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Imaging intramolecular forces and wave functions in diatomic molecules

Ultrashort laser pulses allow for time resolution of the nuclear motion in molecules and the reconstruction of laser-distorted molecular potential curves.

Recent advances in laser technology have enabled the time-resolved imaging of the nuclear wave-packet dynamics of diatomic molecules by employing intense pump and probe laser pulses with pulse durations shorter than the molecular vibration period (Fig. 1). This allows for the nuclear wave-packet dynamics in diatomic cations to be investigated experimentally by analyzing fragment kinetic-energy-release (KER) spectra as functions of the pump-probe delay. Ro-vibrational wave packets can be described as a coherent superposition of stationary ro-vibrational states in adiabatic electronic states of the molecular cation, which undergoes cycles of dephasing and revival. For heavier molecules, such as O_2, a large number of adiabatic potential curves of the molecular ion can typically be populated in the initial ionization by the pump pulse. Consequently, tracing adiabatic states that are involved in the pump-laser-induced nuclear dynamics in heavier molecular ions is significantly more complicated than in H_2^+ (and its isotopes).

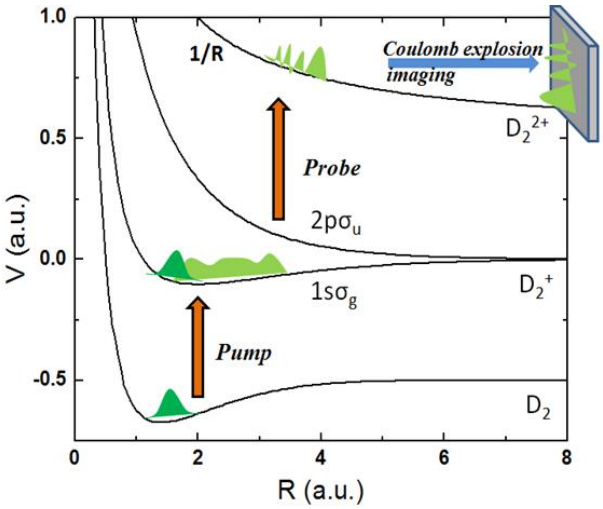

Figure 1. The pump-laser pulse launches a nuclear vibrational wave packet from the electronic and vibrational ground state of the neutral molecule onto the D_2^+ ($1s\sigma_g$) potential curve by ionizing D_2 and starts the molecular clock. After a variable time delay, an intense short probe pulse can cause fragmentation by Coulomb explosion, allowing for the detection of the fragment kinetic energy distribution and reconstruction of the D_2^+ vibrational wave packet dynamics.

Recent quantum-mechanical simulations (Magrakvelidze M *et al* 2014 J. Phys. B **47**) provided delay-dependent fragment KER spectra for the laser-induced dissociative ionization of selected diatomic molecules in both the time domain and as a function of the beat frequency (QB) between molecular vibrational levels (Fig. 2). This combined time and "QB analysis" was shown to better reveal the characteristics of the bound and dissociating molecular motion than the time-domain analysis alone. While the time-domain KER spectra display oscillation periods, revival times, and nuclear-probability-density evolution, QB imaging in the frequency domain reveals QB frequencies and the nodal structure of vibrational states and laser-electric-field-dressed molecular potential curves. Identifying the potential curves by the complimentary analysis of measured and simulated KER spectra in the time and frequency

domains provides a powerful tool for (a) analyzing nuclear dynamics in molecules, (b) identifying dissociation and reaction pathways, and (c) scrutinizing commonly used assumptions in quantum mechanical models by mapping laser-dressed potential curves. The imaging of potential curves has been demonstrated in recent proof-of principle experiments. The reconstruction of the nuclear wave packet amplitudes (up to an overall phase), in principle possible by QB imaging, is still awaiting experimental verification.

About the authors

The epicenter of this work was the Department of Physics and the J. R. Macdonald Laboratory for laser-based atomic, molecular, and optical physics at Kansas State University (KSU). It was part of the Ph.D. thesis completed by M. Magrakvelidze at KSU and the work of NSF-REU summer student Alex Kramer under the supervision of Professor Uwe Thumm. It was performed in collaboration with Professor Klaus Bartschat at Drake University and builds on previous collaborations with experimental research groups at the J. R. Macdonald Laboratory, the Max-Planck Institute for Nuclear Physics in Heidelberg (Germany), and the Institute for Nuclear Physics at the Goethe University in Frankfurt (Germany).

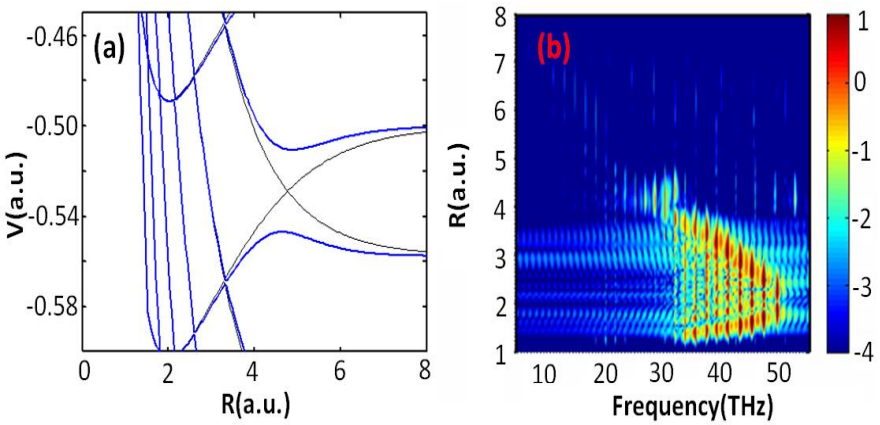

Figure 2. (a) Field-dressed adiabatic potential curves ($1s\sigma_g$ and $2p\sigma_u$) for D_2^+ in an 800 nm, 10^{13} W/cm^2, 20 fs probe laser pulse. (b) Power spectrum of the field-dressed molecular ion for a delay range of 5 ps.

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