NON-ADIABATIC ULTRAFAST ELECTRON-ION DYNAMICS NEAR ALUMINUM SURFACES

Allocation: Blue Waters Professor/240 Knh
PI: André Schleife[1]
Co-PI: Alina Kononov

[1]University of Illinois at Urbana-Champaign

EXECUTIVE SUMMARY

Computational physics and materials research greatly benefits from high-performance computing, since modern first-principles approaches provide insight with unprecedented precision. Currently, highly accurate simulations of real-time quantum dynamics are coming within reach, allowing researchers to overcome the Born-Oppenheimer approximation systematically. We explore the feasibility of Ehrenfest molecular dynamics, based on real-time time-dependent density functional theory, to investigate secondary electrons near surfaces of aluminum. We study the dependence of electronic stopping and secondary-electron emission on the velocity of fast projectile ions. From non-adiabatic electron-ion dynamics emerging after the impact, we gain detailed insight into the materials physics on an attosecond (10-18 secs) time scale. These simulations allow us to determine precisely the transition from "surface-like" to "bulk-like" systems. While large simulation cells and short simulation time steps are required, Blue Waters enables these simulations and allows us to study complex quantum dynamics quantitatively.

INTRODUCTION

Fast particles entering a material with kinetic energies on the order of keV or MeV produce effects on various length and time scales. At early stages, deposition of energy into the electronic system of the target is the dominating mechanism. It is mediated by the electron-ion interaction and leads to excitations of the target's electronic system. Details of these processes are not well understood: Existing theoretical models require ad-hoc assumptions and rely on separate theories, e.g. for the charge state of the projectile. Early models suffer from large uncertainties and are not suitable for the computational materials design needed for novel technological applications.

Scenarios of practical interest include fusion reactors, where highly energetic particles interact with the reactor wall. At initial stages, the vast majority of energy is deposited into the electronic system of the host. Near the surface, excited electrons can overcome the work function of the material and escape into the reactor. These secondary electrons detrimentally affect the plasma properties. Another example of paramount importance for technological applications is modern helium microscopy [1]; secondary electrons emitted from the sample are collected and create image contrast. Helium microscopy is an appealing technique, especially for insulating uncoated samples. A better understanding of the dependence of secondary-electron emission on the velocity and impact angle of the projectile is necessary. Accurate first-principles techniques that are capable of describing these effects are, thus, desirable for a large community of researchers.

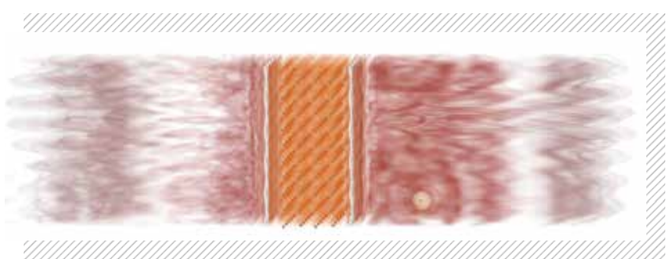

FIGURE 1: Aluminum slab of 200 atoms after being hit by a fast hydrogen projectile. Electrons are localized around the hydrogen projectile and secondary electrons are ejected into the vacuum.

METHODS & RESULTS

In this project we explore Ehrenfest molecular dynamics: we use our new large-scale implementation within the Qbox/Qb@ll code [2], based on real-time time-dependent density functional theory (RT-TDDFT). Nuclei are treated as classical point charges coupled to electrons via Coulomb interaction and electrons are treated quantum-mechanically. The electron-ion interaction is described using non-local pseudo-potentials and the electron-electron interaction is a sum of Hartree and exchange-correlation potential. Kohn-Sham wave functions are expanded into plane waves, and we also use the common adiabatic local-density approximation.

Using this technique we investigated the transition from "surface like" to "bulk like": We varied the number of atoms that constitute an aluminum slab between 100 and 500 (Fig. 1) and computed position-dependent electronic stopping. Figure 2 shows that immediately after entering the material, the projectile (velocity = 1.0 at. u.) experiences larger stopping, which we attribute to the fact that its charge has not equilibrated after entering the slab. Once the projectile traveled about 20 aB, equilibration is observed as the electronic stopping approaches the bulk value [3]. At this point, the projectile attracted some of the host electrons, reducing its initial fully ionized state. Our simulations allow extracting length and time scales of this equilibration, and we can reconstruct the effective charge state of the projectile as it travels through the material.

By fitting the charge-density distribution around the projectile to hydrogen orbitals, we compute the number of electrons around the projectile. For a velocity of v = 1.0 at. u., we found that about 0.3 electrons move with the projectile after it leaves the material. We are analyzing this data for different vacuum lengths, numbers of slab atoms, and projectile velocities to develop a comprehensive picture. We also extract the total number of emitted secondary electrons. These simulations directly provide insight into the underlying physics and allow us to extract quantitative information that can be used to parameterize larger-scale numerical models. Based on our accurate, quantitative data we envision multi-scale computational design of materials under radiation conditions.

WHY BLUE WATERS

First-principles Ehrenfest simulations come with high computational cost and are only possible due to massively parallel implementations of this technique and using high-performance supercomputers with low-latency communication. Thousands of time steps (as short as 0.1 atto-seconds) are needed, and many simulations are required to study vacuum size, numbers of slab atoms, and projectile velocities. Since our code scales very well to hundreds of thousands of cores on Blue Waters, we make efficient use of this machine when running for many hours on hundreds of nodes. Through the Blue Waters team, we have excellent access to visualization and data analysis experts with whom we discussed interpretation and analysis of the vast amount of data (hundreds of megabyte per time step).

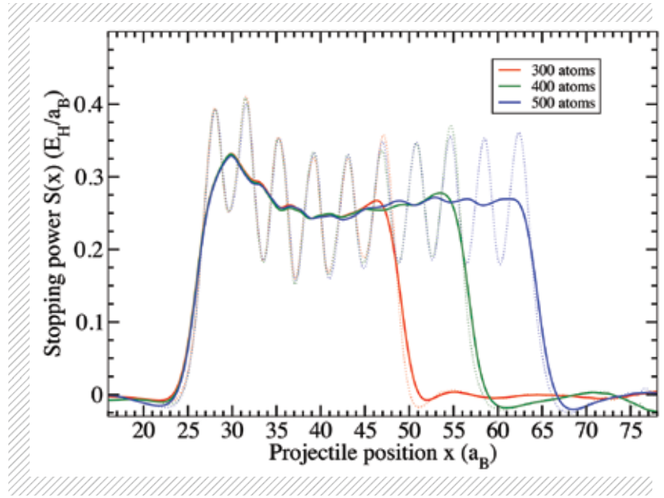

FIGURE 2: Instantaneous electronic stopping of a hydrogen projectile in aluminum slabs of different thickness.

NEXT GENERATION WORK

From our studies, we found that including the effect of excited electrons in first-principles molecular-dynamics simulations is critically important in many cases of practical relevance. Ehrenfest molecular dynamics can be a practical first step towards achieving this. However, the computational cost is high. Using Blue Waters, we can study systems of practical importance. However, we envision that next-generation Track-1 systems will allow us to explore larger length and time scales. They will inevitably be needed to achieve our vision of routinely using quantum-dynamics simulations and of going beyond Ehrenfest molecular dynamics for computational materials design.