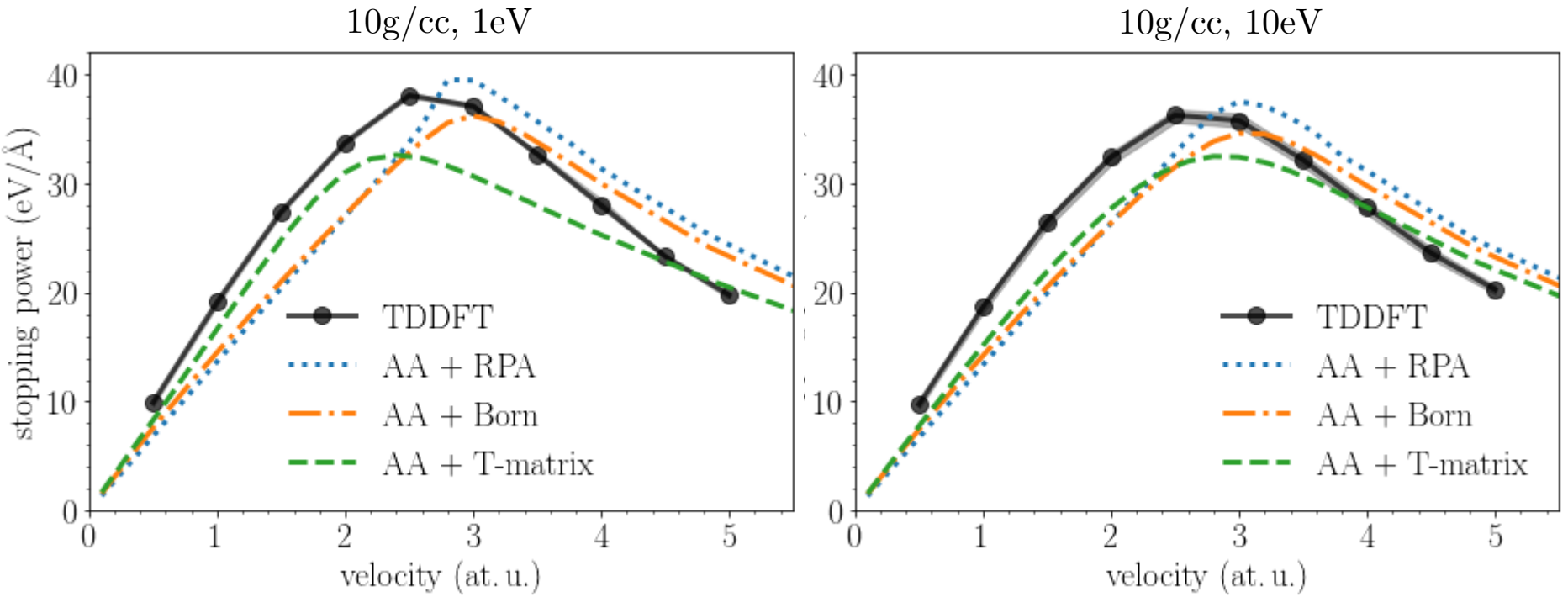






Proton Stopping in Liquid Carbon: comparing to AA

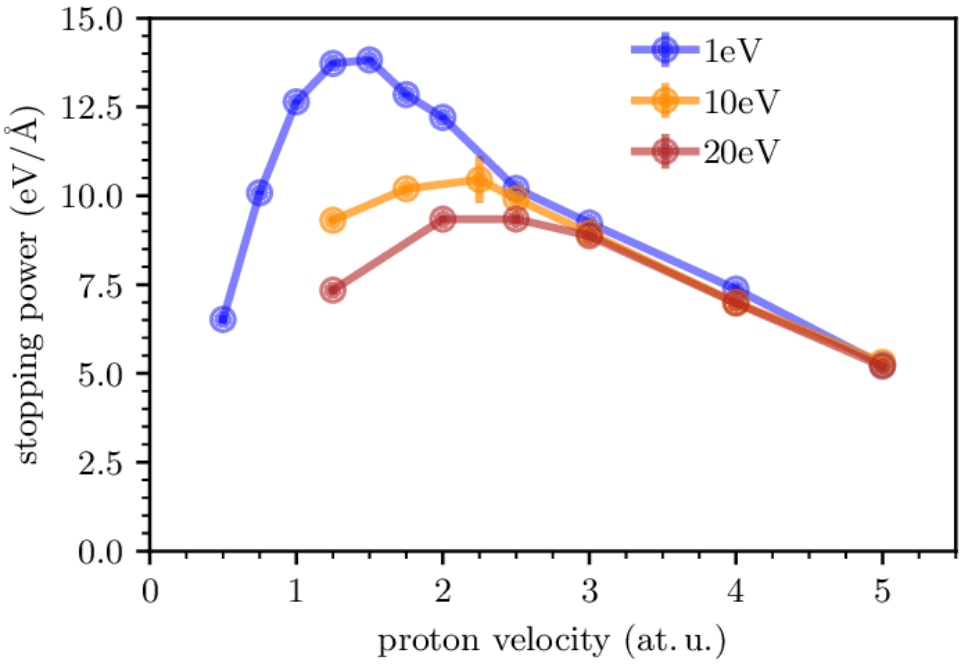
- Stopping power from dielectric models: $S(v) = \frac{2Z_I^2}{\pi v^2} \int_0^\infty \frac{dk}{k} \int_0^{kv} d\omega\, \omega\, \mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right]$
- RPA, Born predict more accurate peak height
- T-matrix predicts more accurate peak position, low-v slope
- Significant discrepancies with all AA models

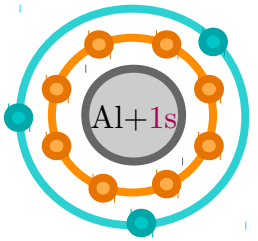


Proton Stopping in Aluminum: electron temperature

- At high T_e, Bragg peak lowers and shifts to higher velocities
- Different pseudizations offer rough insight:
  - 11e PP: total stopping

- 256 Al atoms
- ~70 bands per atom
- ~20k bands
- ~10 million CPU-hours
- per data point





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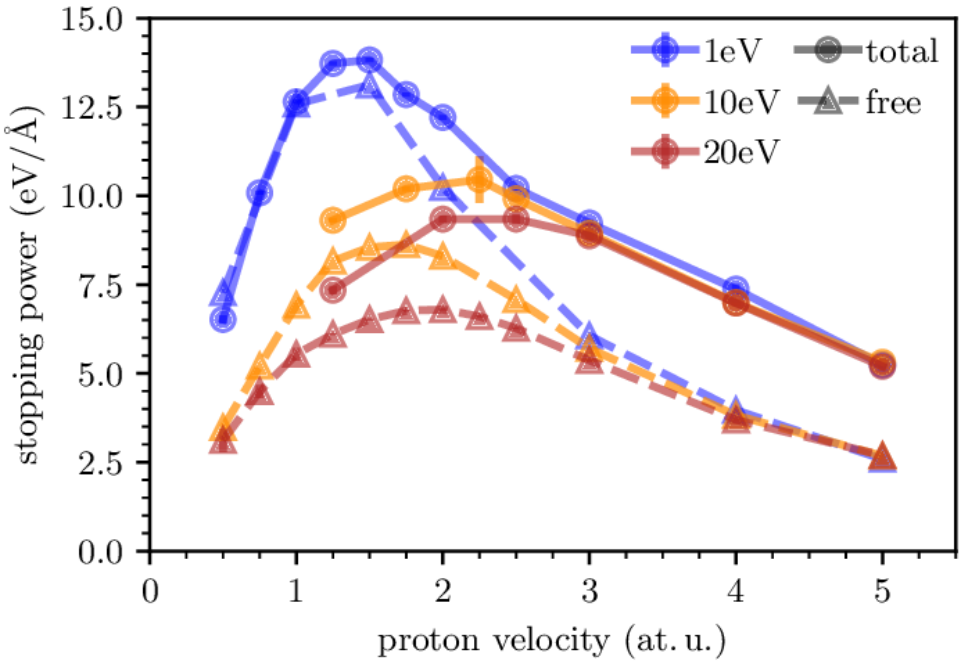

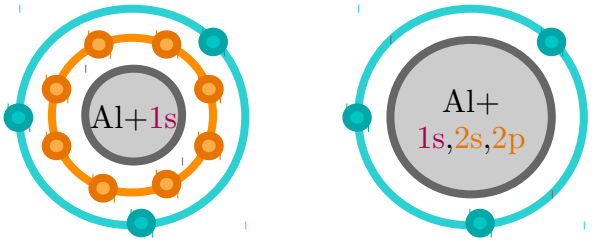

Proton Stopping in Aluminum: electron temperature

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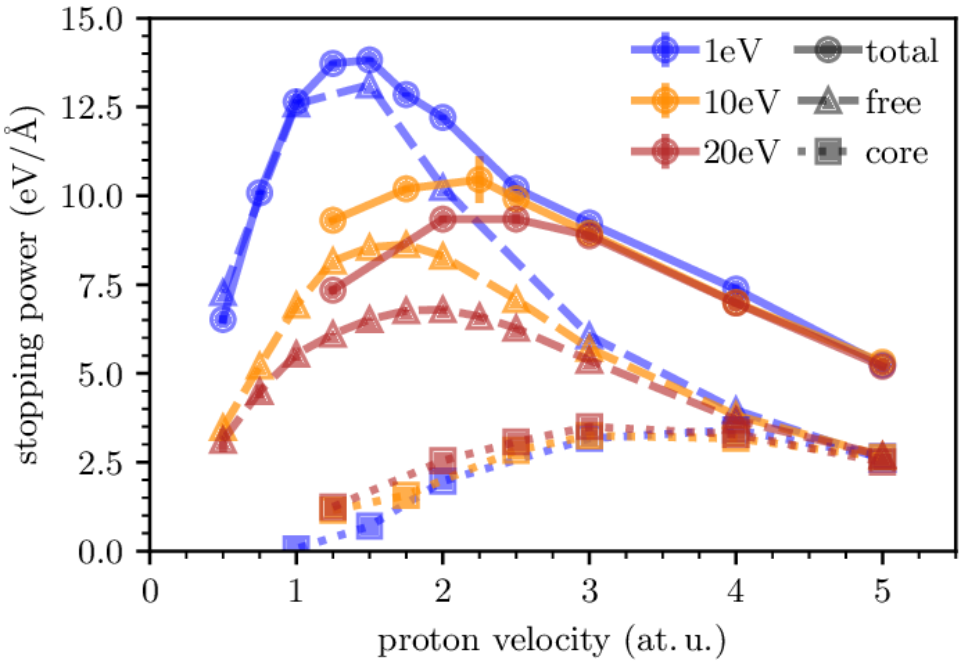
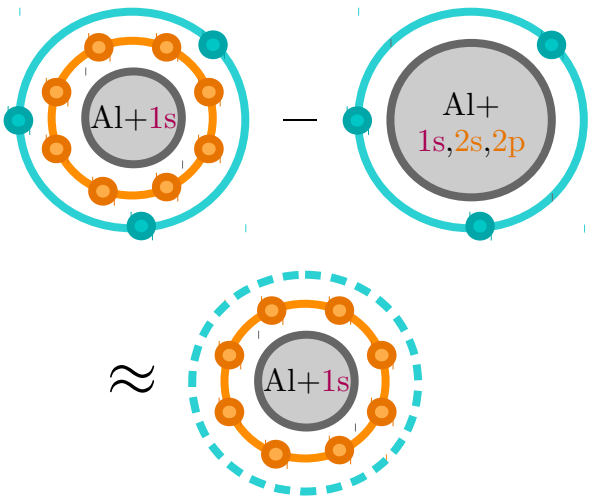
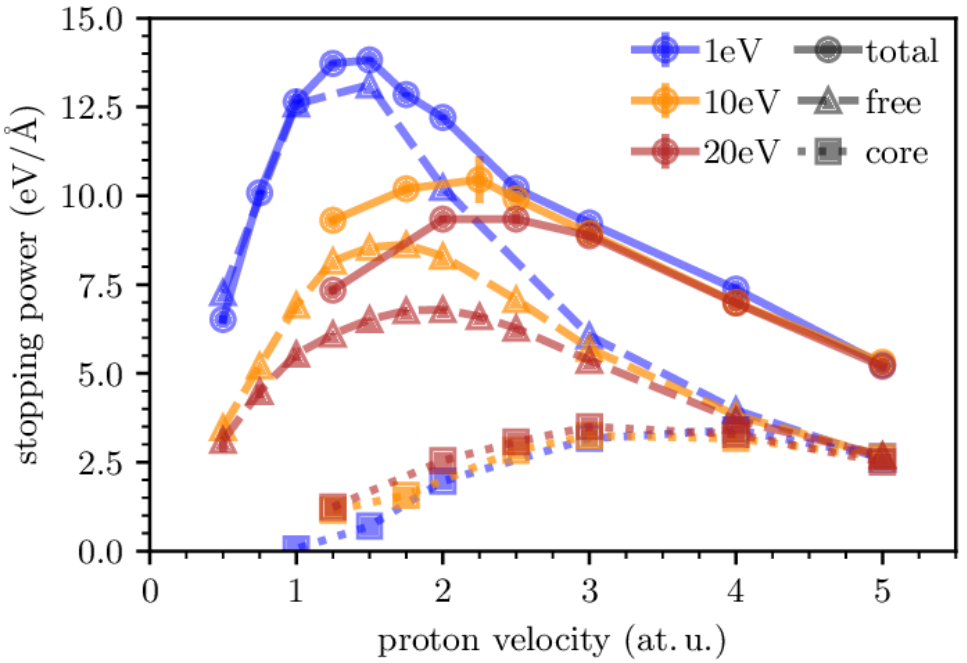
- Different pseudizations offer rough insight:
  - 11e PP: total stopping
  - 3e PP: ~free-electron contribution follows same trend as total
  - 11e PP – 3e PP: ~core contribution not sensitive to T_e, but accounts for increasing fraction of total





Proton Stopping in Aluminum: electron temperature

- Competing effects obfuscated!
- Thermal excitations increase free-electron density
  - 3ePP underestimates free-electron contribution at 20eV



	1eV	10eV	20eV
free electrons per atom	3.00	3.02	3.61

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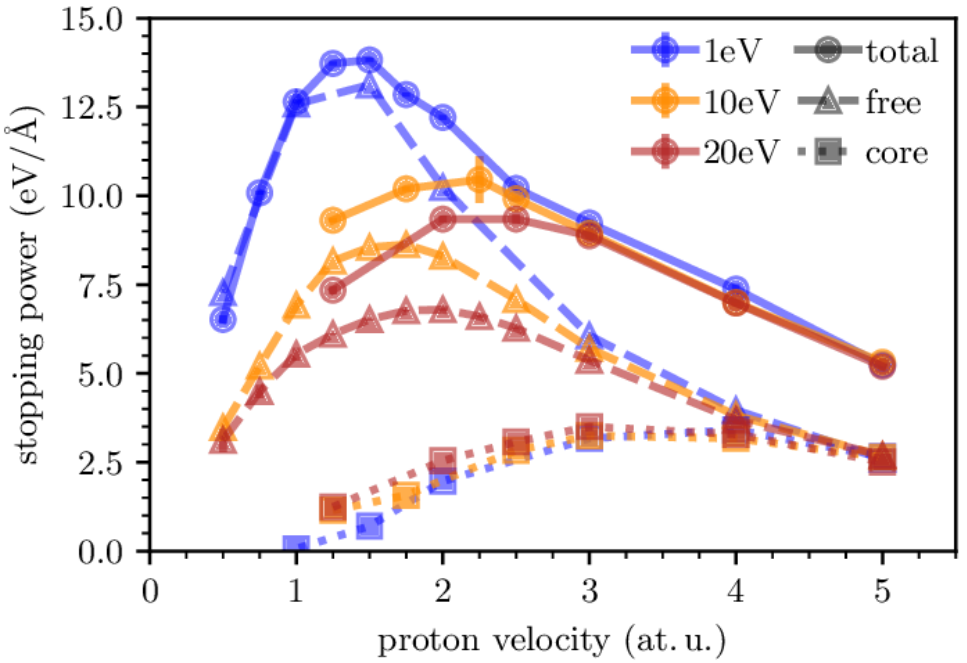
- Thermal depletion of low-energy free states and deeper 2p binding alter 2p $\rightarrow$ free energetics



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2p – free energy difference (eV)	65	55	62.5

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- Thermal excitations increase free-electron density
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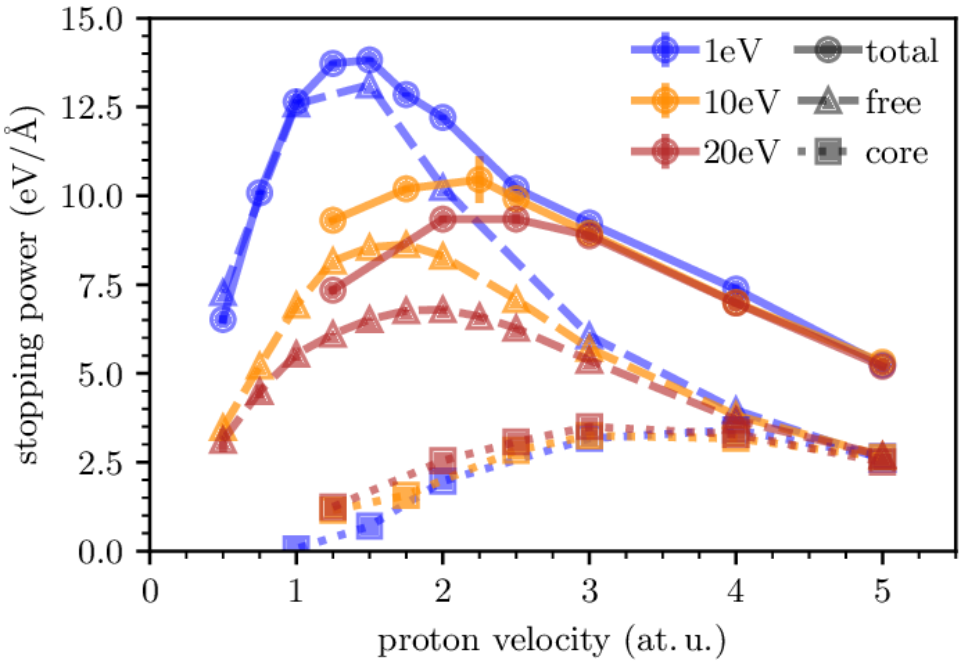
- Thermal depletion of low-energy free states and deeper 2p binding alter 2p → free energetics
- Thermal depletion of 2p allows 2s → 2p at 20eV
- Working to disentangle these processes



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free electrons per atom	3.00	3.02	3.61
2p – free energy difference (eV)	65	55	62.5
2p vacancy (%)	0	0.5	9.6

Summary and Outlook

- Developed metric for trajectories in TDDFT stopping calculations
  - most important in high-Z and/or isochoric systems like Al
- Found only slight trajectory, configurational effects in liquid C
- Benchmarking AA + dielectric models for free-electron stopping
- Studying core electron contributions in Al at high T
- Ultimately interested in mixtures relevant to fusion