# Structure and Dynamics of Atoms, Ions, Molecules and Surfaces: Atomic Physics with Ion Beams, Lasers and Synchrotron Radiation

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## 1. Laser-molecule interactions (with T. Niederhausen and B. Feuerstein)

**Project scope:** We seek to develop numerical and analytical tools to efficiently predict the effects of a strong laser field on the bound and free electronic and nuclear dynamics in small molecules [1] and in laser-assisted collisions [2].
**Recent progress:** We continued our investigation of the dissociation and ionization of $H_2^+$ in short intense laser pulses by applying wave-packet propagation methods. We have started to extend our wave-packet propagation calculation to the single and double ionization of neutral $H_2$. As a complementary approach, we are developing a close-coupling method that coherently includes both ionization steps.
**Example:** Vibrational Wave-Packet Revivals in $D_2^+$. Figure 1 (a) illustrates our reduced-dimensionality calculations [1] for the nuclear wave packet dynamics in $D_2^+$ following ionization of $D_2$ (v = 0) in a 5 fs, $10^{15}$ W/cm$^2$ pulse. After a few optical cycles, the wave packet collapses due to the dephasing of its stationary vibrational state components. The autocorrelation function shows quarter and half revivals of the wave packet 100 and 200 optical cycles, respectively, after the wave packet has been launched (Fig.1 (b)). A second laser pulse, short and strong enough to ensure instantaneous and complete ionization of $D_2^+$, can probe the time evolution of the wave packet. Pump-probe

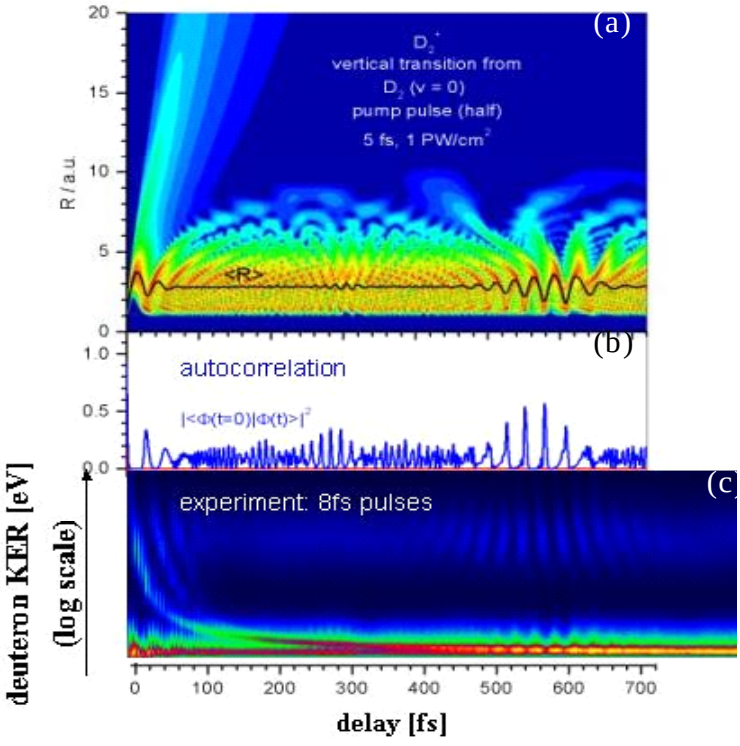


**Fig. 1.** Coherent motion of the nuclear vibrational wave packet in and dissociation of $D_2^+$.
**(a)** Probability density and expectation value <R> of the internuclear distance.
**(b)** Autocorrelation function.
**(c)** Measured kinetic energy release, showing quarter and half revivals [3].

experiments with 8 fs laser pulses on $D_2$ recorded the delay-dependent kinetic energy

release of the deuteron fragments [3] and reproduce the first quarter, half, and full wave packet revivals of our model calculation.
**Future plans:** For delays beyond the first full revival, the comparison between theory and preliminary experiments becomes inconclusive. We intend to investigate whether this discrepancy is due to insufficient experimental resolution or originates in the onset of rotational dephasing.

**2. Neutralization of negative hydrogen ions near vicinal metal surfaces** (with Himadri Chakraborty, Thomas Niederhausen, and Boyan Obreshkov)

**Project scope:** We attempt to understand the resonant transfer of a single electron, initially bound to the projectile, during the reflection of a slow ion or atom at an arbitrarily shaped, nanostructured metal surface as a function of the collision parameters, the surface electronic structure, and crystal orientatioin of the surface [4].
**Recent progress:** We developed a new set of computer programs to calculate the ground-state electronic structure of arbitrarily shaped metallic surfaces and tested our codes in applications to flat and vicinal Al, Cu, Na, and K surfaces [5]. We modeled the surface electronic structure within a statistical Thomas-Fermi approach, including a gradient (von Weizsäcker) correction to the kinetic energy and a local density approximation for the exchange and correlation energy. We self-consistently calculated the projected density of states (PDOS) of the ion-surface system for arbitrary (but fixed) positions of the ion by direct numerical propagation of the time-dependent Schrödinger equation for the motion of the active electron. From the PDOS, we obtained the static shift and width of the ion-affinity level. Our results for the work function changes of vicinal (vs. flat) metal surfaces agree with experiments and Kohn-Sham calculations.
We derived the negative ion-survival probability near vicinal surfaces by integrating a rate equation including both electron capture and loss processes. Our results show a pronounced "step-up" versus "step-down" scattering asymmetry, even if averaged over many scattering trajectories, that is caused by enhanced electron loss along the outgoing part of ion trajectories, which approach a step from below.
**Future plans:** We will investigate resonance formation and charge exchange near vicinal and other structured surfaces. In particular, we will investigate the importance of lateral confinement effects (evidence for which was found in photo-emission experiments).

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**Other DoE-sponsored publications (2003-2006):** A.A. Khuskivadze, I.I. Fabrikant, and U. Thumm, Phys. Rev. A **68**, 063405 (2003). M. Zamkov, H.S. Chakraborty, A. Habib, N. Woody, U. Thumm, and P. Richard, Phys. Rev. B**70**, 115419 (2004). M. Zamkov, N. Woody, S. Bing, H.S. Chakraborty, U. Thumm, and P. Richard, Phys. Rev. Lett. **93** 156803 (2004).