Probing ultrafast molecular dynamics with high-order harmonic generation

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We report theoretical calculations of high-order harmonic generation (HHG) from a dynamically evolving molecular system on the example of N_2O_4. A large amplitude ground state vibrational wavepacket along N-N axis of N_2O_4 can be generated using impulsive stimulated Raman scattering with a short laser pulse (the pump). A second, more intense laser pulse (the probe) is used with some time delay to generate high-order harmonics, which have been observed to oscillate as a function of time delay [1]. We present an extension of the quantitative rescattering theory [2,3] to incorporate the internal vibrational degrees of freedom. We also show that the probe pulse has very strong effect on the vibrational wavepacket, which cannot be neglected in the realistic simulation.
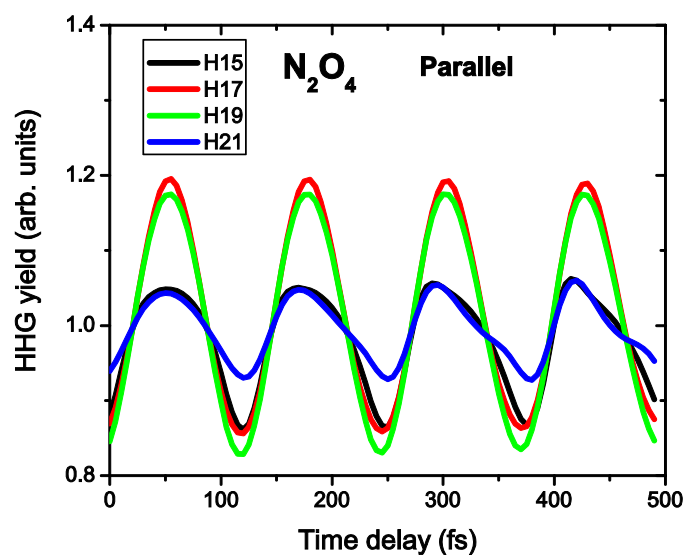
Fig. 1 HHG yield for few harmonic orders as a function of time delay between the pump and probe pulses. The harmonic yield is normalized to the yield generated without the pump pulse.

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