

## Strong-field dissociation dynamics of molecular dications

This content has been downloaded from IOPscience. Please scroll down to see the full text.

2015 J. Phys.: Conf. Ser. 635 112044

(<http://iopscience.iop.org/1742-6596/635/11/112044>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 129.130.37.147

This content was downloaded on 24/03/2016 at 14:59

Please note that [terms and conditions apply](#).

# Strong-field dissociation dynamics of molecular dications

Bethany Jochim<sup>\*1</sup>, T. Severt<sup>\*2</sup>, M. Zohrabi<sup>\*</sup>, U. Ablikim<sup>\*</sup>, Ben Berry<sup>\*</sup>, B. Gaire<sup>\*</sup>, K.J. Betsch<sup>\*</sup>, F. Anis<sup>\*</sup>, Tereza Uhlíková<sup>†</sup>, K.D. Carnes<sup>\*</sup>, E. Wells<sup>‡</sup>, B.D. Esry<sup>\*</sup>, I. Ben-Itzhak<sup>\*</sup>

<sup>\*</sup>J.R. Macdonald Laboratory, Department of Physics, Kansas State University, Manhattan, Kansas 66506 USA

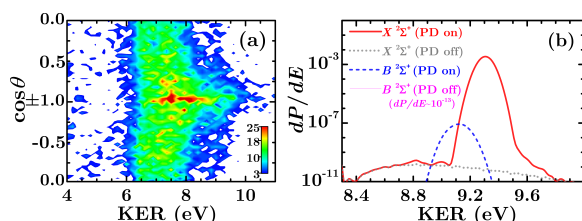
<sup>†</sup>Department of Analytical Chemistry, Institute of Chemical Technology, Prague Technická, Czech Republic

<sup>‡</sup>Department of Physics, Augustana College, Sioux Falls, South Dakota 57197 USA

**Synopsis** We focus on the dissociation of metastable molecular dications induced by intense, ultrafast laser pulses. In particular, we demonstrate the dominant role of commonly-neglected permanent-dipole transitions and drive dissociation via a pump-dump-like mechanism within a single laser pulse.

Many doubly-charged molecules have ground electronic states with lifetimes spanning from picoseconds to seconds [1], thus enabling unique studies. We focus on dissociation of several-keV beams induced by intense, ultrafast laser pulses. Specifically, we demonstrate that permanent-dipole (PD) transitions can play a key role and that the structure of the metastable electronic ground state can delay stimulated emission, leading to pump-dump-like transitions.

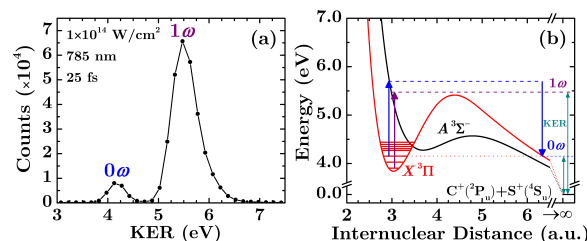
In  $\text{NO}^{2+}$ , dissociation parallel to the laser polarization is prominent at high intensities, as shown in Fig. 1(a). Contrary to common intuition dictating that electronic transitions always prevail, we find that permanent-dipole transitions are extremely important in this system. We demonstrate this with time-dependent Schrödinger equation (TDSE) calculations [2], shown in Fig. 1(b), together with the most likely number of photons involved in the transition, derived from fitting the measured angular distribution in Fig. 1(a) [3]. Specifically, we find that the dominant pathways leading to the aligned feature at high kinetic energy release (KER  $\geq 8.9$  eV) are multiphoton permanent-dipole-driven vibrational excitations on the electronic ground state leading to its continuum.



**Figure 1.** (a) Density plot of  $\text{NO}^{2+} \rightarrow \text{N}^+ + \text{O}^+$  dissociation as a function of KER and  $\cos\theta$  for  $5 \times 10^{15} \text{ W/cm}^2$  laser intensity. (b) Calculated KER spectra for transitions starting from  $X^2\Sigma^+(v=3)$  and dissociating on the indicated state.

In  $\text{CS}^{2+}$ , photodissociation via  $X^3\Pi \rightarrow A^3\Sigma^-$  transitions yield the main KER peak, marked as  $1\omega$  in Fig. 2(a). In addition, a more intriguing

peak, marked  $0\omega$ , appears at about one-photon energy lower KER. This  $0\omega$  peak can be understood as a pump-dump process driven by one laser pulse, where the same  $X^3\Pi \rightarrow A^3\Sigma^-$  excitation (“pump”) is followed by delayed stimulated emission (“dump”). This delay, on the order of a few tens of fs, is due to the potential barrier of the electronic ground state, which makes stimulated emission energetically forbidden until the dication has stretched far enough, as indicated in Fig. 2(b). Once the Condon point is reached, stimulated emission is most likely, and the pump-dump-like process results in  $\text{C}^+ + \text{S}^+$  fragments with a KER value reflecting no net energy exchanged with the laser field. Finally, we also observe this “pump-dump-like” mechanism in  $\text{CO}^{2+}(v=0)$ .



**Figure 2.** (a) Measured KER spectrum for  $\text{CS}^{2+} \rightarrow \text{C}^+ + \text{S}^+$ . (b) Potential energy curves for  $\text{CS}^{2+}$  [4] with dissociation pathways for the one-photon ( $1\omega$ ) and pump-dump ( $0\omega$ ) transitions.

This work was supported by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy. BJ is also supported by DOE-SCGF (DE-AC05-06OR23100). TU is supported by GACR and MetaCentrum.

## References

- [1] D. Mathur 2004 *Physics Reports* **391** 1
- [2] F. Anis and B. D. Esry 2008 *Phys. Rev. A* **77** 033416
- [3] A. M. Sayler *et al* 2007 *Phys. Rev. A* **75** 063420
- [4] T. Šedivcová *et al* 2006 *J. Chem. Phys.* **125** 164308

<sup>1</sup>E-mail: [bjochim@phys.ksu.edu](mailto:bjochim@phys.ksu.edu)

<sup>2</sup>E-mail: [tsevert@phys.ksu.edu](mailto:tsevert@phys.ksu.edu)

