Abstract

Interferences in vibrational wave packets of Br_2 molecules are controlled in presence of a solid Ar environment that provides decoherence. By applying a negatively or positively chirped excitation pulse, one can set the clock backward respectively forward in the wave packet propagation. Based on this mechanism, we present a general scheme to record vibrational decoherence. Wave packets are spatially focused at T_{opt} by applying negatively chirped pulses. From the focussing contrast, we determine a vibrational dephasing time on the B state of $T^{vib}_{deph} = 3$ ps. We use positively chirped pulses to bring the formation of fractional revival structures forward with respect to T^{vib}_{deph}. By exciting four vibrational levels with such a pulse, we observe a 1/6 revival indicating the vibrational coherence time T^{4}_{vib} for exactly four levels. The required chirp prolonges the pulse duration by a factor of ten to $\Delta\tau = 300$ fs. Electronic dephasing T^{el}_{deph} restricts the revival control efficiency to parts of the pulse with $\Delta\tau < T^{el}_{deph}$, which allows to derive $T^{el}_{deph} > 300$ fs.