

Photodissociation of Na_2^+ in intense femtosecond laser fields

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Abstract. Photodissociation of Na_2^+ in a well defined vibrational level is studied for laser intensities ranging from 10^{11} W/cm² up to 10^{14} W/cm². Two intensity dependent competing processes are observed: photodissociation by laser-induced potentials and by Coulomb explosion.

1. Introduction

High intensity multiphoton physics is now well established and a great deal of work has concentrated to understand processes within and beyond the regime of perturbation theory. Recently observed features of small molecules subjected to intense laser fields are: above-threshold dissociation, molecular bond softening, vibrational population trapping, Coulomb explosion, etc. [1, 2]. The model system H_2^+ and more recently Ar_2^+ in an intense laser field have been intensively studied. However, a detailed comparison between experiment and theory is very difficult because due to the experimental preparation of these molecular ions a number of different vibrational states contribute to the same spectrum of photofragments. If the interest is to study the influence of the laser pulse duration on the photodissociation dynamics, H_2^+ is again not a perfect candidate because the time scale of the vibrational period is about 15 fs and intense laser fields of such time durations are not yet generally available. Therefore we have chosen the molecular ion Na_2^+ to study the different processes arising from the interaction of the molecule with the intense and ultrashort laser field.

The molecular ion Na_2^+ can be prepared in a well defined vibrational level by nanosecond stepwise ionization in a molecular beam. Laser pulses with a duration of 90 fs, much shorter than the vibrational period of the Na_2^+ -molecule (300 fs) are well suited for studying the photodissociation dynamics. This diatomic molecule is an ideal candidate to study photodissociation processes with ultrashort and intense laser pulses and in particular to investigate the influence of the laser pulse duration on the dissociation dynamics [3].

2. Experimental setup

The experimental setup is shown in Figure 1. The neutral diatomic molecule Na_2 was produced vibrationally cold in its electronic ground state by expanding pure sodium from an oven operated at 1000 K. With a ns-laser Na_2^+ was prepared predominantly in the vibrational state $v'' = 0$ of the ionic ground state by resonant

two photon ionization. These ions were extracted from the molecular beam by a static electric field and illuminated in a second interaction region with the intense femtosecond laser pulses. Due to the separation of the two interaction regions space charge effects play no role. The resulting charged photodissociation products were detected in a linear time-of-flight (TOF) spectrometer.

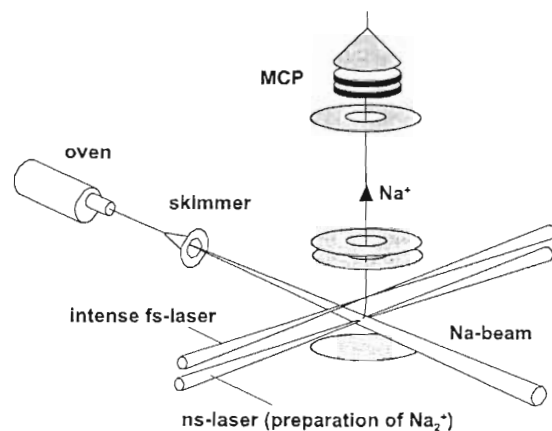


Fig. 1. Experimental setup. Two separate interaction regions are used. In the first region the Na_2^+ -ions are prepared by resonant two photon ionisation. The ions are transferred by a small electric field into the second interaction region where the Na_2^+ ($v''=0$) interact with intense femtosecond laser pulses.

3. Results and Discussion

The measured TOF-spectrum is shown in Figure 2(a) for different intensities of the fs-laser. With the laser polarization parallel to the TOF-axis we observe four peaks in the TOF-spectrum which we assign to different dissociation channels. To determine the released kinetic energy of the fragments and the total signal of the different dissociation channels we deconvoluted the measured TOF-spectrum into an energy spectrum. In Figure 2(b) the fragment distribution between the four channels and the corresponding energy release is displayed for several laser intensities.

The photodissociation processes can be understood in the picture of “adiabatically field-dressed state potentials” [4]. The molecule dissociates by moving from the field-free equilibrium distance towards larger internuclear distances on the laser-field induced repulsive potentials. Comparison of the results with theoretical calculations [3] show that the fragments with a released energy of 1.3 eV and of 0.5 eV originate from resonant 1- and 2-photon absorption in the field-dressed potential curves, respectively. At the lowest laser intensities (10^{11} W/cm²) only these two channels are observed. At intermediate laser intensities (10^{12} - 10^{13} W/cm²) a third channel with a total released energy of 2.9 eV starts to emerge. We assign this channel to a resonant 3-photon absorption with subsequent ionization and formation of Na_2^{2+} , which undergoes Coulomb explosion. At this intensity the molecule moves on the light-induced potential curves to larger internuclear separations and reaches a distance where the ionization probability is close to unity [4], still within the duration of the laser pulse. From the measured fragment energy we conclude that this Coulomb explosion takes place at an internuclear separation of about 4.8 Å. At the highest

laser intensity (10^{14} W/cm²) a strong contribution from a fourth channel with a released energy of 4.7 eV dominates the fragment energy distribution. From the measured energy we assign this channel to multiphoton ionization to the Na₂⁺⁺-Coulomb curve taking place close to the Na₂⁺ internuclear equilibrium distance of 3.6 Å.

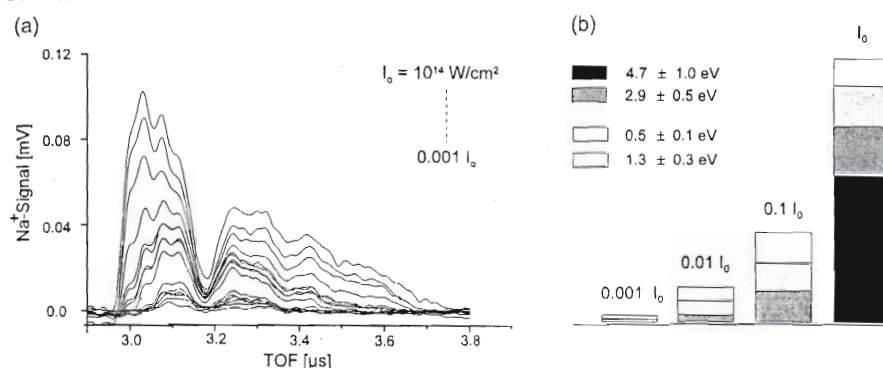


Fig. 2. (a) TOF-spectra of Na₂⁺ obtained with an intense 790 nm, 90 fs laser and for different laser intensities. (b) Total kinetic energy of the fragments as obtained from the deconvolution of the TOF-spectra and distribution between the fragmentation channels for several laser intensities.

From these results we conclude that depending on the laser intensity, dissociation of Na₂⁺ competes with ionization to Na₂⁺⁺. For low laser intensities dissociation clearly dominates. At moderate intensity of 10^{12} W/cm² ionization comes into play so that dissociation after 3-photon absorption cannot proceed entirely. At the highest laser intensities direct ionization to Na₂⁺⁺ and fragmentation via the Coulomb curve dominates.

4. Conclusions

In conclusion the photodissociation of Na₂⁺ in a given vibrational level and interacting with an intense femtosecond laser field has been studied. A comparison with recent theoretical results enabled us to assign each observed fragmentation channel. Additionally, the interplay between the two competing processes of dissociation via laser-induced potentials and ionization with subsequent Coulomb explosion was observed.

References

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