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# First-principles insights for high-resolution ion microscopy of graphene

Presented by: Alexandra Olmstead[1]

Coauthors: Alina Kononov[1], Andrew Baczewski[1], Andre Schleife[2]

Affiliations: [1]Sandia National Laboratories and [2]University of Illinois at Urbana-Champaign

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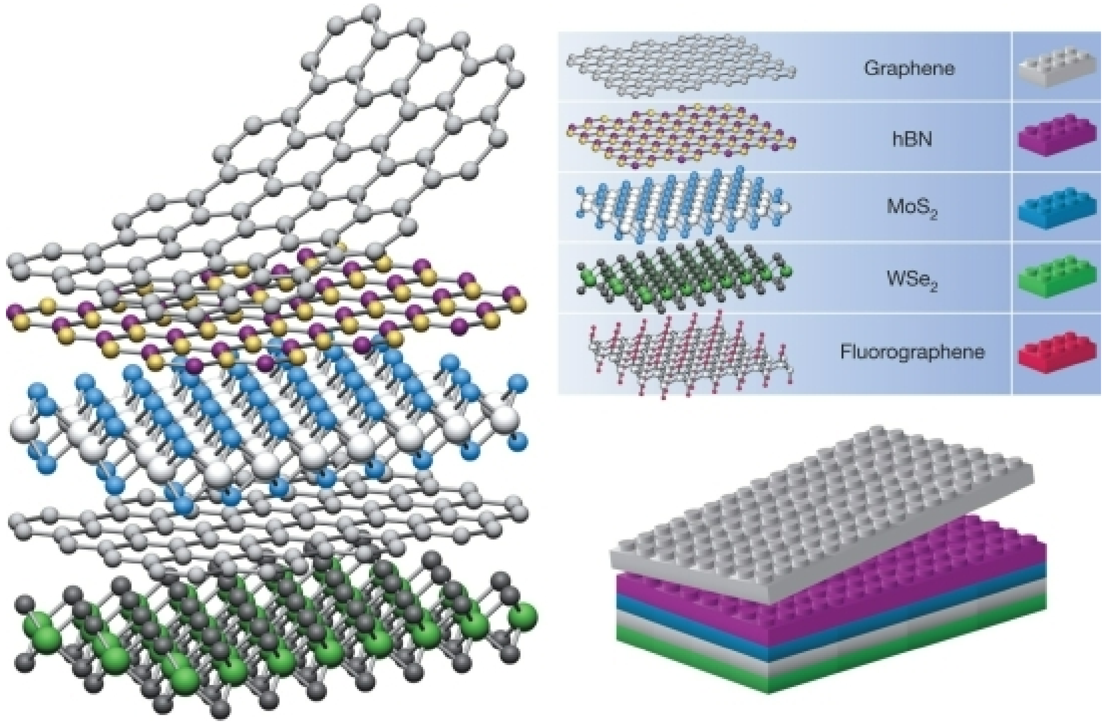
# Motivation

- High-resolution, nondestructive imaging is important for 2D materials and their applications in emerging electronic devices
- Other simulations to determine damage mechanisms usually don't account for the role of electron excitation in damage processes
- Recent studies predict enhanced energy deposition in 2D compared to bulk materials
- Energy deposition rates influence the ion beam's ability to damage the material
- To determine whether deposited energy leads to defects, charge dynamics and atomic forces in the material are analyzed




A. Geim and I. Grigorieva, Nature 2013

arXiv:2201.05207

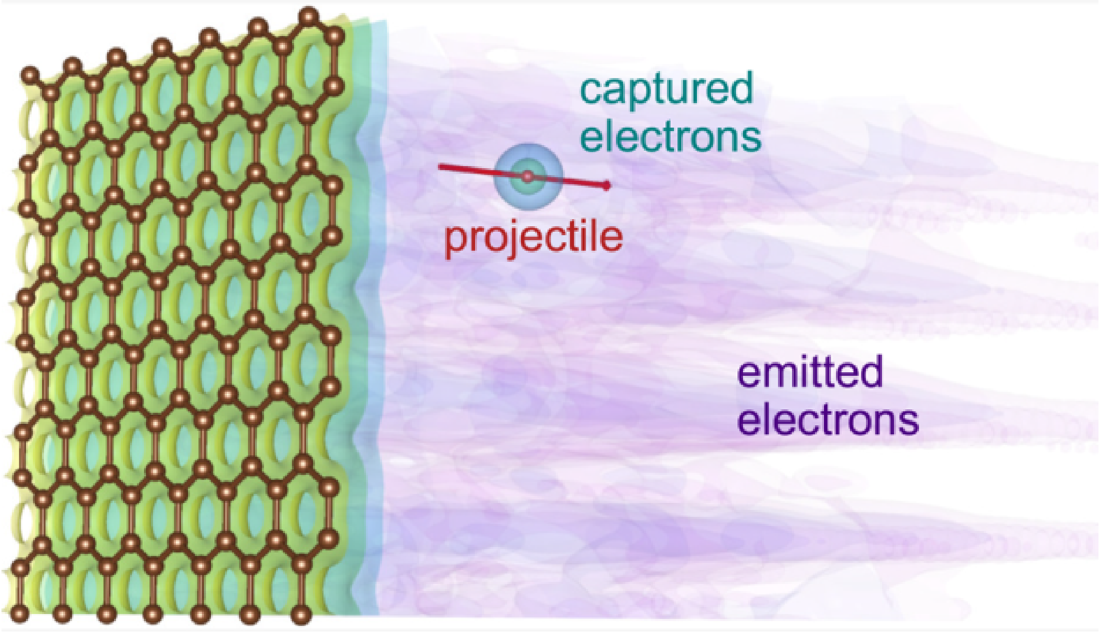
# Computational Methods

- Real-time time-dependent density functional theory (TDDFT) was used to perform the first-principles simulations of 10-200 keV light ions in free-standing graphene

- TDDFT calculations performed using qbox/qb@ll code

- Proton and $He^{2+}$ projectiles inserted and approached the graphene sheet at constant velocity

- At a distance of 10.5 a0 from the carbon atoms, the electron density was integrated in the entrance and exit sides of the vacuum

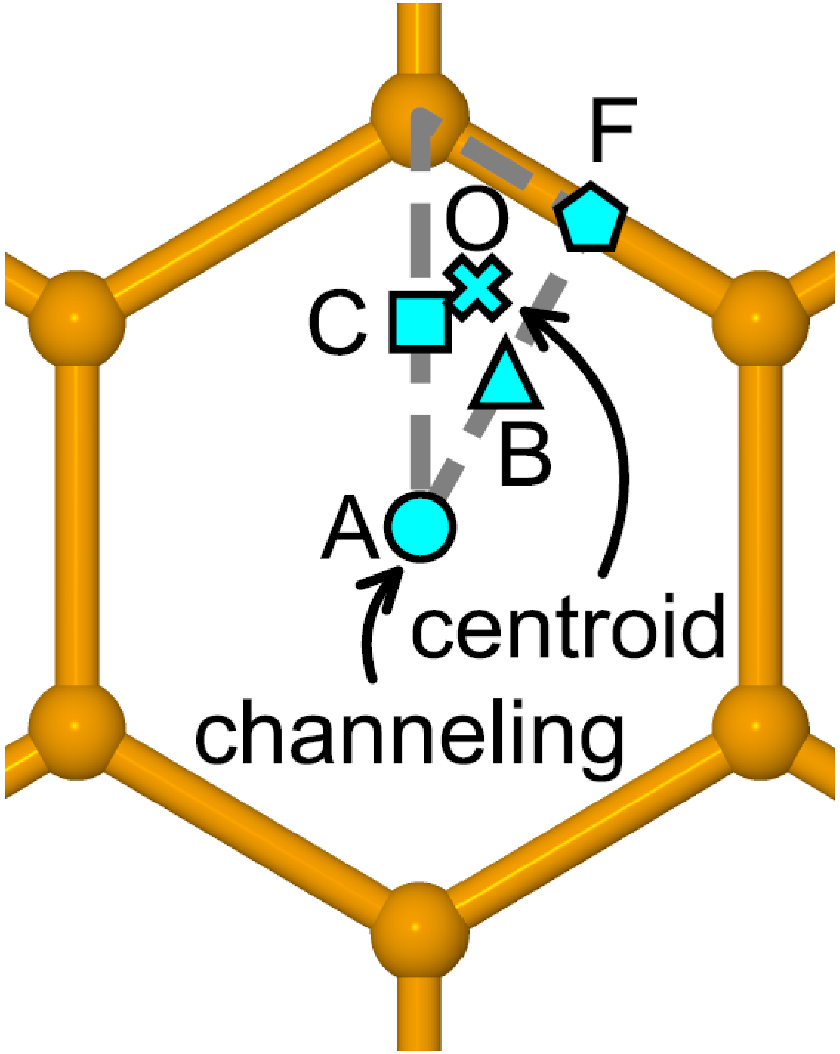



A. Kononov and A. Schleife
Nano Letters 2021

For more details see: arXiv:2201.05207

# Impact Points

- Three (OFA) of the five impact points are sufficient to model the microscopy
- Emitted electron yields from entrance and exit sides were extracted
- Distribution of emitted electrons is larger near the C-C bonds
- The charge distribution for the emitted electrons is linearly interpolated between the impact points

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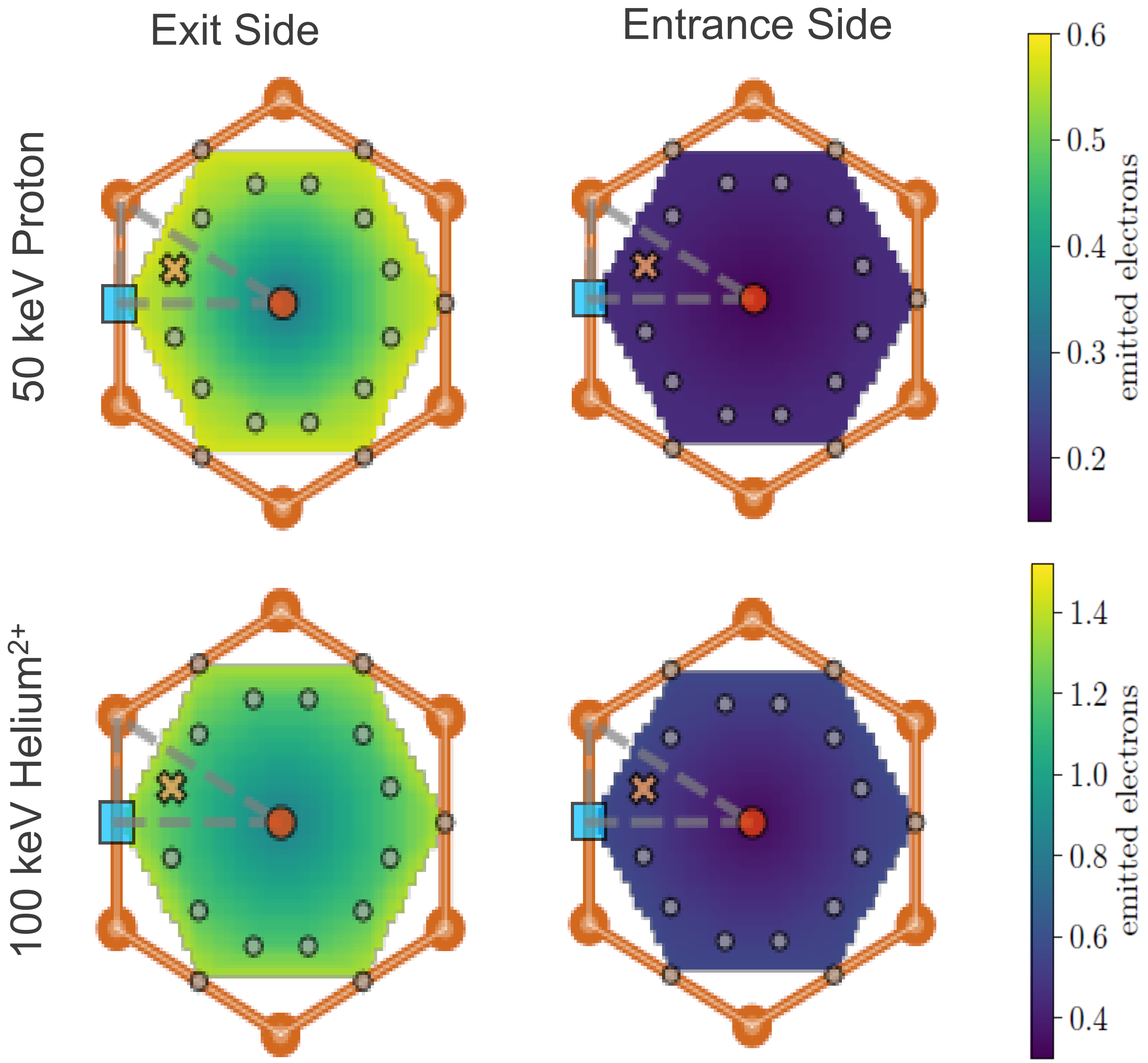
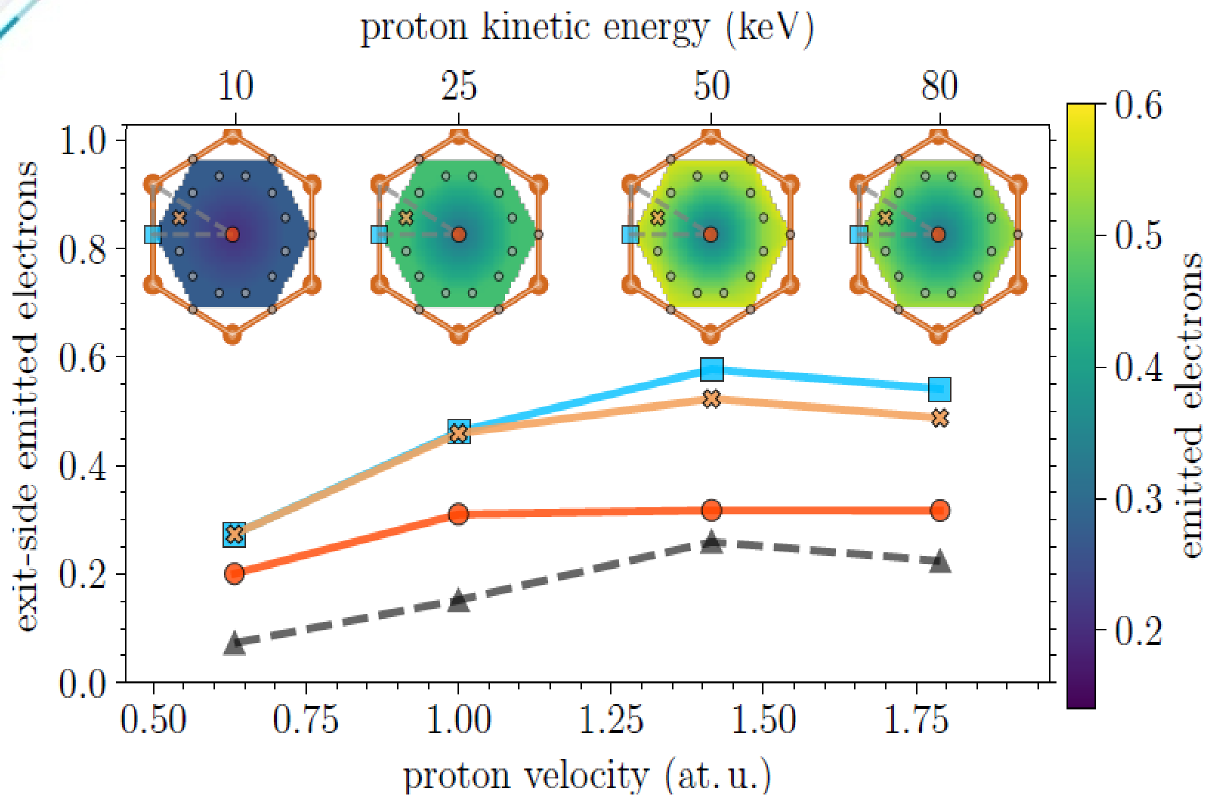
# Microscopy Images

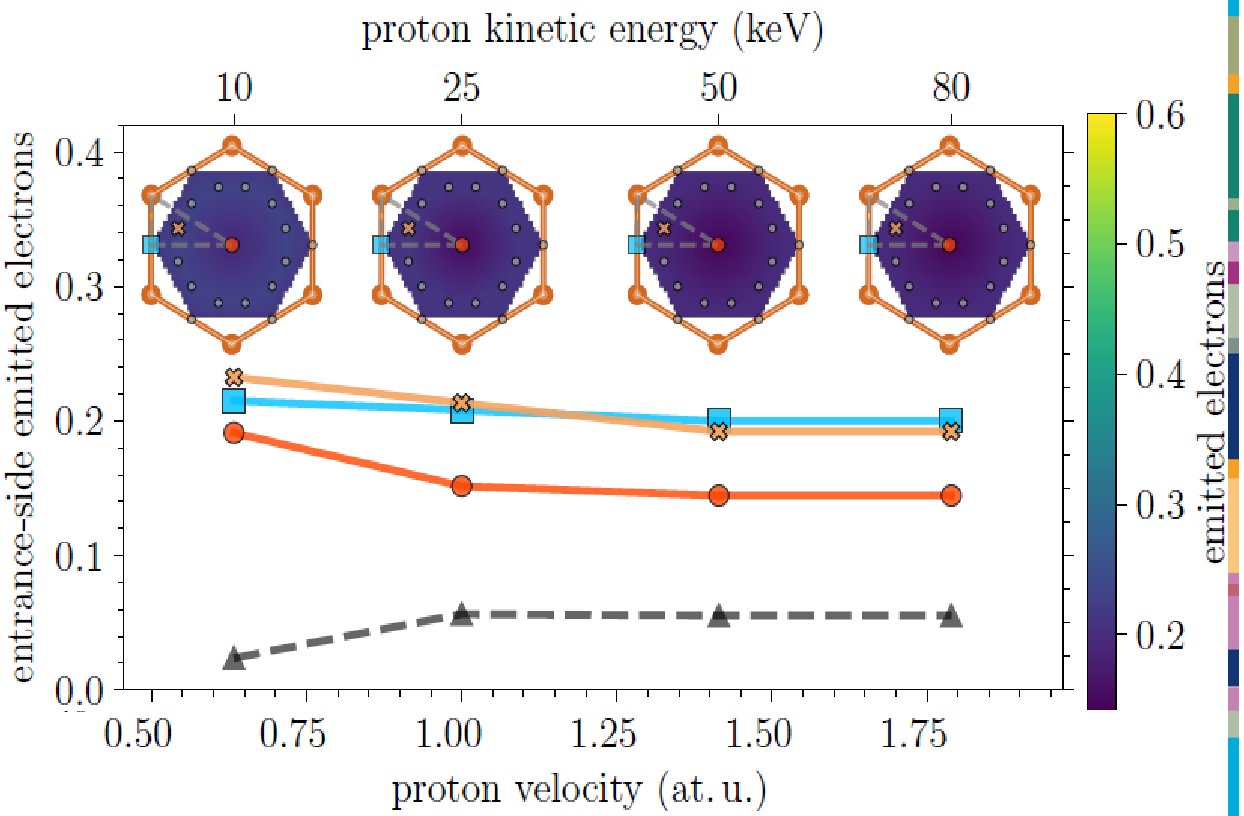
- Emitted exit-side electrons produce higher contrast than entrance-side in both protons and $He^{2+}$ ions
- 50 keV maximizes exit-side electron emission and contrast for protons
- 100 keV maximizes exit-side contrast for $He^{2+}$




arXiv:2201.05207

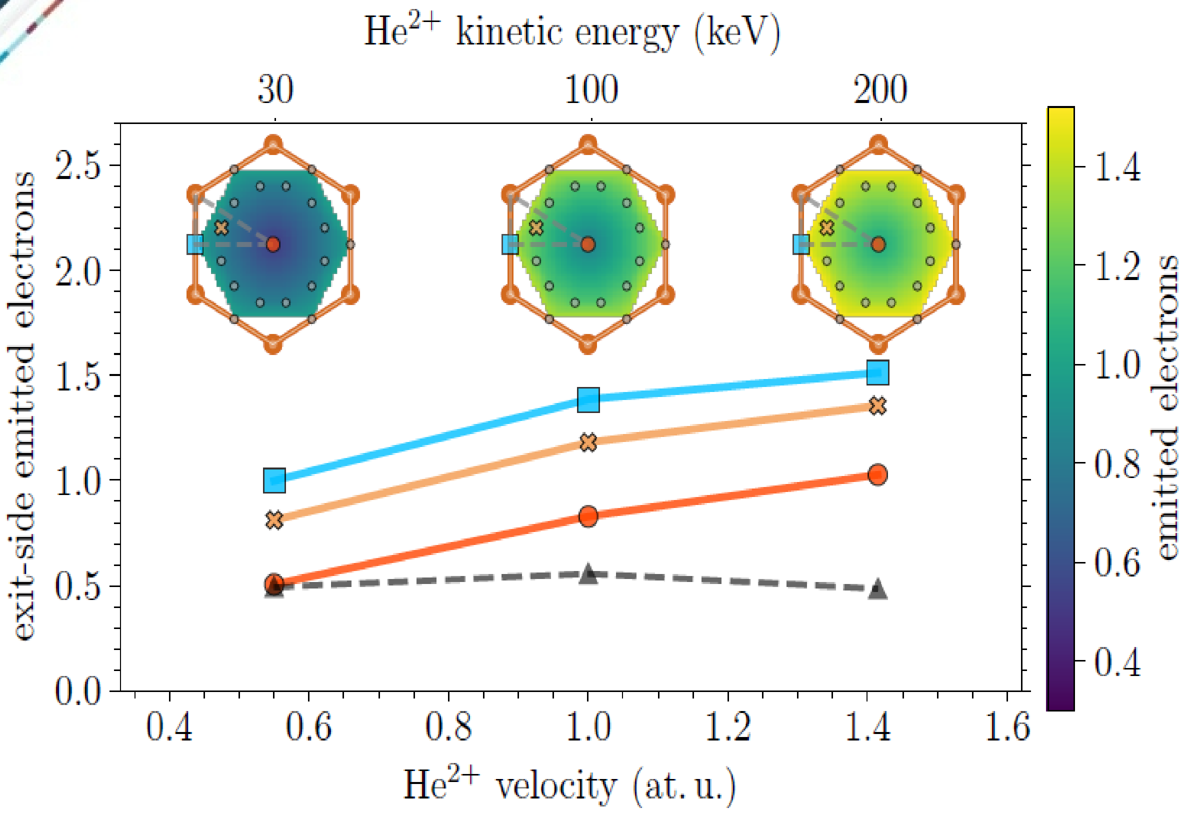
# Proton Velocity Dependence

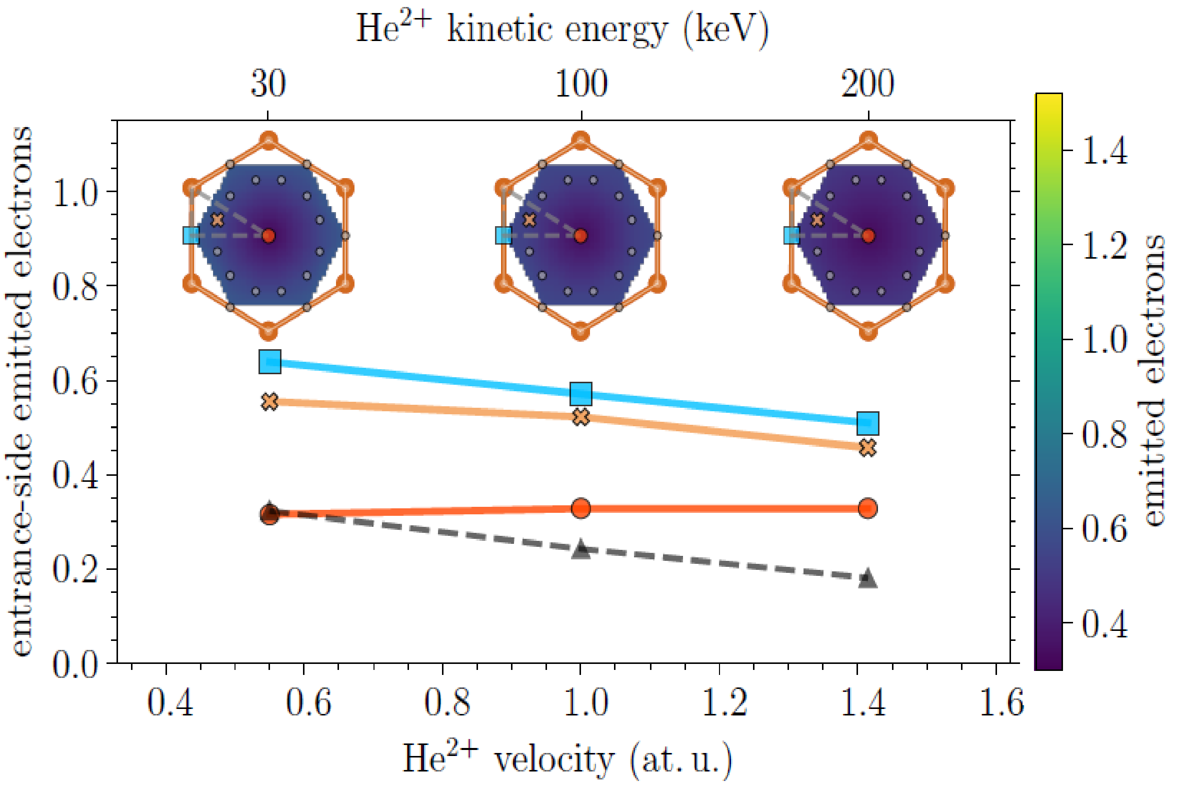



- Contrast metric (black) shows exit-side contrast maximized at 50 keV
- Exit-side produces higher contrast and overall stronger emissions
- Stronger exit-side emissions allow for a lower ion dose

# Helium Velocity Dependence



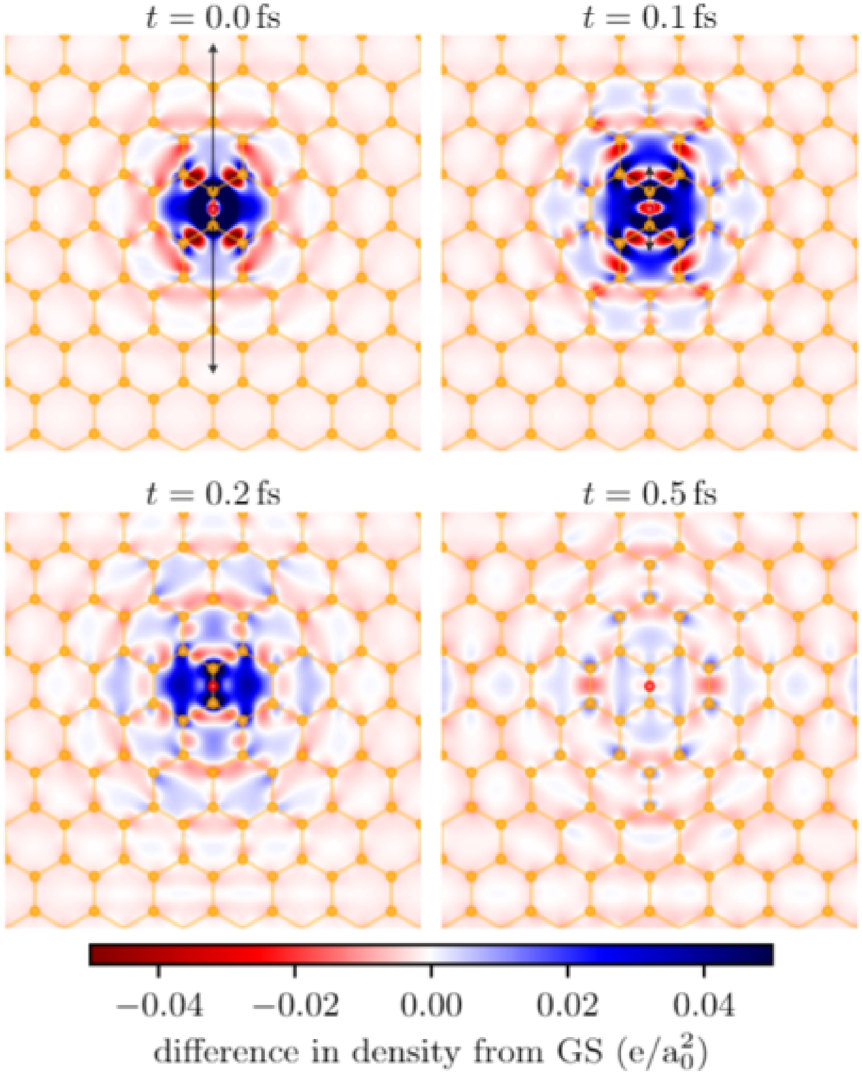




- Contrast metric (black) shows maximized exit contrast at 100 keV
- Entrance-side contrast maximized at lower energies
- Image contrast optimized by higher beam energy for exit-side than entrance-side emission

# Damage Indicators: charge equilibration

- Successful imaging techniques must avoid damaging the atomic structure
- Impacts along the C-C bonds are most likely to cause damage
- The fs-scale rate of equilibration of the ion's induced charge suggests these impacts are unlikely to damage the atomic structure
- Equilibration times are 1.3 times longer for a $He^{2+}$ impact than a proton impact

Charge distribution after impact from $He\ 2^{+}$

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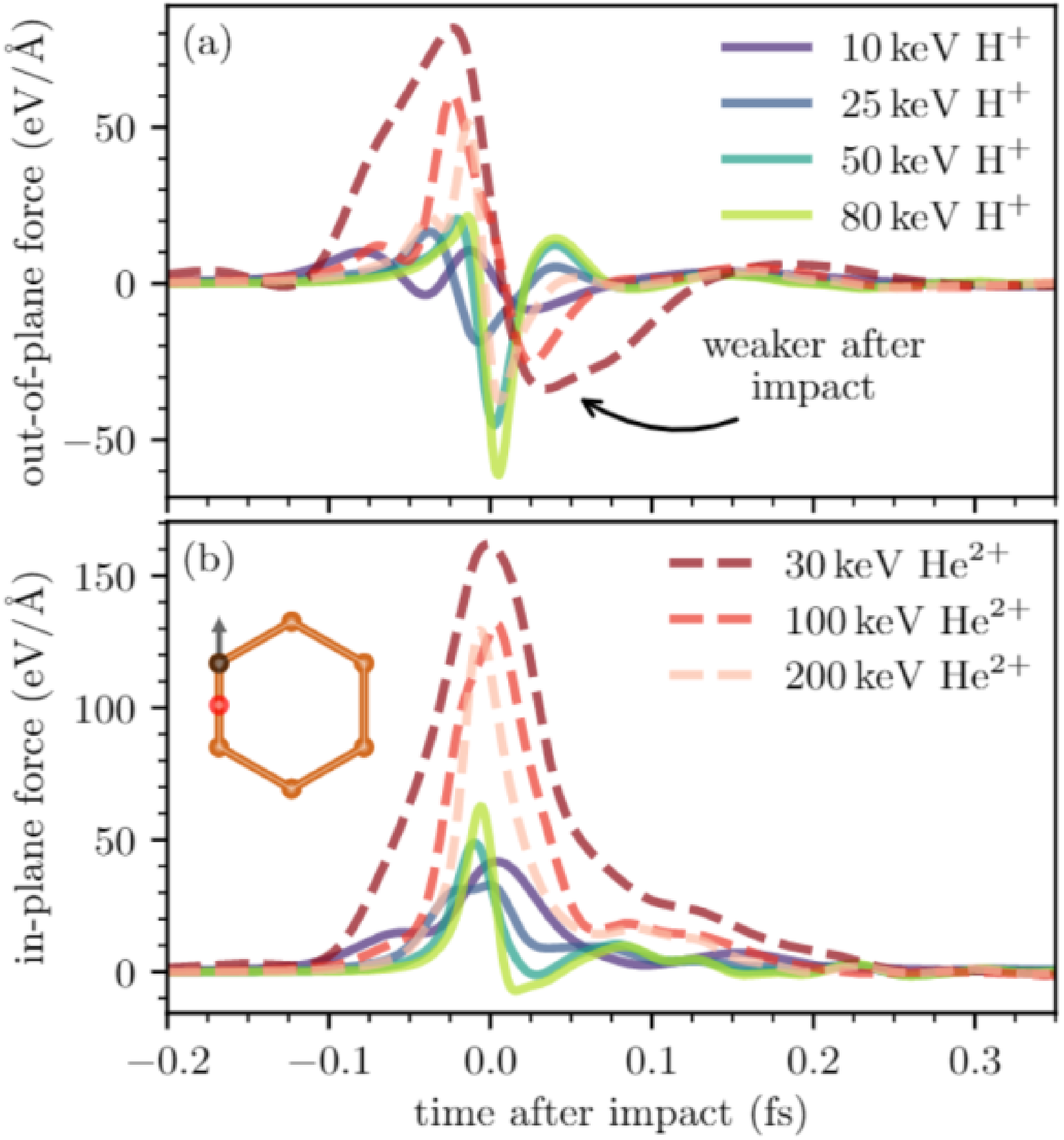
# Atomic forces results

- After an ion impacts the midpoint of a C-C bond, the in-plane and out-of-plane forces experienced by the nearest carbon atom are shown
- Within 0.35 fs of an ion's impact, the forces acting on the carbon atoms decay
- Kinetic energy transfer is over ten times smaller than defect formation energies
- Electronic excitations caused by single impacts of light ions are unlikely to cause defects

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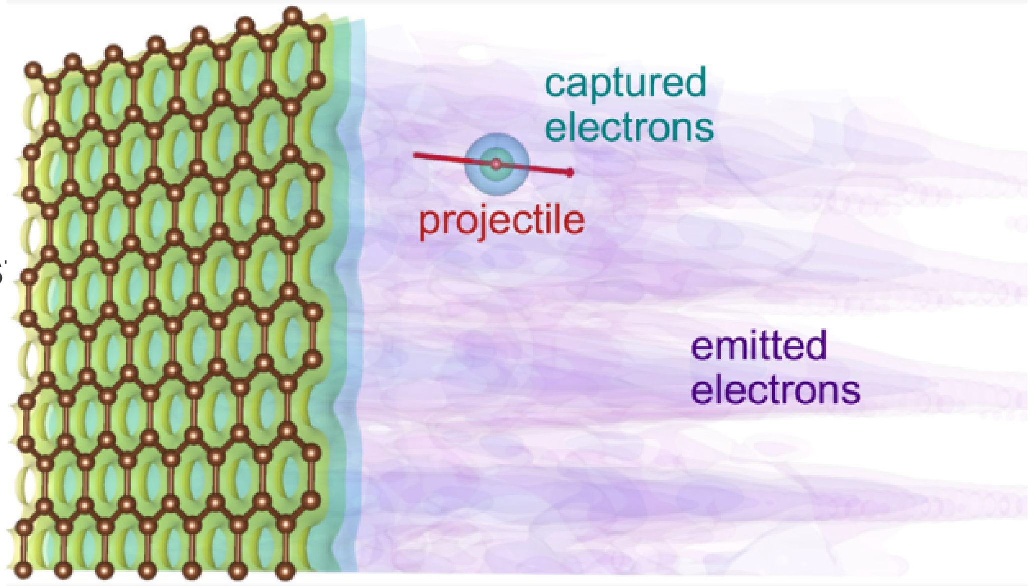
# Conclusions

- This theoretical work predicts that exit-side electron emission produces higher contrast images of graphene
- Higher beam energy is needed to optimize image contrast for exit side than what is needed to optimize image contrast for entrance side
- Electronic excitations caused by single impacts of light ions are unlikely to cause defects

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