MAPPING OF MOLECULAR DYNAMICS IN INTENSE LASER FIELDS

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Abstract. Femtosecond time resolved molecular multi-photon-ionization studies with photoelectron detection on the Na_2 prototype are reported. In intense laser fields electronic transitions at non Franck-Condon allowed internuclear distances are observed.

1. Introduction

Photoelectron spectroscopy in combination with femtosecond pump-probe and molecular beam techniques is a powerful tool to map molecular vibrational wavepacket motion along the internuclear coordinate with sub-Ångström spatial resolution and femtosecond time resolution [1]. This method allows therefore to study the interaction of molecules with intense laser fields as a function of the internuclear separation.

2. Experiment

In the experiments reported here the pump laser (70 fs, $I_0=10^{11}$W/cm^2) prepares the population on the electronic states of the sodium dimer molecule ($X\ ^1\Sigma_g^+$, $A\ ^1\Sigma_u^+$, $2\ ^1\Pi_g$) that are involved in the multiphoton ionization process at an excitation wavelength of 620 nm. A time delayed probe laser of variable intensity ionizes the molecule. The wave-packet motion on the electronic states. is mapped via energy resolved photoelectron detection using a magnetic bottle spectrometer.

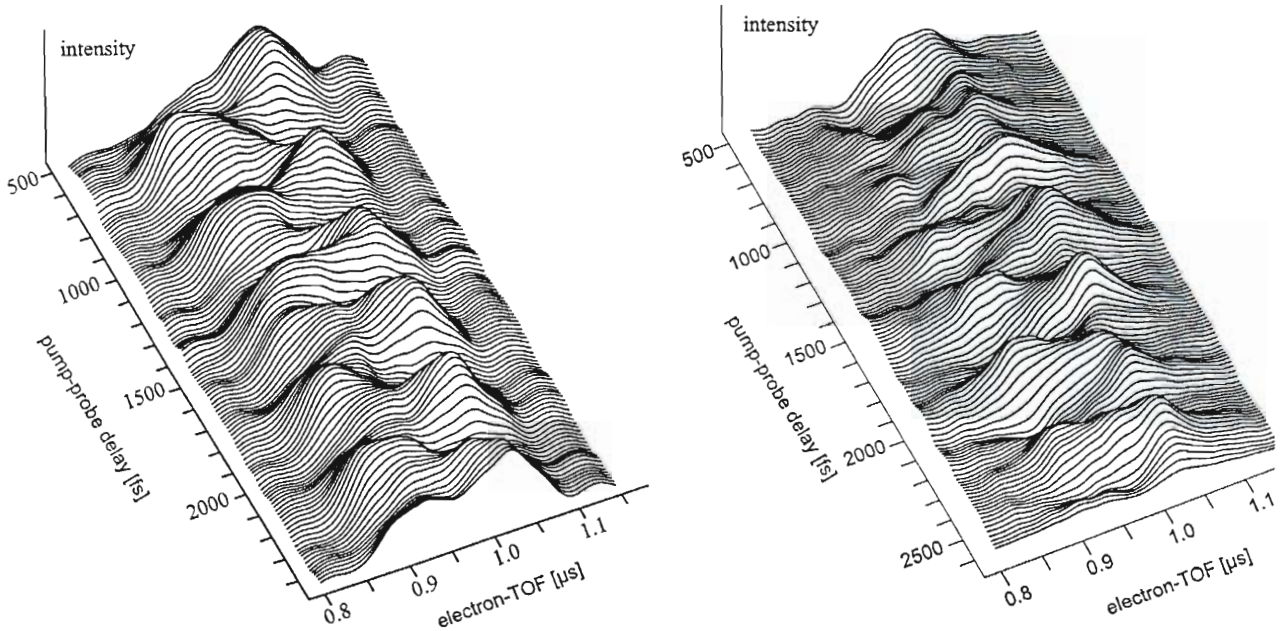

Fig. 1. Mapping of vibrational wave-packet motion in intense probe laser fields at fixed attenuated pump laser intensity. Left: Probe laser intensity I_0 (10^{11} W/cm^2); right: probe laser intensity 10 I_0.

3. Results and Discussion

The pump laser prepares the wavepackets on the involved electronic states, where the intensity determines the population. After excitation the wave-packet motion evolves on unperturbed potentials. Increasing the intensity of the probe laser (70 fs, 620 nm) at fixed pump laser intensity leads to different mapping spectra (see fig. 1).

In order to analyze the mapping spectra we performed Fourier transformations at 0.90 μs electron time-of flight corresponding to small internuclear distances [1], and at 1.02 μs corresponding to large internuclear distances (see fig. 2).

We observe two dominant contributions to the dynamics corresponding to the wavepacket motion on the A $^1\Sigma_u^+$ state (ca. 110 cm^{-1}) and the 2 $^1\Pi_g$ state (ca. 90 cm^{-1}). Using attenuated femtosecond probe laser pulses the wave-packet dynamic is probed on unperturbed molecular potentials. This can be seen from the upper part in fig.2. At small internuclear distances (fast electrons) there is a resonant enhanced ionization out of the A $^1\Sigma_u^+$-state via the 2 $^1\Pi_g$ state and we observe a frequency contribution of the A-state-wavepacket to the overall dynamics. At large internuclear distances (slow electrons) the resonant transition out of A $^1\Sigma_u^+$ via the 2 $^1\Pi_g$ is not Franck-Condon allowed. At attenuated probe laser intensity the dynamic at large internuclear distances is therefore dominated by the wavepacket motion on the 2 $^1\Pi_g$ state. By increasing the intensity of the time delayed probe laser fields we observe an onset of the A-wavepacket frequencies. This contribution becomes dominant at a probe intensity of 10 I_0.

We attribute this observation of electronic transitions at non Franck-Condon allowed internulcear distances to light induced molecular potentials during the probe laser interaction (see for ex. [2]). Note that a non-resonant transition out of the A-state would lead to the formation of "fast" electrons

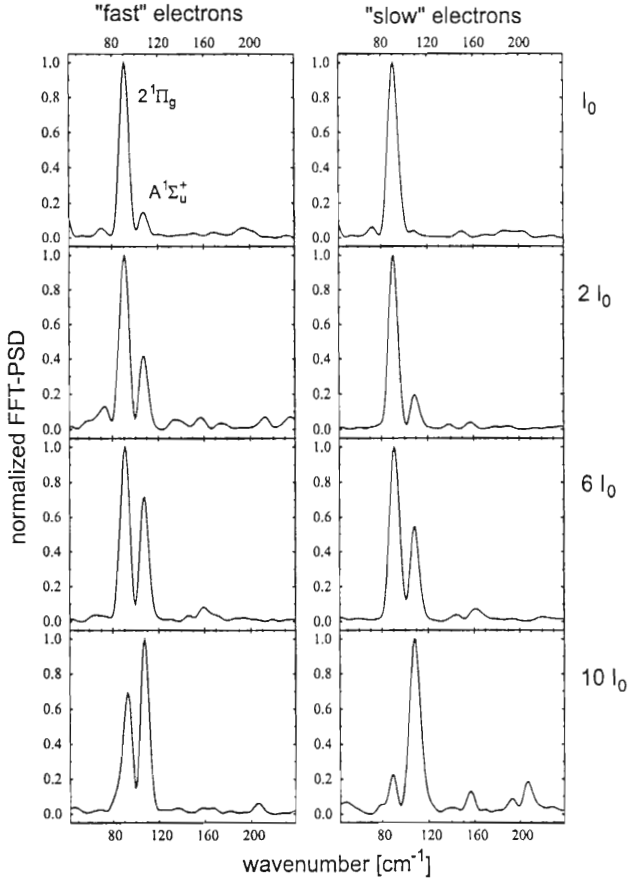

Fig. 2. Normalized FFT-PSD spectra at two fixed TOF, representing the inner (left, 0.90μs) and the outer (right, 1.02μs) turning point of the wavepacket in the Π-state. For the "fast" photoelectrons we observe a frequency contribution of both wavepackets (A and the Π-state). For "slow" photoelectrons the contribution of the A-wavepacket starts to appear at intermediate intensities and increases very fast with laser intensity. This behavior is attributed to the effect of perturbed molecular potentials in high laser fields.

4. Conclusions

Electronic transitions at non Franck-Condon allowed internuclear distances can be achieved by fs-pump-probe experiments with adjusted probe laser intensities, where the pump-laser intensity determines the amount of population on the electronic state of interest. This result is particularly important for coherent control schemes.

References

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