Electron-Nuclear Coupling through Autoionizing States after Strong-Field Excitation of H_2 Molecules

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When an H_2 molecule is exposed to a strong laser field, the following two single ionization channels may occur:

$$H_2 \xrightarrow{IR} H_2^+ + e^- \tag{1}$$

$$H_2 \xrightarrow{IR} H^+ + H + e^- \tag{2}$$

The first channel is called bound ionization, in which one electron is freed by the laser field and the molecule ends up as a bound molecular ion H_2^+ and a free electron. The second channel is named dissociative ionization, where the H_2 molecule breaks up into a proton, a hydrogen atom and a free electron.

By using a Reaction Microscope (REMI) [1] and ultrashort laser pulses, we investigated channel-selective electron emission from strong-field ionization of H_2 [2]. Measuring of electrons and ions in coincidence, we distinguished the bound and dissociative ionization channels and extracted electron momenta for the two channels. According to the two-step mechanism [3], the laser-induced dissociation of H_2 molecules is described as follows: the molecule is ionized by the laser field in the first step and then dissociates as the vibrational wave packet is excited from the bound $1s\sigma_g$ state to the repulsive $2p\sigma_u$ state in most cases, which means that the dissociative ionization process is independent from the bound ionization process. On first approximation, the electrons from the two channels should thus be the same. As shown in Fig. 1, the fanlike stripes and the above-threshold ionization rings appear for both channels, as expected from the two-step mechanism. However, an enhancement of the photoelectron yield in the low-energy region for the bound ionization channel can be clearly observed.

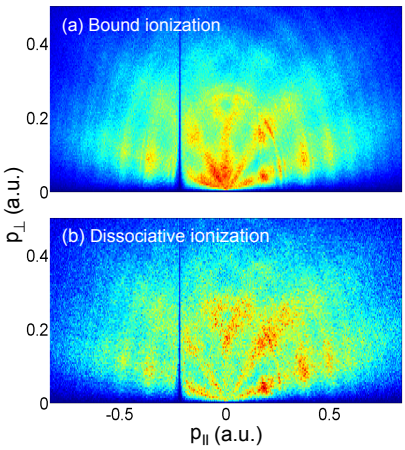

Fig. 1: Electron momentum distribution for (a) bound ionization and (b) dissociative ionization.

We interpret the low-energy enhancement in the electron yield for bound ionization as being due to the population and subsequent decay of autoionizing states. For H_2 molecules, the autoionzing states represent a series of high-lying Rydberg states and are characterized by a simultaneous electronic and vibrational excitation. Instead of populating the H_2^+ bound state and being excited to the repulsive state for dissociation, the molecule can be excited to the above-mentioned autoionizing state located above the ionization limit. Therefore, an autoionization process occurs, in which the vibrational energy of the nuclei is transferred to the excited electron, which becomes free and appears in the photoelectron momentum spectra.

References

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