The influence of nuclear motion on the electron dynamics in an efficient sub-cycle control of the molecule K_2

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With precise control of the temporal phase of femtosecond laser pulses direct manipulation of valence electron motion is possible. The control scheme relies on the selective population of dressed states first demonstrated for atoms [1]. In a combined experimental and theoretical approach, we show that the control scheme can be transferred to molecules. For the example of the potassium dimer specific target states can be addressed through the control of the electron dynamics. The key element is the oscillating dipole created by the superposition of the $X^1\Sigma_g^+$ ground state and the $A^1\Sigma_u^+$ first excited state. We highlight the effect of the nuclear motion on the control scheme and in particular on the oscillating dipole.

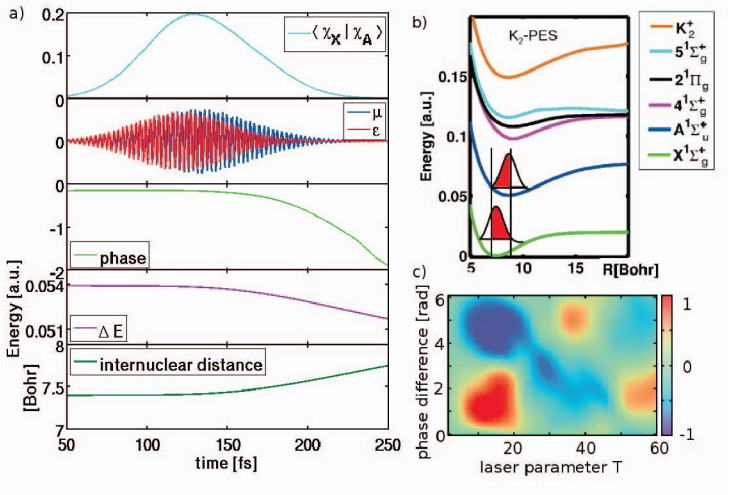

Fig. 1: a) Properties affected by nuclear motion (top to bottom): nuclear overlap (cyan), oscillating dipole (blue), laserfield in red (not affected), phase of the oscillating dipole (green), energy difference of the ground and first excited state (magenta), average nuclear distance (dark green). b) Potential energy surfaces of K_2 with nuclear wave packets on the first two states. Their the overlap is shown in red. c) control landscape dependent on the time delay T and the phase difference between the two laser sub pulses.

The scheme consists of two sub pulses, the first prepares the superposition and the second one guides the system to the target state. Figure 1 a) shows the effect of the first subpulse (second panel, red). In case of atoms the superposition would stay on, only influenced by external perturbations, but in a molecule nuclear wavepacket motion is induced and effects the oscillating dipole. The wavepacket motion is visualized in Figure 1 b) on the relevant potential energy surfaces of K_2. It reduces the nuclear overlap (Figure 1,cyan) and thus damping the oscillating dipole (blue) and fixing the time window for control [2,3]. Secondly, the oscillating dipole changes its phase (green) due to the change in energy difference (magenta) between the first two states experienced along the reaction coordinate (dark green). This gives rise to the control landscape (Figure 1 c)), recorded for the two critical parameters, time delay between the two subpulses (T) and their phase difference.

References

[1] M. Wollenhaupt and T. Baumert "Ultrafast laser control of electron dynamics in atoms, molecules and solids." Faraday Discuss. **153**, 9-26 (2011)
[2] M. Wollenhaupt and T. Baumert "Ultrafast strong field quantum control on K2 dimers." J. Photochem. Photobiol. A **180**, 248-255 (2006).
[3] P. v. d. Hoff,M. Kowalewski and R. de Vivie-Riedle "Searching for pathways involving dressed states in optimal control theory." Faraday Discuss. **153**, 159-171 (2011).

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