

Molecular ATI and ATD with femtosecond laser pulses

A. Assion, T. Baumert, J. Helbing, V. Seyfried and G. Gerber

Physikal. Institut, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany

Abstract. Time-resolved observation of molecular ATI is reported. We show both in single pulse and pump-probe experiments, that threshold and above threshold ionization signals are sensitive to the vibrational wavepacket dynamics. We also present first results of non-resonant multiphoton dissociation (ATD) of Na_2^+ in intense ultrashort laser fields.

ATI has been extensively studied in atoms both experimentally and theoretically [1]. When molecules interact with intense laser fields an additional degree of freedom is given by the motion of the nuclei. In the Na_2 molecule vibrational wavepackets can be excited and the influence of vibrational motion on molecular ATI can be studied [2]. The absorption of excess photons may also lead to above threshold dissociation (ATD) [3].

Fig. 1 shows ATI spectra of Na_2 recorded with 80fs laser pulses at 618nm ($I_0 \approx 10^{12}$ W/cm²). Three peaks separated by the photon energy can be seen at 0.9eV, 2.9eV and 4.9eV. These energies correspond to ionization of Na_2 with 3 (threshold), 4 and 5 photons of the molecule in the $v''=0$ level of the electronic ground state. In addition electrons of slightly lower energy are formed with increasing laser intensity and eventually dominate the spectrum. By the Franck-Condon principle, the nuclear kinetic energy is conserved during electronic transitions. Taking into account the two intermediate neutral electronic states $A^1\Sigma_u^+$ and $2^1\Pi_g$ which resonantly enhance ionization, the photoelectron kinetic energy is seen to decrease from 0.94eV to 0.8eV with increasing nuclear separation and therefore serves as a measure of the internuclear distance at which ionization occurs [4]. Excitation with an ultrashort laser pulse of only moderate intensity leaves the nuclei in the molecule essentially fixed since they have no time "to move apart" before the laser is turned off. Threshold and above threshold ionization therefore take place with the nuclear wavefunction peaked at small internuclear distances. In much more intense laser fields,

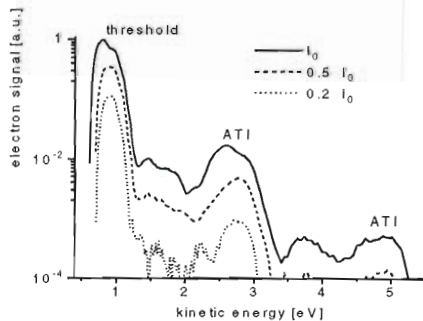


Fig. 1: Threshold and above threshold ionization electron spectra of Na_2 .

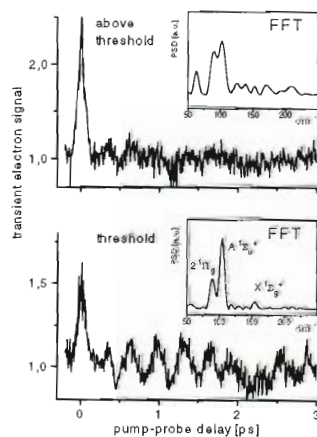


Fig. 2: Transient electron signal of electrons from above threshold ionization (top) and from threshold ionization (bottom).

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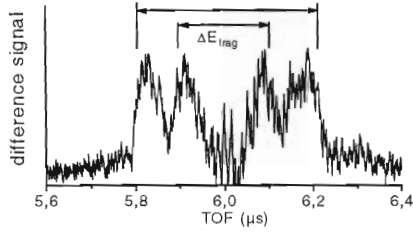


Fig. 3: Time of flight (TOF) spectrum of Na^+ fragments originating from the dissociation of Na_2^+ by an intense 800nm 80fs-laser pulse.

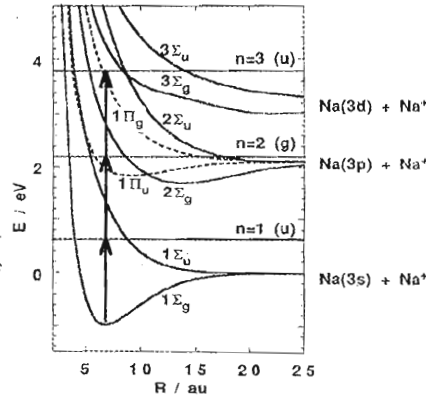


Fig. 4: Lowest lying electronic states of Na_2^+ .

however, population is transferred to the resonant excited states already during the leading edge of the laser pulse. The vibrational wavepackets in these states spread earlier and propagate to larger internuclear distances during the molecule-laser interaction, leading to less energetic threshold (0.8eV) and above threshold photoelectrons seen in Fig. 1. In a time resolved experiment we have for the first time applied pump-probe techniques to the detection of threshold and ATI electrons using 40fs pulses at 618nm. The transient threshold electron signal, shown in the lower part of Fig. 2, is composed of frequencies of vibrational wavepackets propagating in the $A^1\Sigma_u^+$, $2^1\Pi_g$ and the ground state $X^1\Sigma_g^+$ of Na_2 [5]. At a five times higher intensity ($\approx 10^{11}\text{W/cm}^2$) an oscillating transient is also observed for electrons from above threshold ionization which directly shows that ATI is sensitive to the wavepacket motion in the $A^1\Sigma_u^+$ and $2^1\Pi_g$ states (upper part of Fig. 2).

In a further experiment we studied the effects of altered potential curves in intense laser fields on above threshold dissociation (ATD) as proposed in a theoretical publication [6]. Fig. 3 shows a TOF spectrum of Na^+ fragments from the dissociation of Na_2^+ by 800nm 80fs-laser pulses. Na_2^+ was prepared in low vibrational levels of its electronic ground state by resonant two photon ionization of Na_2 with a nanosecond laser. The signals recorded with each laser alone were subtracted from the two laser signal. Two total fragment energies of $0.28\pm 0.1\text{eV}$ and $0.85\pm 0.1\text{eV}$ can be determined and assigned to two and one photon fragmentation with the asymptotes $\text{Na}(3p)+\text{Na}^+$ and $\text{Na}(3s)+\text{Na}^+$ respectively. None of the lowest lying excited electronic states of Na_2^+ can be reached with 800nm photons (Fig. 4). However, in the intense laser field, population is observed to “leak out” into dissociating channels, which can be explained by the strong deformation of the corresponding adiabatic dressed states [6].

References

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