

Interactions of Ions and Photons with Atoms, Surfaces, and Molecules

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1. Neutralization of Negative Hydrogen Ions near Metal Surfaces (T. Niederhausen, H. Chakraborty, U. Thumm)

We have applied wave-function propagation techniques to charge-transfer processes in ion-surface interactions, using self-consistent potentials to represent the electron-surface interaction. Apart from contributing to the qualitative understanding of the interaction mechanism through computer animations, this project has led to the quantitative assessment of charge transfer and to the characterization of surface resonances in terms of level shifts and decay widths as functions of the ion-surface distance. Using the split-operator Crank-Nicholson propagation technique in conjunction with carefully adjusted absorbing potentials near the numerical grid edges, we have performed 3D propagation calculations for the decay of negative hydrogen ions near metal surfaces.

In particular, we have completed calculations for the neutralization of negative hydrogen ions near two copper surfaces of different symmetries, Cu(100) and Cu(111). In these (adiabatic) calculations, we kept the ion-surface distance D fixed and propagated the electronic wave function. To start with, we used a 1D propagation scheme where the active electron is restricted to move along the surface normal. To model the electron-surface interaction, we used suitable parametrizations of self-consistent LDA potentials. For numerical convenience, we performed these calculations for 100 monolayer thick Cu films and extended the numerical grid 200 atomic units on the vacuum side. In order to test the accuracy of our Cu potentials, we diagonalized the effective one-electron Hamiltonian and were able to reproduce the experimentally known energies of the upper and lower limits of the surface projected L-band gap, the surface-state energy, and the energies of the few lowest image states.

We obtained resonance positions and widths by first propagating the initial negative hydrogen state (one-electron approximation) for fixed D . Next, we calculated the projected density of states (PDOS) by Fast Fourier Transformation of the autocorrelation function, i.e., the overlap of the electronic wave function at time t with the initial electronic state. We repeated this calculation for various D and identified peaks in the PDOS as resonance states that asymptotically (for large D) correspond to the affinity level of the negative hydrogen ion, to image states, surface states, and bulk states of the 100 layer thick film. The lifetimes of those resonance states were extracted from the line widths of the corresponding peaks in the PDOS. We compared our results for the Cu(100) and Cu(111) surfaces with results for a structureless Cu-jellium potential.

By plotting the level shifts and widths for Cu(100), Cu(111), and Cu(jellium) as a function of D , we noticed several avoided crossings between image-state, surface-state and affinity-level resonances (Fig.1). Interestingly, the detailed behaviour of both level widths and shifts differs strongly for decay into Cu(100) and Cu(111). Qualitatively, we

understand this difference in terms of the different energetic location of image states, surface state, and surface band gap.

We plan to extend this 1D adiabatic propagation calculations to two and three dimension. We will try to quantify to what degree the inclusion of the active electron's motion in the surface plane affects resonance widths, and shifts. Next, we intend to include the motion of the projectile in order to provide neutralization probabilities for the scattering of negative ions near surfaces and to understand the interplay of elementary processes at different time scales (decay into surface states, decay of surface states, effects of the electron's motion parallel to the surface, the role of lifetimes and interaction times).

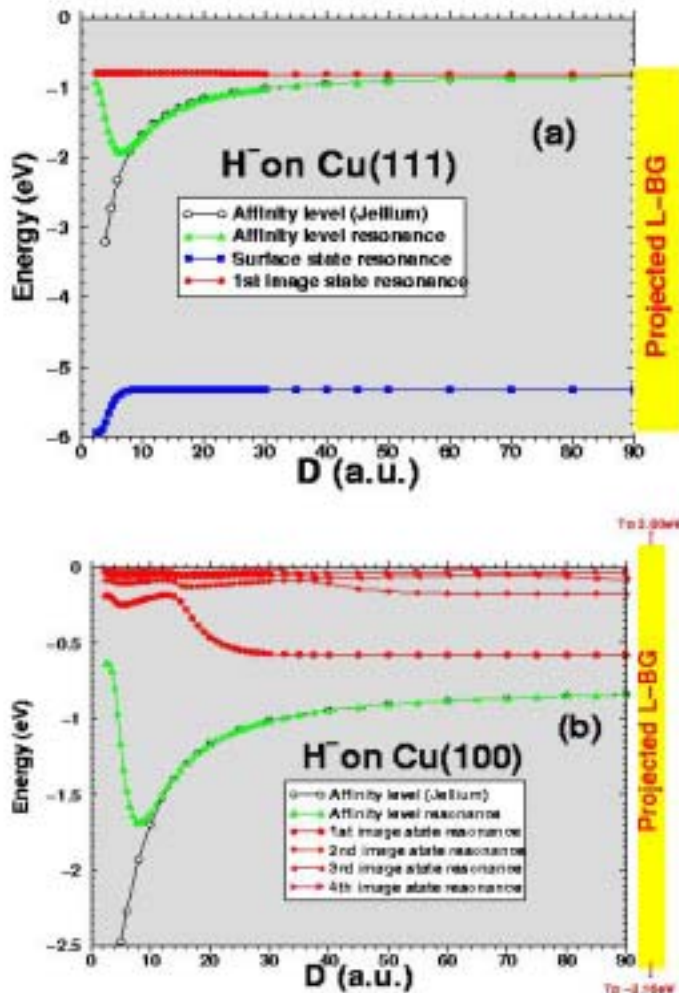


Fig.1: Affinity level, surface state, and image state peak position as a function of the ion-surface distance D . Results obtained from the projected density of states for a Cu(111) surface (a) and a Cu(100) surface (b).

2. Laser-Matter Interactions (M. Alcantara Ortigoza, B. Feuerstein, U. Thumm).

We have investigated the interaction of 25 fs, 0.2 PW/cm², 780 nm pulses with H₂⁺ and D₂⁺ molecular ions within a reduced-dimensionality model that represents both the nuclear and electronic motion by one degree of freedom. We carefully adjusted the adiabatic molecular electronic potential by introducing a „soft-core function“ $a(R)$ in the electron-nucleus interaction potentials $1/(x \pm R/2 + a(R))$ that depends on the internuclear distance R instead of the commonly used fixed soft-core constant a . We obtained molecular model potentials that reproduce accurate three-dimensional results for the known number of 19 vibrational states in the electronic ground state and for the dipole oscillator strength.

We solved the time-dependent Schrödinger equation on a two-dimensional numerical grid and designed a simple, but as far as we know new, method to calculate the flux of emitted electrons and protons by means of „virtual detectors“ for electrons and protons. These detectors are placed outside the excursion range of the electron and at a distance R where the amplitudes of bound vibrational states have become irrelevant.

Our results reproduce the main features of measured kinetic-energy release spectra, support the „charge-resonance enhanced“ ionization mechanism, and allow us to clearly distinguish between molecular dissociation (MD) into field-dressed final channels and fast, ionization-induced Coulomb explosion (CE). Both MD and CE appear as distinct peaks in the kinetic energy release spectra. We find that MD dominates for molecular ions that are prepared in the two lowest vibrational states only, while CE becomes increasingly dominating for higher vibrational states (Fig.2).

For two short laser pulses of variable delay, we started to resolve in time the interplay between MD and CE. We intend to further investigate the pump-probe dynamics for two short pulses (Fig.3). We will explore the possibility of adding a further dimension to the electronic motion. Motivated by new experiments in the Macdonald Laboratory, we have stated to investigate the ionization of model atoms under the combined influence of a few-cycle intense Laser pulse (10^{14} to 10^{15} W/cm²) and a significantly longer and less intense pulse (about 10^{12} W/cm²). The two pulses may or may not be phase coherent.

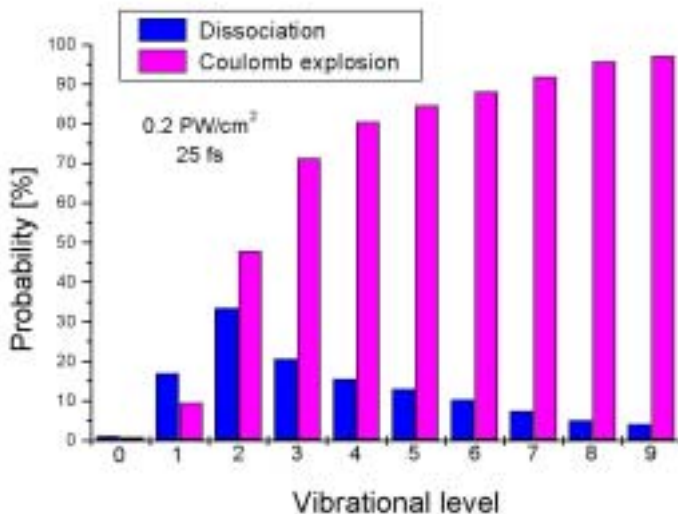


Fig.2: Dissociation and Coulomb explosion fractions for H₂⁺ and 25fs, 0.2 PW/cm² Laser pulses as a function of the initial vibrational state of the molecular ion.

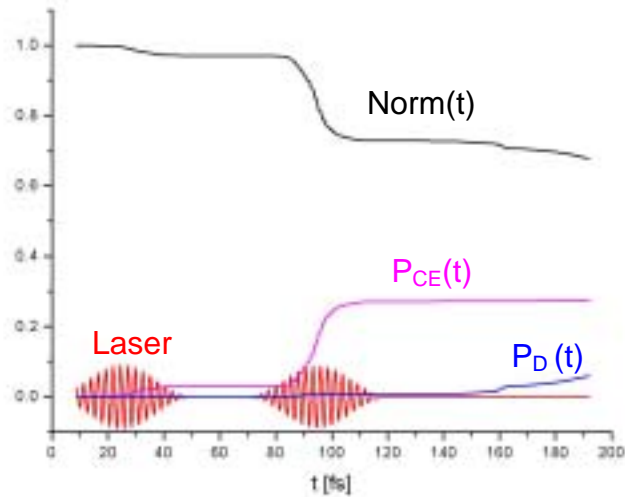
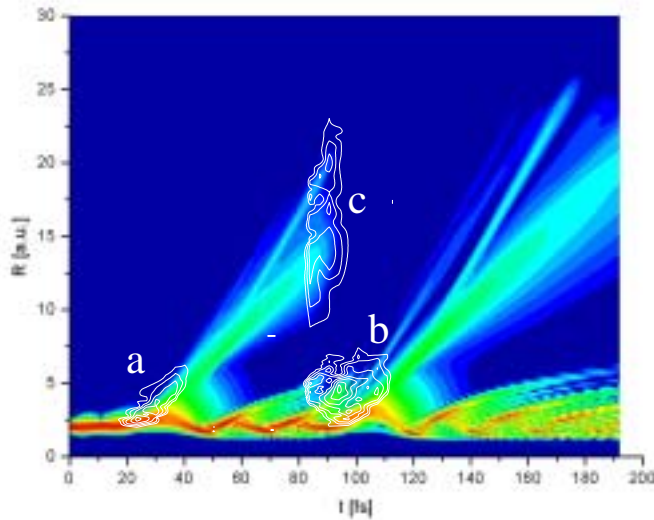
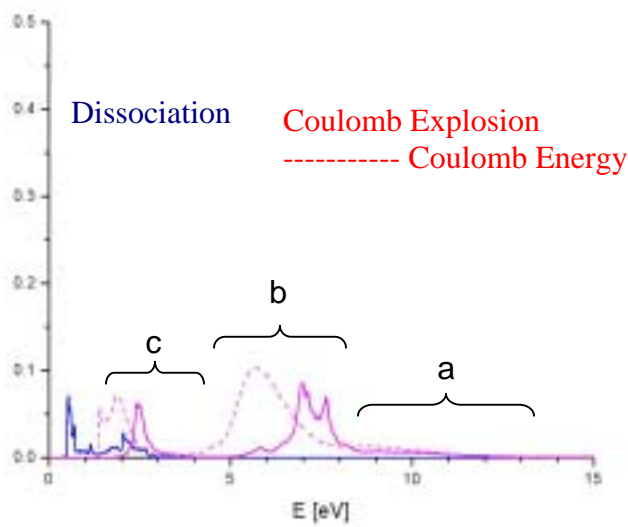


Fig.3: Pump-probe results for D_2^+ molecular ions, initially in the vibrational ground state ($v=0$), and two 25fs, $0.3PW/cm^2$ pulses with a delay of 70fs.

Top: Norm of the molecular wave function on the numerical grid, laser pulses, and probabilities for Coulomb explosion (P_{CE}) and dissociation (P_D) versus time.



Middle: Probability density of the molecular wave function, integrated over the electronic coordinate x . The jets correspond to dissociation. The contour lines represent the rate of Coulomb explosion (a,b) and dissociation followed by Coulomb explosion (c).



Bottom: Kinetic energy release spectrum corresponding to the middle graph. The dashed line shows Coulomb explosion results where the initial kinetic energy of the nuclei is neglected.