Core contributions to stopping powers in warm dense matter

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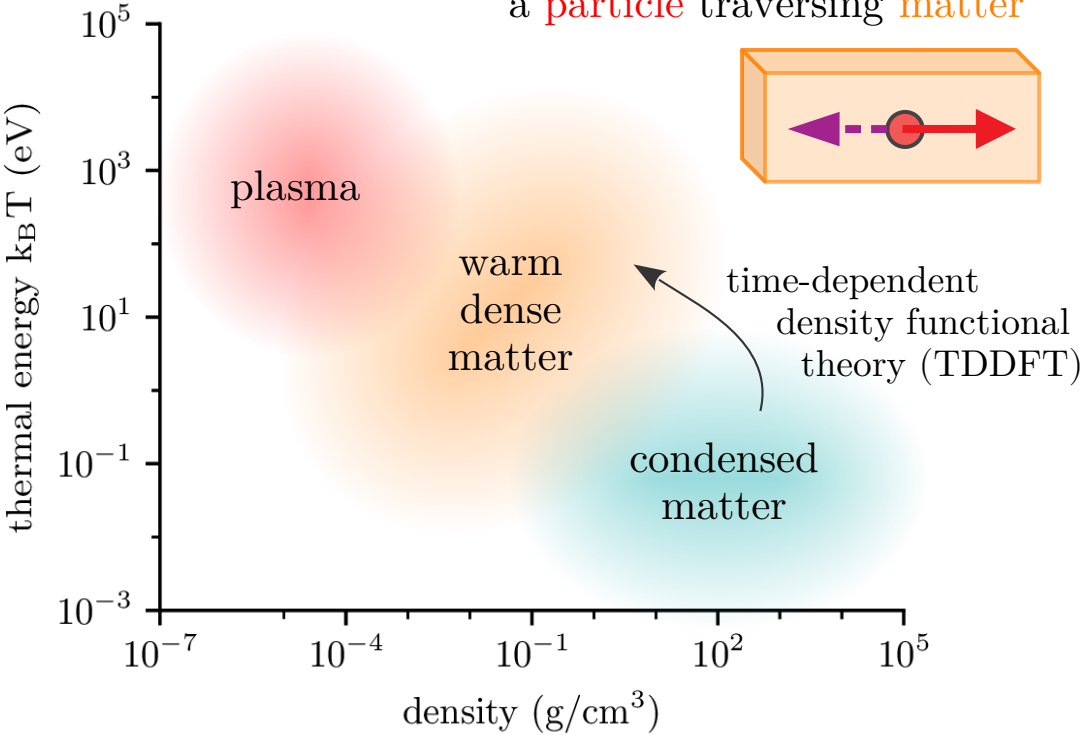
Bridging the gap between plasma and condensed matter

- Fusion fuel passes through poorly understood WDM regime on the way to ignition
  - competing physics challenge models
  - scarce experimental data, uncertain conditions limit validation
- Self-heating through electronic stopping influences ignition requirements
- This work: first-principles benchmark data using TDDFT
- Also studying other transport properties
  - XRTS: arXiv:2109.09576
  - conductivity: Robinson, B61.07

stopping power:
friction force experienced by
a particle traversing matter



Electronic stopping powers from TDDFT

- Mean-field model of quantum electron dynamics
- Initial condition: equilibrium state from Mermin-DFT
- Evolve electron density $n(\mathbf{r}, t)$ over time
- Stopping power ~ $\mathbf{F}_{\alpha}[\mathrm{n}](\mathrm{t})$

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

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

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

kinetic energy

external potential due to ions

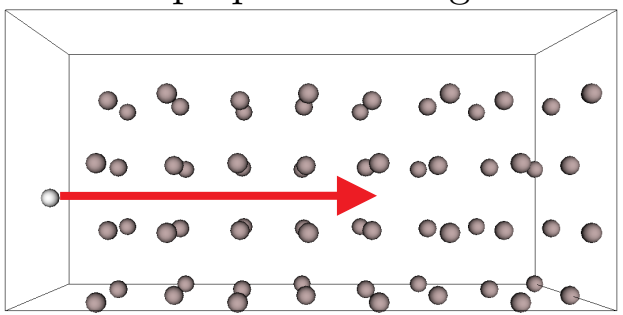
Coulombic e-e interaction

exchange-correlation

projectile trajectory
is an approximation!

Olmstead, N59.05

arXiv preprint coming soon

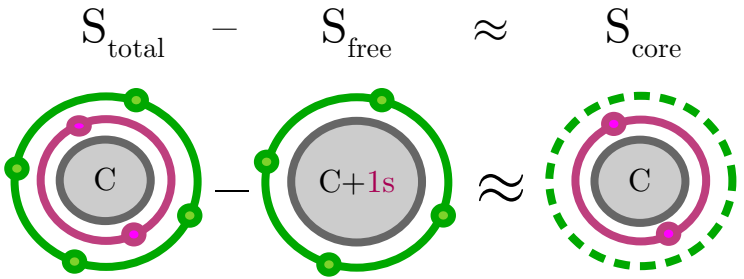


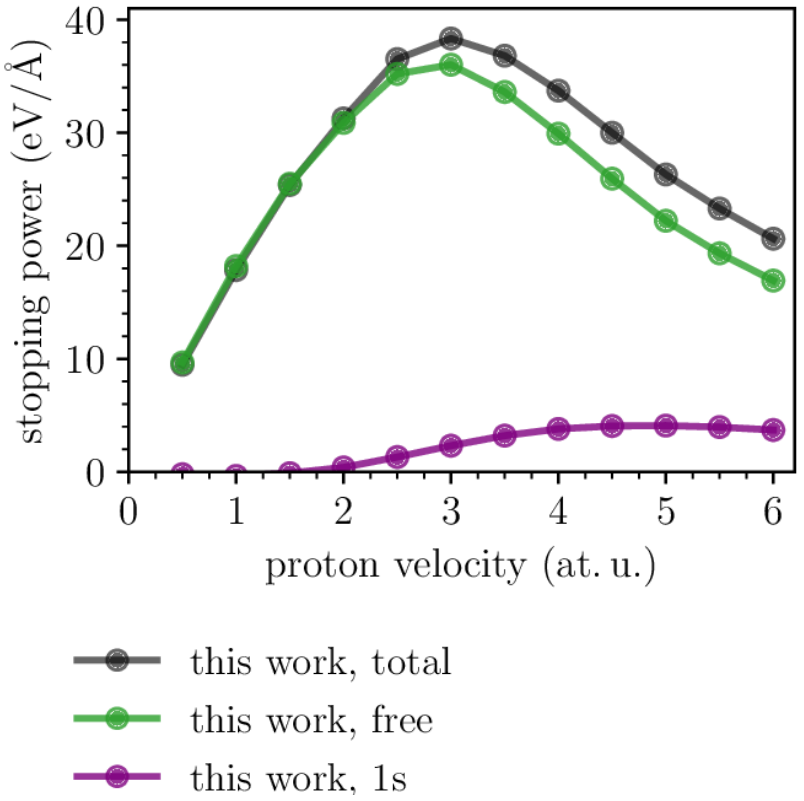
pseudopotential approximation allows detailed insights

adiabatic local density approximation

Orbital-resolved contributions through pseudization

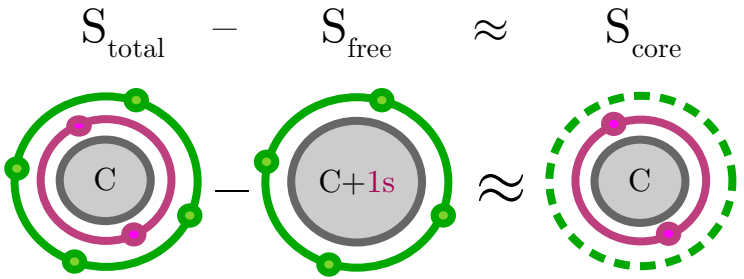
- C at 10 g/cc, 10 eV

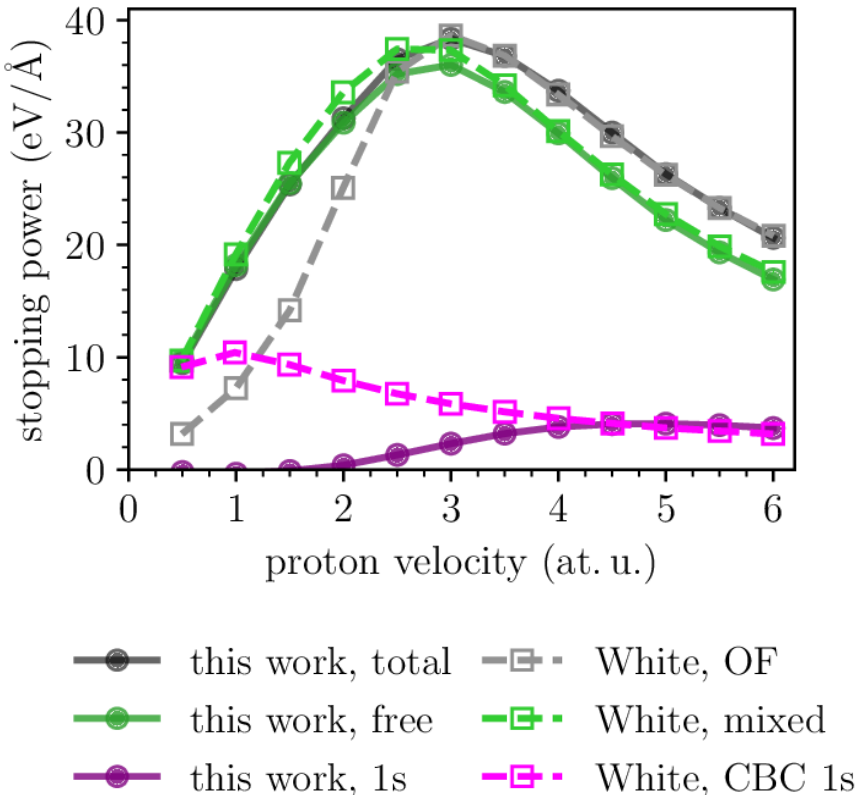




Simplified models of core contributions lose accuracy near/below Bragg peak

- C at 10 g/cc, 10 eV
- Reasonable agreement across TDDFT flavors
- Opposite trends in TDDFT and CBC 1s contributions
- Core contributions warrant further scrutiny

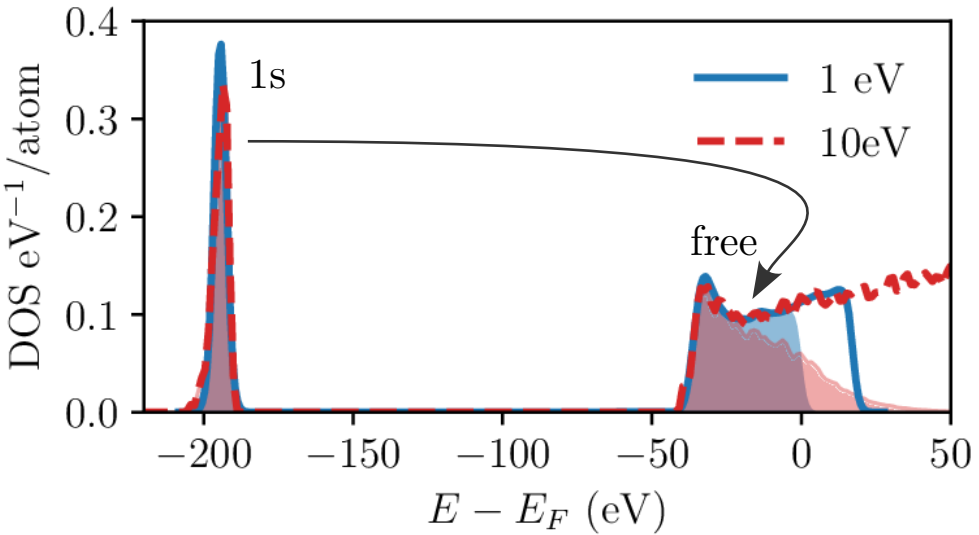


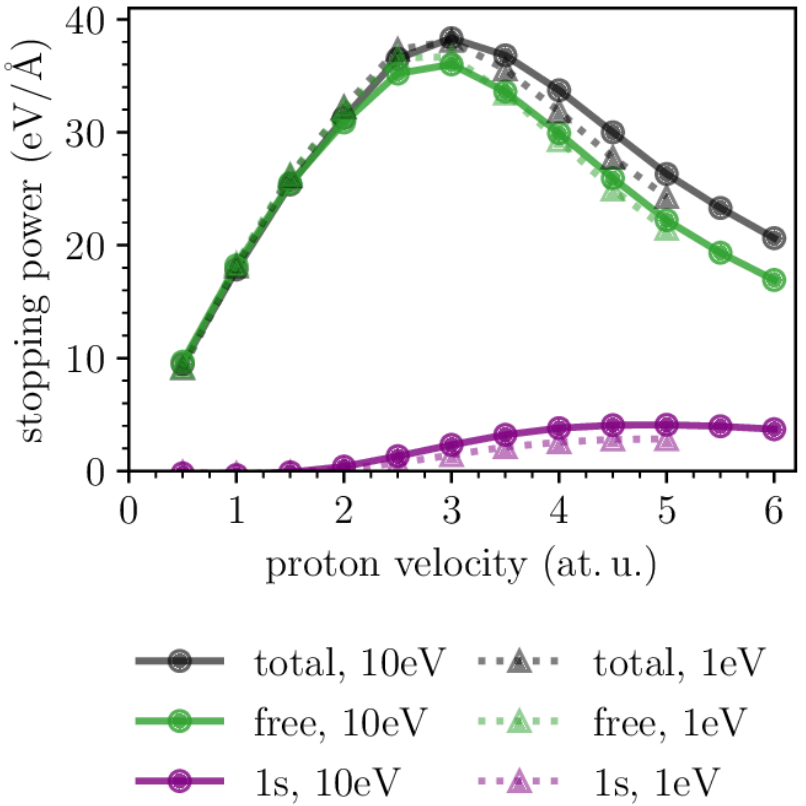


White, J. Phys. Condens. Matter 34 (2022)
Barriga-Carrasco and Casas, Laser Part. Beams 31 (2013)

Electron temperature enhances core contributions in C

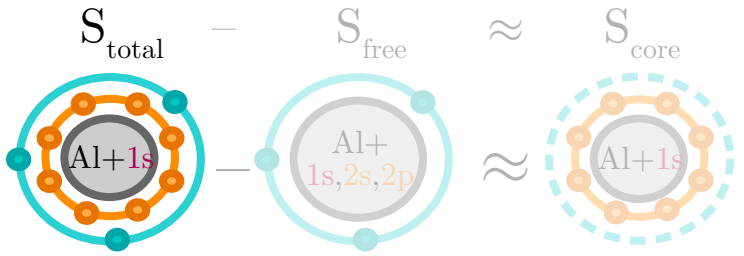
- ~50% higher 1s contribution at T_e =10eV vs 1eV
- Thermal vacancies facilitate 1s → free excitations

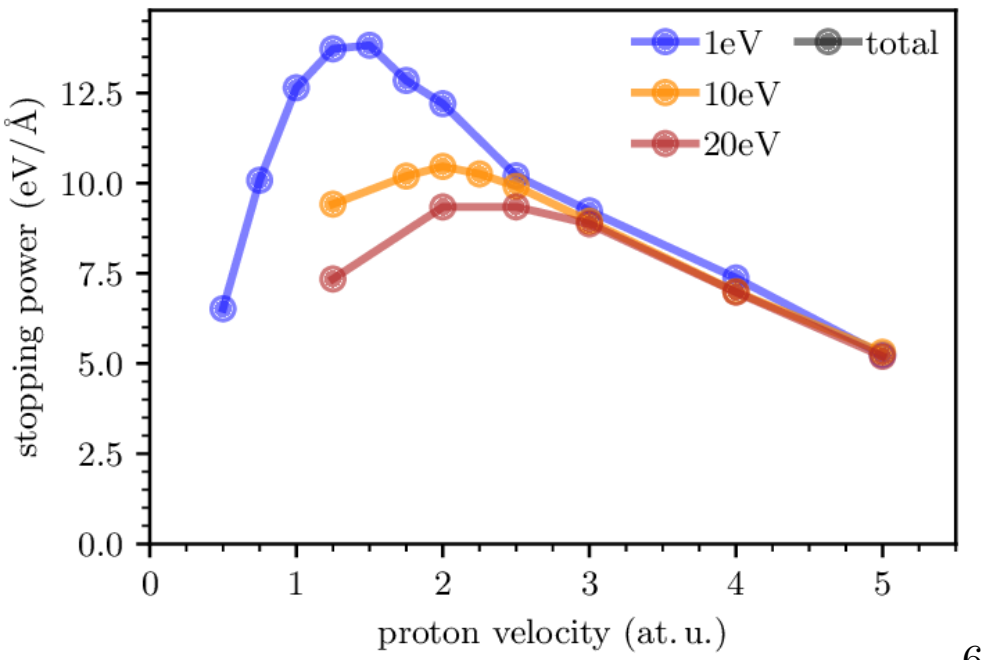




Competing mechanisms affect core contributions in Al

- At high T_e, Bragg peak lowers and shifts to higher velocities





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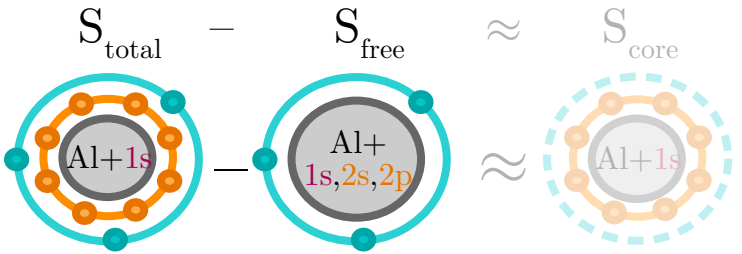
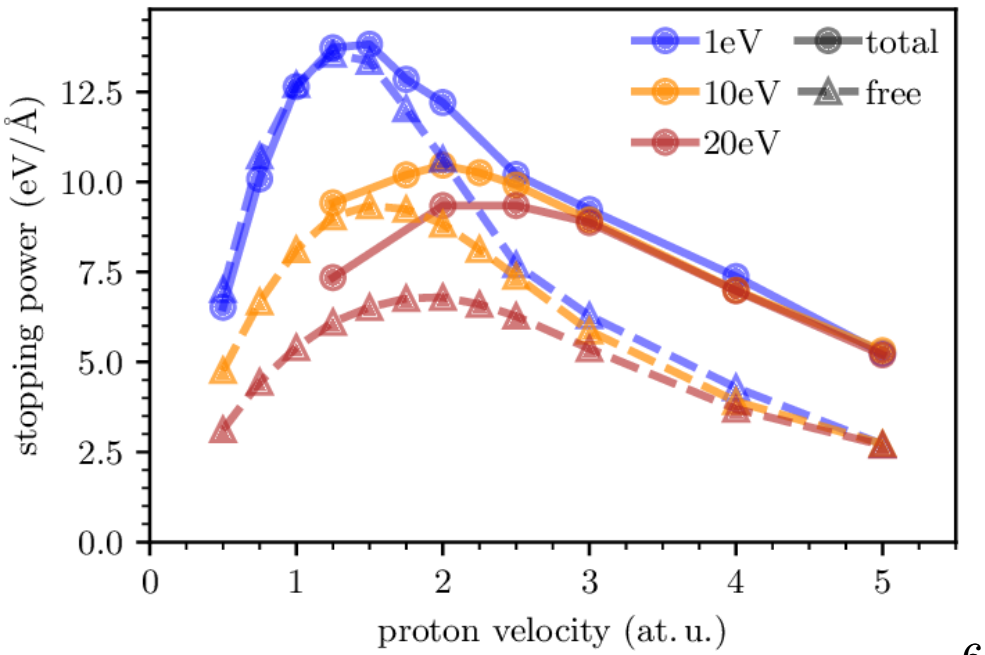
  - 3e PP underestimates S_{free} at T_e=20eV



T_e	1eV	10eV	20eV
free e^- per atom	3.00	3.02	3.61



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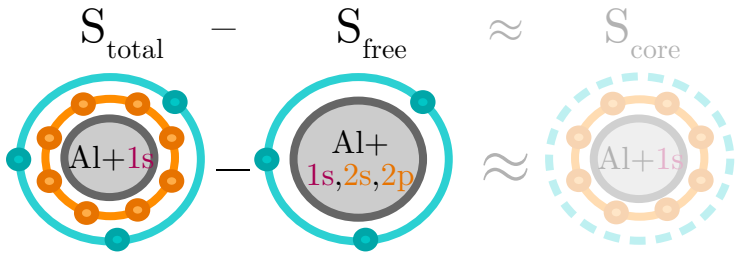

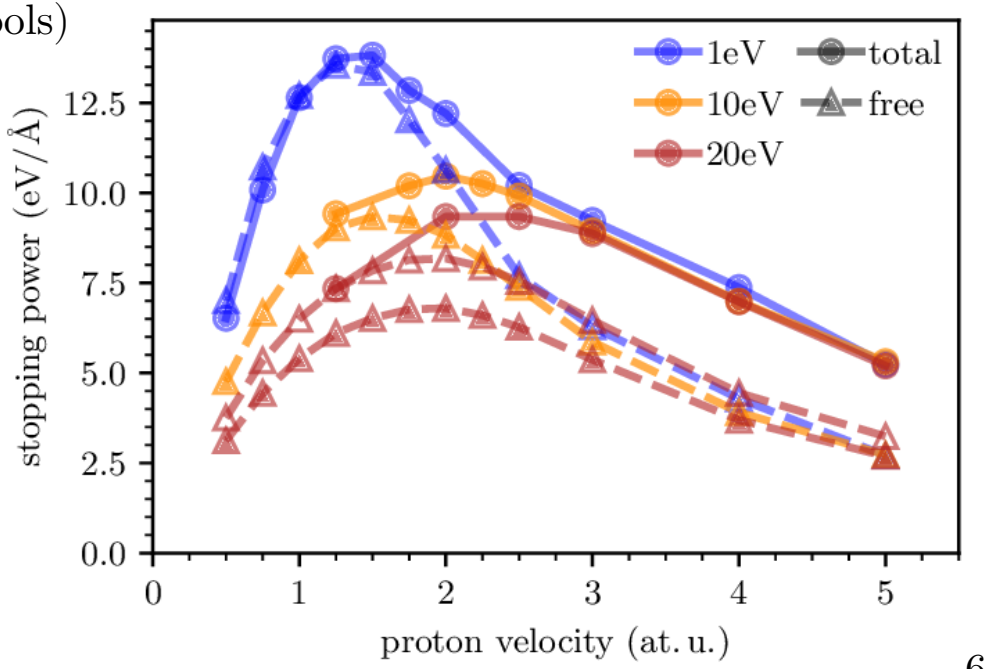

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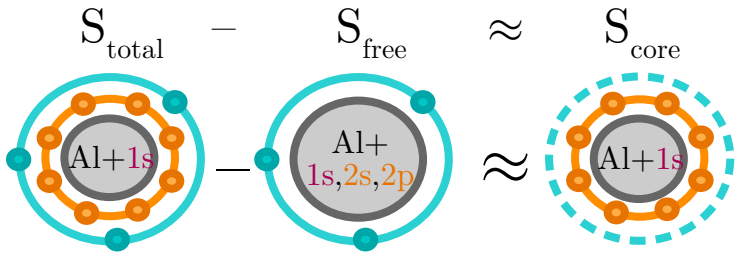

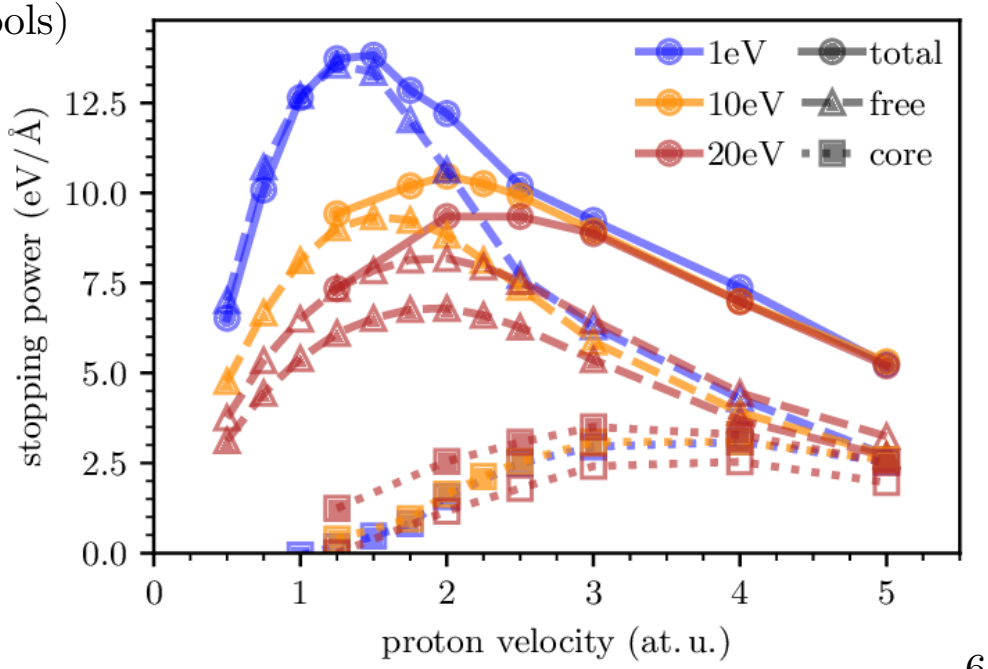

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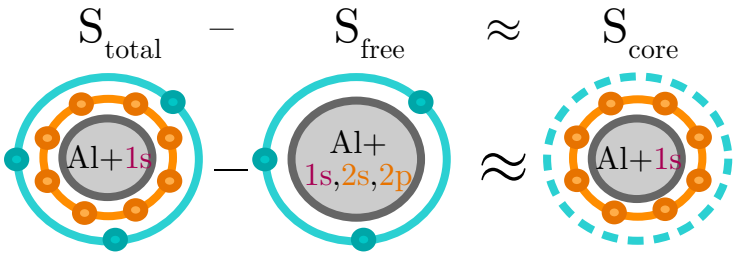
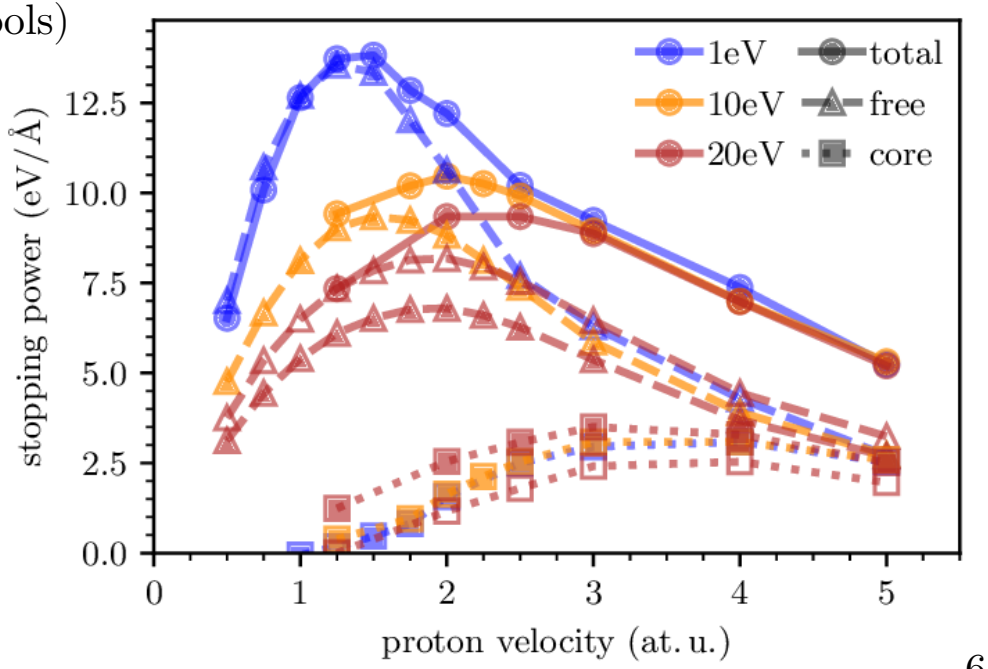
- Thermal depletion of low-energy free states and deeper 2p binding alter 2p → free ΔE

T_e	1eV	10eV	20eV
free e⁻ per atom	3.00	3.02	3.61
$E_{free} - E_{2p}$ (eV)	65	55	62.5





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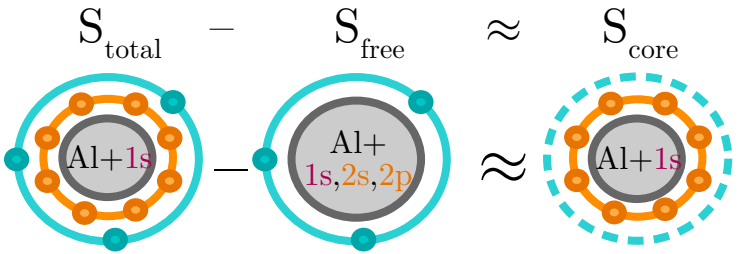
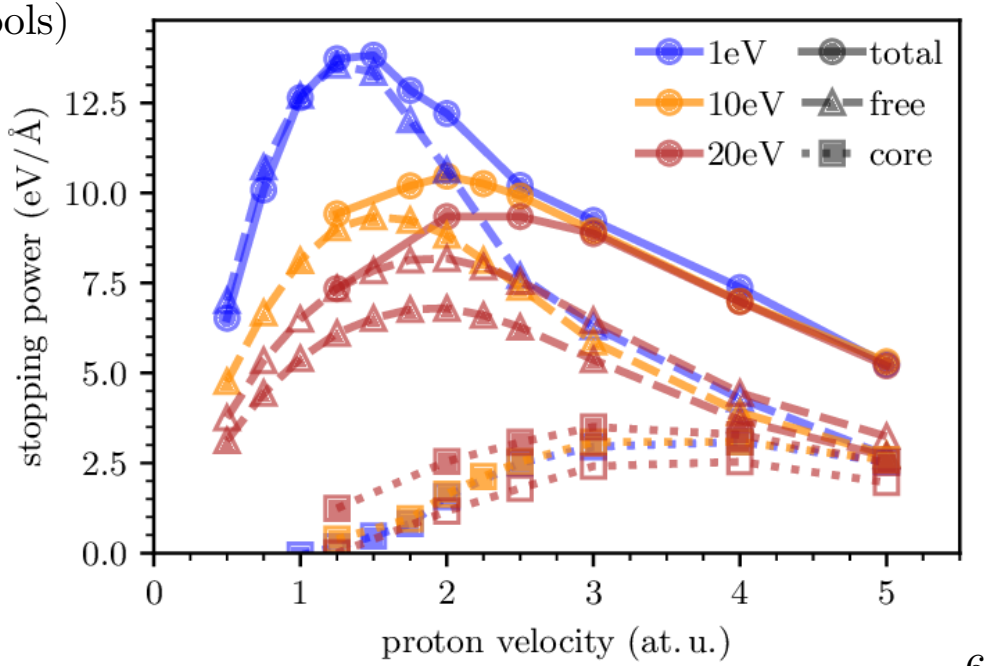
- Thermal depletion of low-energy free states and deeper 2p binding alter 2p → free ΔE
- Thermal depletion of 2p allows 2s → 2p at T_e=20eV

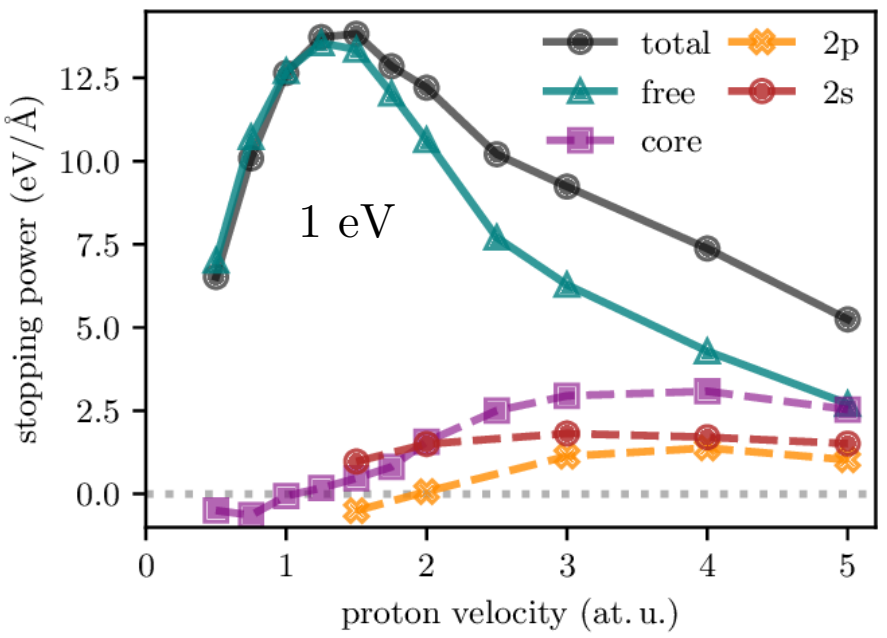
T_e	1eV	10eV	20eV
free e^- per atom	3.00	3.02	3.61
$E_{free} - E_{2p}$ (eV)	65	55	62.5
2p vacancy (%)	0	0.5	9.6

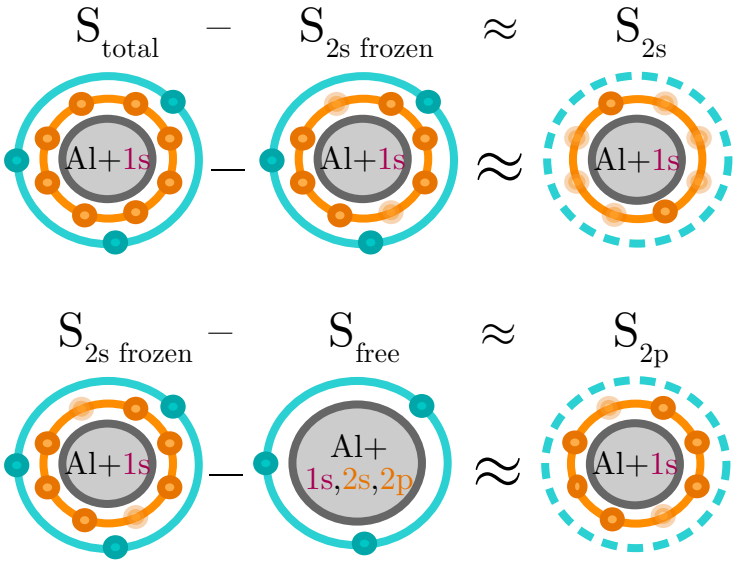




Surprising orbital-resolved core contributions in Al

- Further disentangle by freezing 2s
- 2s contribution surprisingly large: symmetry breaking of 2p excitations
- 2s onset surprisingly low v: inconsistent with threshold models
- Negative values: core excitations can inhibit free response





Finite-size errors beyond plasmonic interpretation

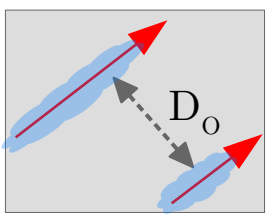
- Trajectory-dependent and non-monotonic finite-size effects inconsistent with plasmonic model

$$S(v) = \frac{2Z^2}{\pi v^2} \int_{\cancel{0}}^{\infty} \frac{dk}{k} \int_0^{kv} d\omega\, \omega\, \mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right]$$

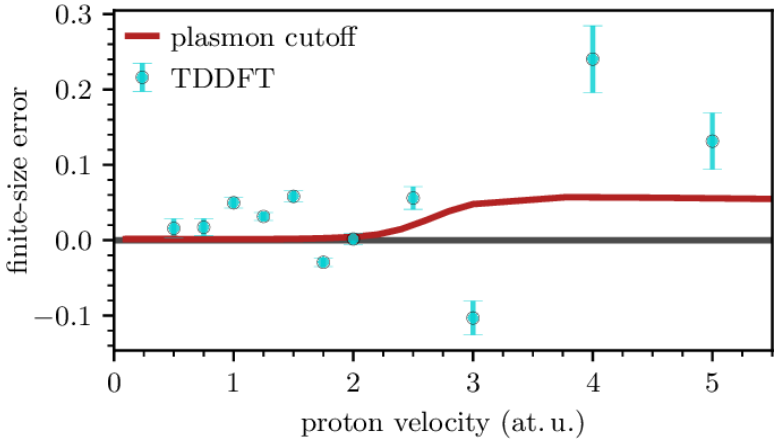
$$k_{\mathrm{cut}} = 2\pi/L$$

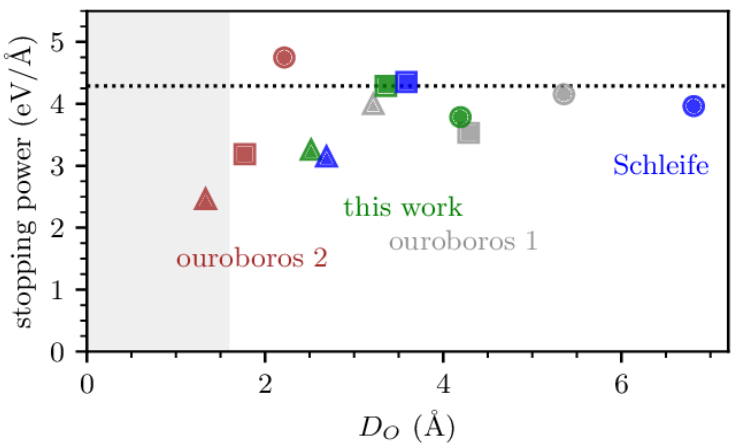
see Hentschel et al., arXiv:2301.09700

- Attribute to "ouroboros" effects
- Propose converging with increasing D_O



arXiv preprint coming soon!





Schleife et al., PRB 91 (2015); Kononov et al., in prep. (2023)

Summary and Outlook

- Simplified models incorrectly describe onset of core contributions to stopping power
- Isolating orbital-resolved contributions through pseudization in TDDFT
- New interpretation of finite-size errors arising from fictitious interaction with excitations
- In C, thermal vacancies in free electron states enhance 1s contribution
- In Al, competing effects modify 2s+2p contributions at high T_e
- Planning detailed investigation with cheaper systems, e.g., Li and Na

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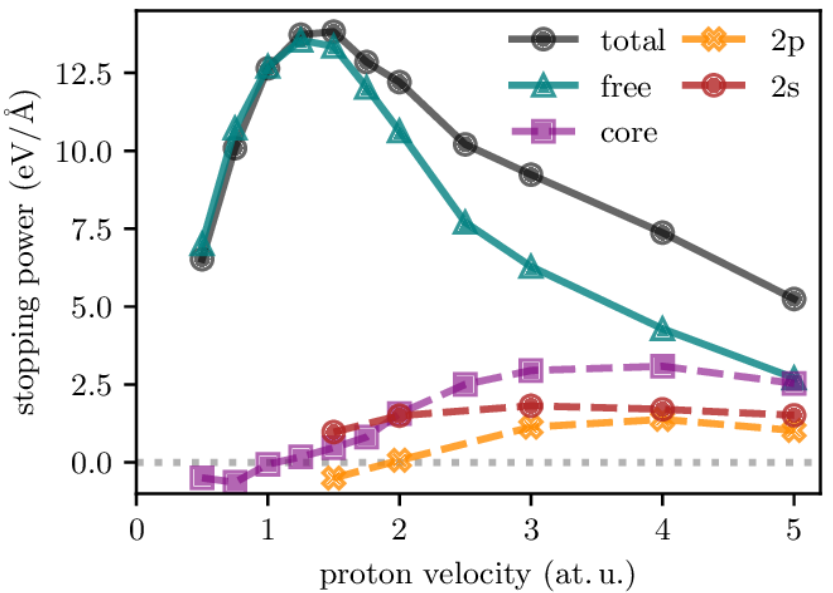

Partitioned finite-size effects allow cost reductions

- Finite-size effects depend on trajectory!
- Core contribution insensitive to supercell size
  - can use smaller supercell, reduce cost
- Behavior in free contribution inconsistent with plasmonic interpretation of finite-size effects

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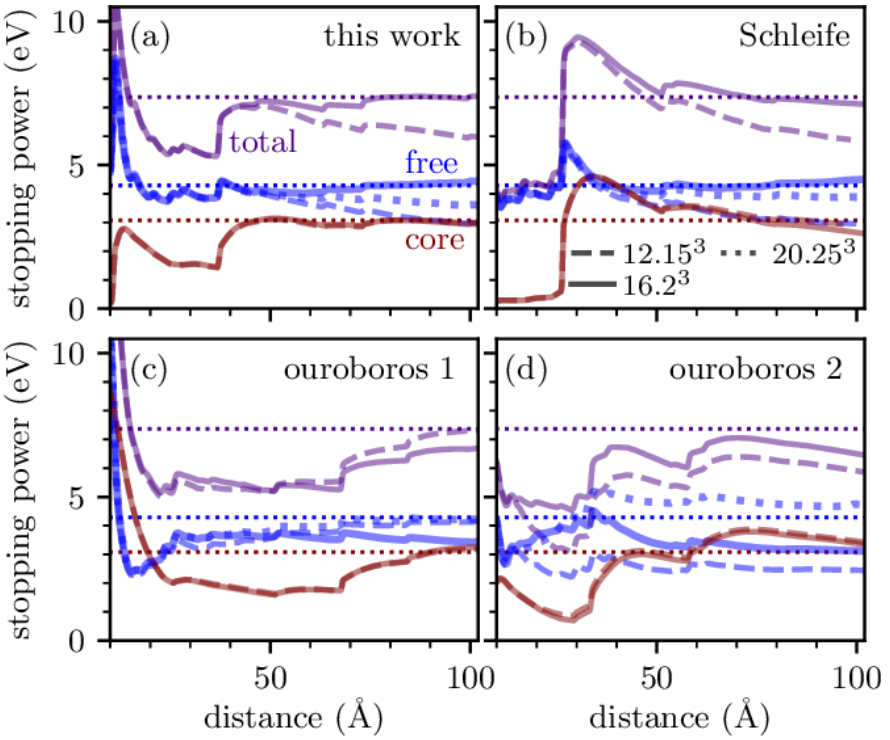

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