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Probing O_2^+ potential curves with an XUV–IR pump–probe experiment

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Synopsis Upon ionization of ground state O_2 molecules in a short XUV pulse, we observe a time-dependent vibrational wave packet in the potential of the binding $O_2^+(a^4\Pi_u)$ state. Our pump–probe delay dependent kinetic-energy-release (KER) spectra are in qualitative agreement with the results of coupled-channel simulations that are based on calculated Born-Oppenheimer potential-energy curves (PECs). Using a Morse potential adjusted to the experimental data most features of the experimental spectra are reproduced quantitatively.

In an XUV–IR pump–probe experiment, an extreme ultraviolet (XUV) pulse initiates an oscillating nuclear vibrational wave packet in the binding $O_2^+(a^4\Pi_u)$ potential by single-photon ionization. After a variable time delay population is transferred to the weakly repulsive $O_2^+(f^4\Pi_g)$ state by an infrared (IR) pulse [(Duration (FWHM) \approx 12 fs, Intensity \approx 3×10^{12} W/cm²) (Fig. 1). The initial three-dimensional momenta of the created photoions and photoelectrons are determined using a reaction microscope.

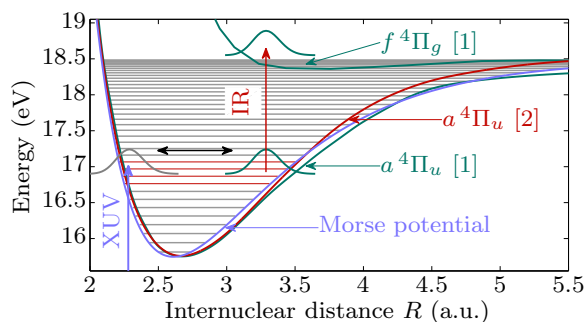


Figure 1. O_2^+ PECs calculated in Refs. [1] and [2] as well as a Morse potential adjusted to the experimental data.

The experimental yield of O^+ ions with $KER < 0.08$ eV is plotted as a function of the pump–probe delay in Fig. 2. Clearly visible is an oscillation with a period of 40 fs. The anharmonicity of the $a^4\Pi_u$ PEC causes the originally well localized wave packet to dephase over a period of approximately 640 fs before it revives after 1270 fs.

With a coupled-channel calculation, we simulate the wave-packet oscillation and the probing process for different sets of PECs. The results are shown in the three lower graphs of Fig. 2.

The revival time of the wave packet is overestimated in the simulations using the PECs calculated in Refs. [1] and [2]. If the wave-packet is propagated within a Morse potential adjusted to the experimental data, the revival time is in very good

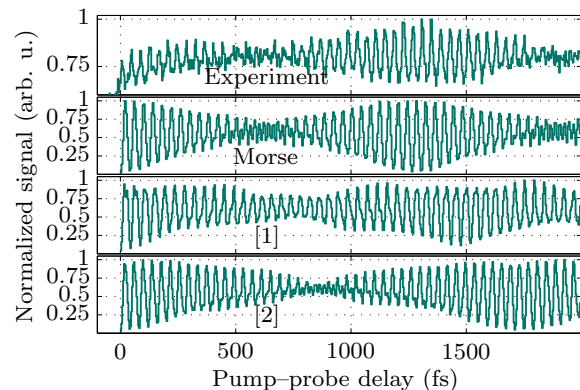


Figure 2. O^+ yield as a function of the pump–probe delay. Experimental data (top) and three simulated spectra using a Morse potential as well as the PECs calculated in Refs. [1] and [2].

The close resemblance between the Morse potential and the predicted PECs in Fig. 1 demonstrates that the revival time and our experimental method is sensitive to small variations in the shape of the binding PEC. Additionally, the KER distribution of the O^+ ions contains information about the shape of the repulsive PEC. This allows us to distinguish between the $O_2^+(f^4\Pi_g)$ PECs calculated in Refs. [1] and [2].

References

- [1] C. M. Marian, *et al.*, 1982 *Mol. Phys.* **46** 779
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