

Tracking hot electrons in nanostructures is key to ultrafast optics and photochemistry

WHY IT MATTERS

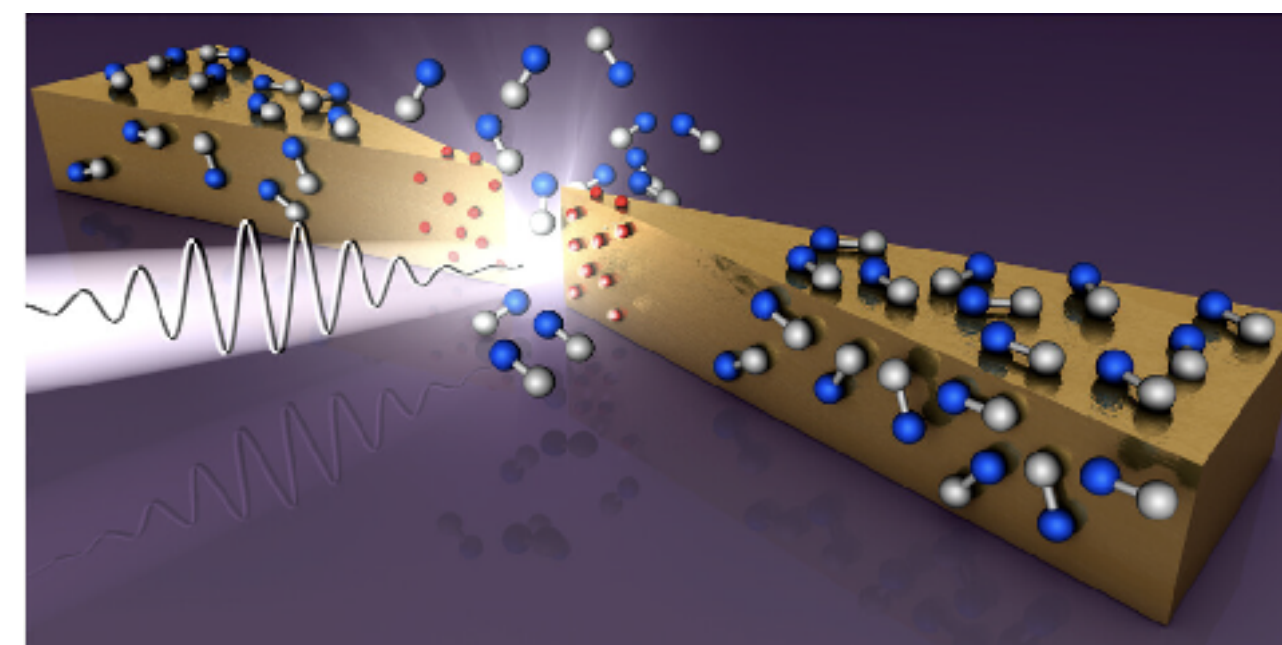
Hot-electron energy, direction, and lifetime govern nanoscale light-matter interactions and drive ultrafast photochemistry.

THE LIMITATION

Standard electromagnetic simulations do not capture **carrier transport and relaxation**.

OUR GOAL

Bridge optical fields and **hot-electron dynamics** to predict measurable quantities.

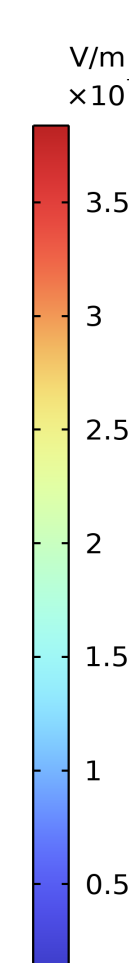
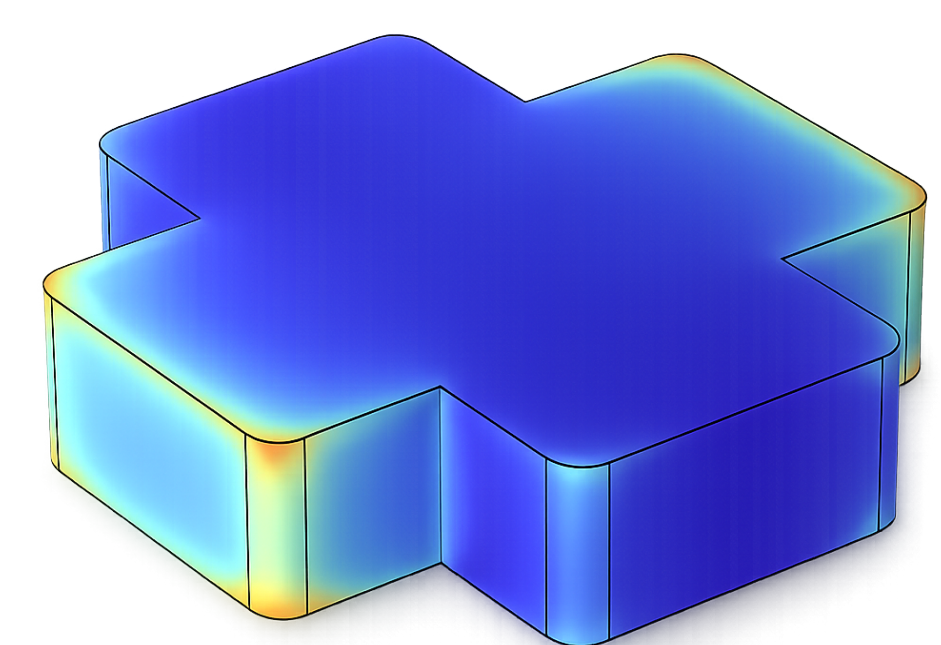


Plasmon-induced hot-carrier generation and transfer at a metal-molecule interface [1].

Electromagnetic fields as the origin of hot-electron dynamics

Localized electric-field enhancement creates hot spots where optical energy is transferred to the material. These regions define the initial spatial distribution of hot-electron generation.

Electric field volume map [V/m]



The electromagnetic field defines where hot carriers are generated, while the mesh provides the discretization required for spatial sampling of generation events.

$$P \propto |\mathbf{E}(\mathbf{r})|^2 \cdot M$$

M is the measure of the discretization element (volume for tetrahedra, area for surface triangles).

LiveLink advantage

COMSOL-MATLAB coupling via LiveLink provides direct access to fields, geometry, and mesh, enabling seamless transfer of model data into the Monte Carlo workflow.

Any change in geometry, mesh, or excitation is **handled directly in COMSOL** and automatically **propagated through LiveLink**.

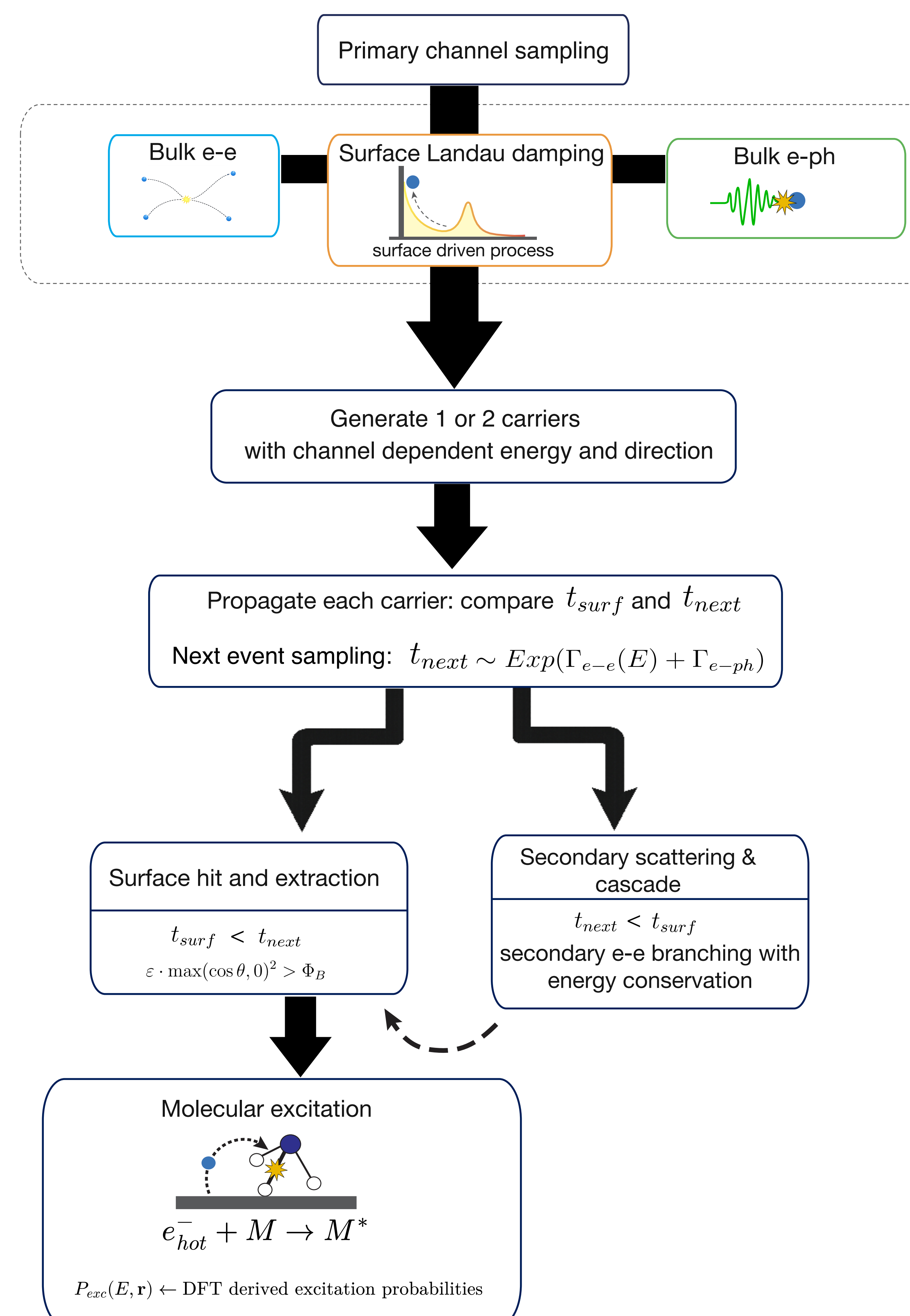
Input to Monte Carlo carrier generation

Research question

How can we design plasmonic nanostructures to control where, when, and with what energy hot carriers reach reactive interfaces?

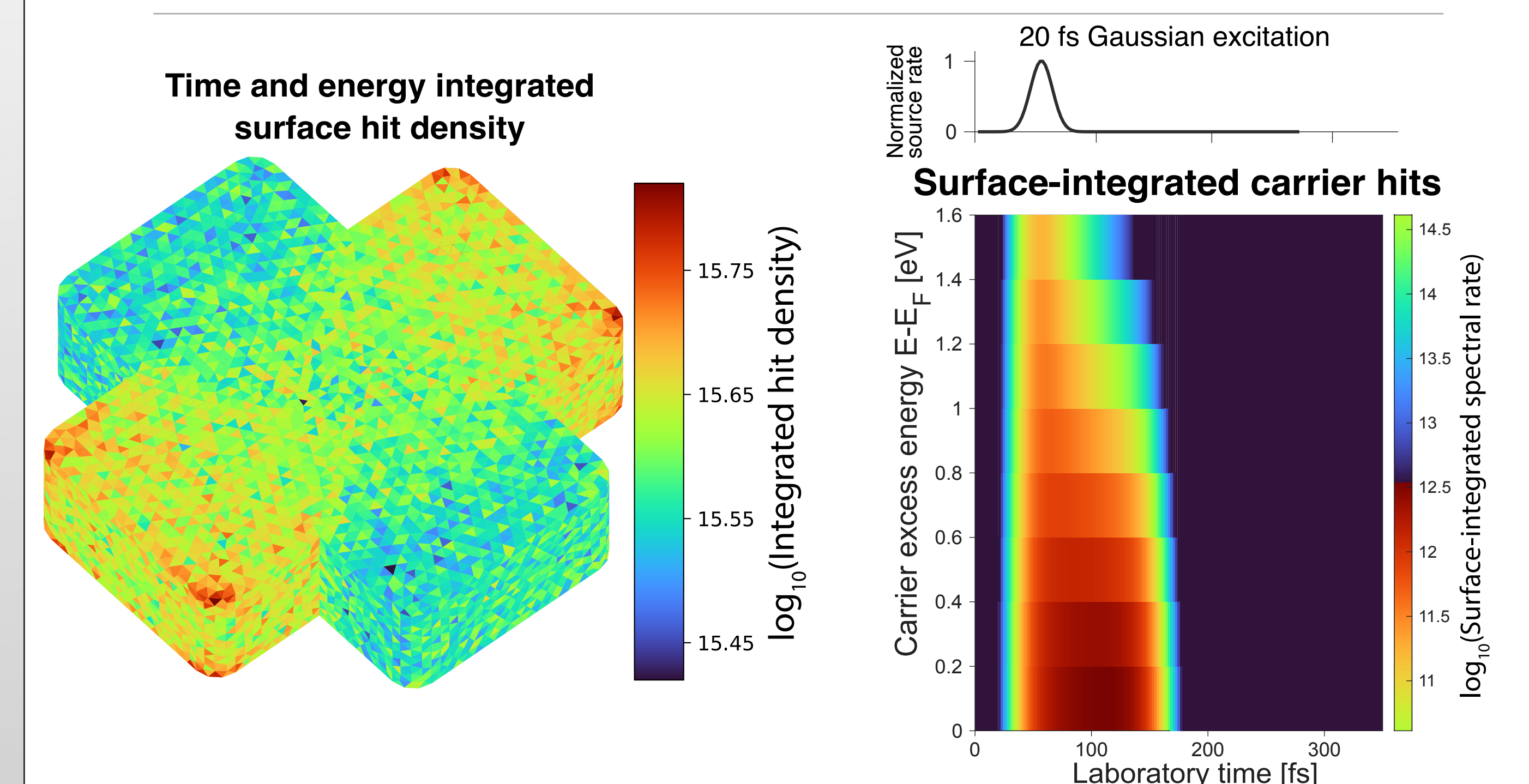
Monte Carlo for hot carrier generation and transport in MATLAB

In the Monte Carlo algorithm, carriers are propagated **step by step** as sketched in the schematics below. Energy and direction distributions are taken from [2].



Space, energy, and time resolved carrier delivery

The Monte Carlo framework resolves when, where, and with what energy hot carriers reach selected interfaces.



Key capability: Impulse-response Monte Carlo kernels enable reconstruction of carrier flux under arbitrary excitation profiles.

Future Research Directions

Towards a predictive multiscale model of hot-carrier-driven chemistry

Full band- and k-resolved transport

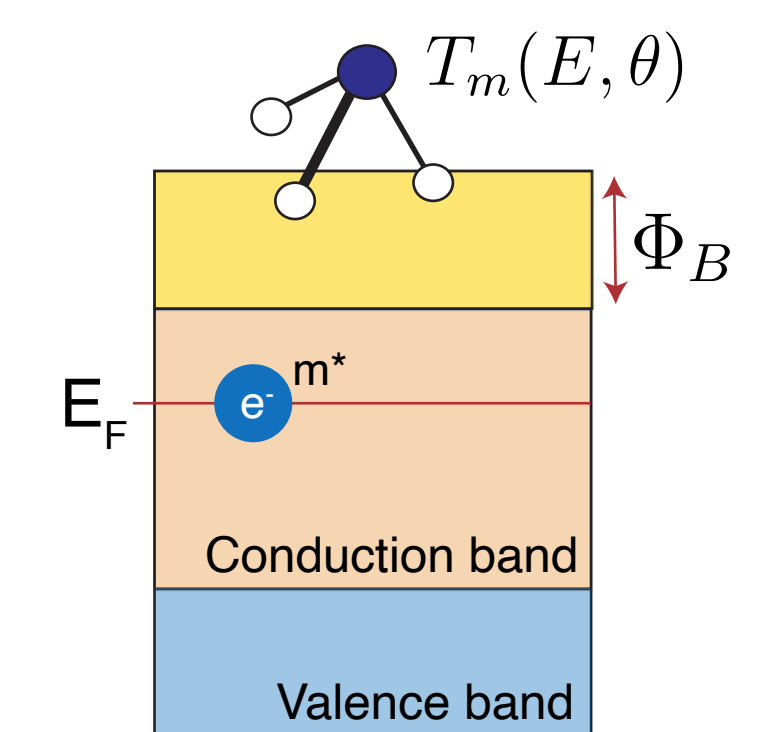
Use $E_{n,k}$, $v_{n,k}$ and state-resolved scattering from electronic-structure calculations.

DFT-informed interfacial chemistry

Use molecule- and interface-specific transition data from first principles.

Quantitative validation

Comparison with **experimental data** and **ab initio models**



Final goal: connect electromagnetic excitation to carrier-resolved interfacial chemistry within a single predictive framework.

References

- [1] Schirato, Andrea, et al. "Quantifying Ultrafast Energy Transfer from Plasmonic Hot Carriers for Pulsed Photocatalysis on Nanostructures." ACS Nano 18, no. 29 (2024).
- [2] Khurgin, Jacob B. "Fundamental Limits of Hot Carrier Injection from Metal in Nanoplasmonics." Nanophotonics 9, no. 2 (2020).

Acknowledgements

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