High harmonic generation in a flat water-jet with double MIR pulses

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High harmonic generation (HHG) is one of the most fundamental processes in strong-field-driven quantum systems. The mechanism of HHG in gas media is well understood by the three-step-model in isolated atoms or molecules, while that in crystalline solids is modelled by the carrier dynamics in the band structure. HHG in liquids is recently demonstrated [1] using the flat liquid-jet technique [2], but the underlying physics is not yet fully clarified because of the dynamic complexity of the molecular coordination.

To gain an insight into the mechanism of liquid HHG, we performed HHG experiments in a flat water-jet with twin MIR pulses and measured the time evolution of the high harmonic spectra. We used a KTA-based optical parametric chirped-pulse amplification (OPCPA) source which produces intense MIR pulses (44 μJ, 120 fs at 3.2 μm, 450 Hz) [3]. With tight focusing using an off-axis parabolic mirror (f=25.4 mm) onto a flat water-jet (1-3 μm thick), we observed odd-order high harmonics up to the 11th (~290 nm). The double pulse excitation was realized by placing a split mirror pair before the off-axis parabola. High harmonics produced by one of the MIR pulses was spatially selected and then measured by a fiber-coupled spectrometer. Fig. 1 shows the time dependence of the observed 5th harmonics at 640 nm. Around the temporal overlap, we observed clear interference fringes that can be understood by high-order autocorrelation. At long delays ($\tau >$ 100 fs), we observed a persisting interference-like signal up to 400 fs, exceeding the MIR pulse duration. This result suggests that a coherently oscillating dipole is induced by the first MIR pulse which interacts with the second MIR pulse to enhance or suppress the HHG process. We also observed slow increase of the signal offset starting at τ~200 fs, indicating the coupling of the HHG process to low-frequency degrees of freedom.

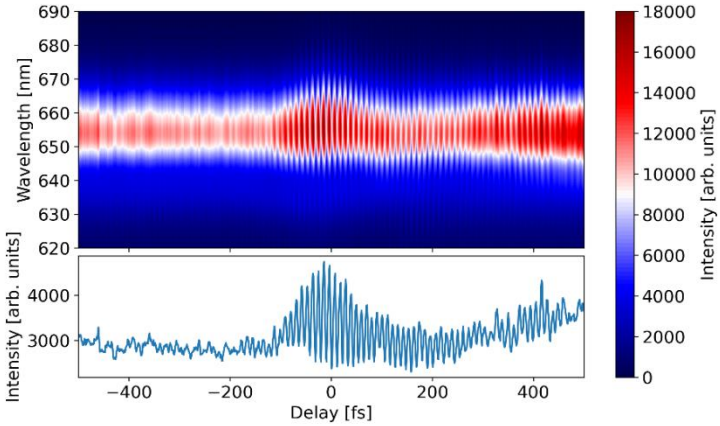

Fig. 1 5th harmonic spectrum from liquid water with the pump-probe delay from -500 to 500 fs

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[2] M. Ekimova et al., *Struct. Dyn.* **2**, 054301 (2015).

[3] F. Lu and P. Xia et al., *Opt. Lett.* **43**(11):2720-2723 (2018).