Supplemental Document

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High-harmonic generation in liquids with few-cycle pulses: effect of laser-pulse duration on the cut-off energy: supplement

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Supplementary Information: High-harmonic generation in liquids with few-cycle pulses: effect of laser-pulse duration on the cut-off energy

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Experimental identification of intensities below plasma threshold

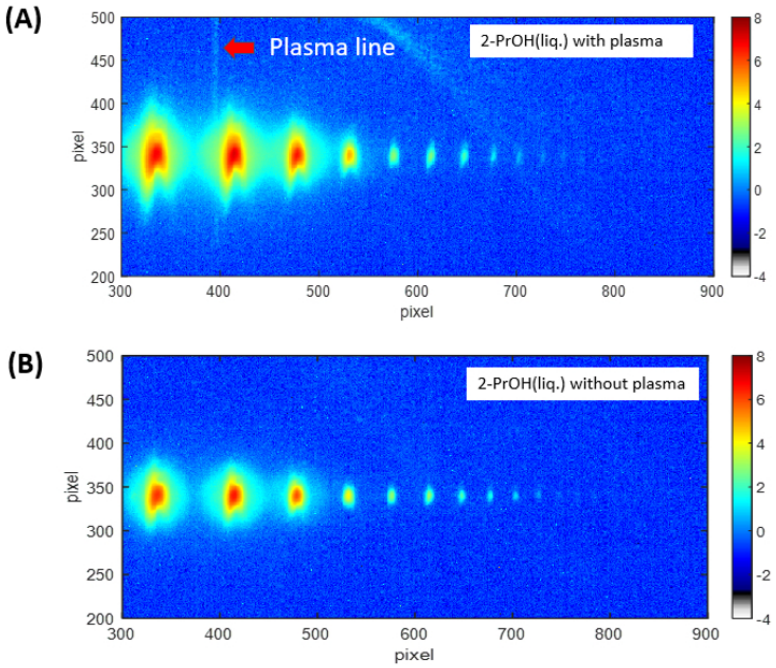

Fig. S1. (A) Shows the raw MCP image of the high-harmonic spectrum of isopropanol generated with a multi-cycle 1800 nm laser pulse in the plasma generation regime. (B) Shows the same high-harmonic spectrum of isopropanol generated with a multi-cycle 1800 nm laser pulse at a lower incident laser power where there is no plasma generation.

For the liquid-phase measurements, the data has been acquired at intensities below plasma generation. This can be clearly determined from the raw MCP image. As seen in Figure S1(A), in the presence of plasma, plasma lines (marked with a red arrow in Figure S1(A)) appears in the raw MCP image. Since plasma emission is isotropic these emissions appear as continuous straight lines in the image as opposed to the high harmonics, which have well-defined divergence properties. Under identical experimental conditions, with reduced intensity below the plasma generation threshold, we see that these isotropic emission lines from plasma completely disappear (Figure S1(B)). Thus, just from the raw MCP spectrum we can clearly identify the onset of plasma generation and keep the intensity below it.

Comparison of isopropanol liquid and gas phase spectra acquired at identical experimental conditions

For longer wavelengths since the harmonic generation efficiency drops drastically with increasing wavelength and the gas phase generated purely by evaporation might not be dense enough to generate detectable harmonic signal. Therefore, lateral scanning of the jet to get a gas reference spectrum is not ideal However, it is known that the gas density decreases monotonically with the distance from the liquid-gas interface and at high enough laser intensity the transmitted laser pulse can generate a harmonic signal from the gas phase near the exiting surface of the flat-jet(from the direction of laser propagation). This in addition to the driving laser itself causes heating of the liquid, which increases evaporation resulting in an increased gas density for the next arriving laser pulse. This effect is absent when we laterally shift the flat-jet out of the interaction region. We have therefore designed a heatable bubbler that gives a higher gas density target in comparison to pure evaporative cooling for acquiring the reference gas spectrum of isopropanol (free of any liquid contribution). In our work (Figure 2C) we have presented the entire high-harmonic spectra showing that the cut-off energy remains fixed at $\sim$ 11.3 eV for liquid isopropanol for different intensities in the few-cycle regime ($\sim$ 12 fs) . In comparison, from Figure 2(D) of the manuscript, we observe that harmonic generation in the gas phase occurs for intensities larger than 3.8×10^{13} W/cm^2. Therefore the spectrum acquired below this threshold intensity from the flat-jet comprises the harmonic signal from the liquid alone. For a higher intensity of 4.15×10^{13} W/cm^2, we present the raw MCP spectrum emitted from the liquid jet (Figure S2(A)) and the gas only spectrum (Figure S2 (B)) for identical experimental parameters. As can be seen from the image, in the low harmonic energy range (up to 600 pixels) the liquid phase signal is at least an order of magnitude higher than the maximum gas-phase signal. The gas-phase signal only becomes comparable and competing for high energy harmonics (beyond 600 pixels in Figure S2 (A) and (B)).

To discuss this in terms of the harmonic energy, we extract the spectra from Figure S2 (A) and (B) and present in Figure S2 (C) at a few different intensities. The blue line shows the harmonic signal from the liquid jet and the orange line denotes the harmonic signal from gas only and the yellow line shows the spectrum obtained from the liquid only (liquid jet - gas only). Here the gas-phase high-harmonic signal generated by the heated bubbler target density is significantly higher and therefore the signal is detectable as compared to that generated by evaporative cooling of the liquid jet at a laterally shifted position. Even then as can be seen, due to the difference in magnitude of the emitted harmonic signal, the cut-off energy $\sim$ 11.3 eV (end of plateau, following the Lewenstein definition) remains unaffected by the addition of the gas phase signals. The harmonics beyond $\sim$ 17 eV in the gas phase and liquid phase signals are comparable. A rudimentary subtraction of the gas signal from the liquid jet signal, represented by the yellow plots curves in Figure S2(C) shows that the position of liquid cut -off energy (end of plateau) is unaffected by the gas signal, However, the same cannot be concluded for the maximum harmonic energy obtained from the liquid jet signal.

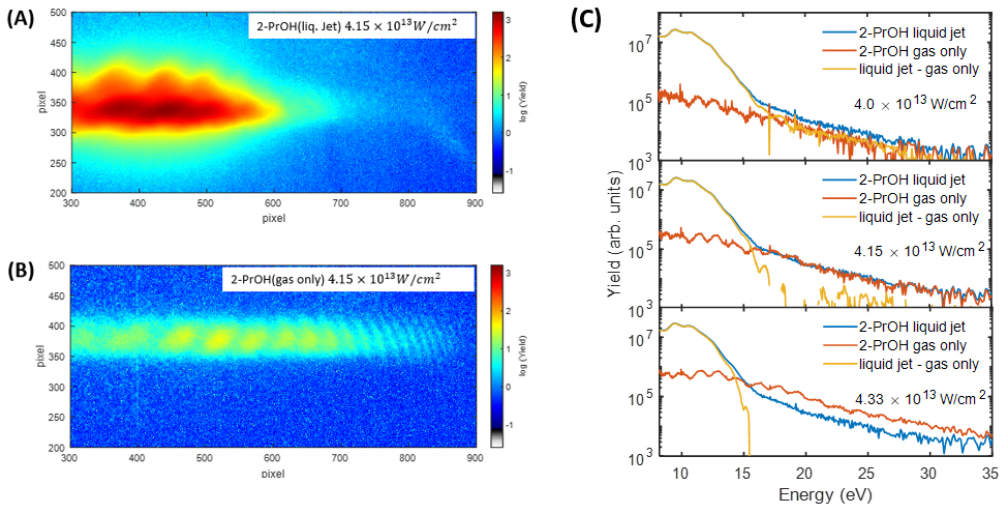

Fig. S2. (A) Shows the raw MCP image of the high-harmonic spectrum of isopropanol generated in the few-cycle regime from the liquid jet. (B) Shows the raw MCP image of the high-harmonic spectrum of gas-phase isopropanol (from bubbler) generated in the few-cycle regime under identical experimental conditions. (C) Shows the comparison of the high-harmonic spectrum of isopropanol from liquid jet (blue line), the high-harmonic spectrum of gas-phase isopropanol (orange line) for a few different intensities. The difference of these two spectra is shown by the yellow line. The middle panel corresponds to the spectra extracted from Figure S2(A-B).

Higher-order non-linear effects from fused silica on harmonic peaks

In our experiment the control of pulse duration was achieved by adjusting the spectral phase of the driving pulse using a pair of fused-silica wedges. The controllable amount of glass was introduced to chirp the pulses and therefore stretch it in time. Although the predominant effect of the glass is the 2nd order phase that yields uniform elongation of the pulse in time, the higher order phase plays a non-negligible role for the broad few-cycle pulses used in this experiment. The combination of residual phase due to self-steepening during the SPM process [1], as well as higher order phase due to glass have an effect on the central wavelength of the produced high-order harmonics. To illustrate this effect in a transparent way, we simulated the spectral response of 2nd and 3rd order process by taking into account the measured few-cycle electric field in time (including the temporal phase) and calculating the FFT[E(t)^2]. Here E(t) is the complex amplitude of the field retrieved from the FROG measurements. Due to the combination of 2nd- and higher-order spectral phase, the effective central wavelength of the compressed part in the spectrum can vary slightly when varying the amount of material. The slight amount of central-wavelength shift leads to the shift of the central frequency of the harmonic of order n by $\Delta\omega_n \sim n\Delta\omega$.

Figure 3(B) shows the HHG spectra in isopropanol for a range of pulse durations. The HHG spectra indicate a blue-shift with increasing pulse width. This effect is consistent with simulated SHG and THG spectra taking into account the pulse shapes obtained from measured and reconstructed FROG traces, shown in Figure S3.

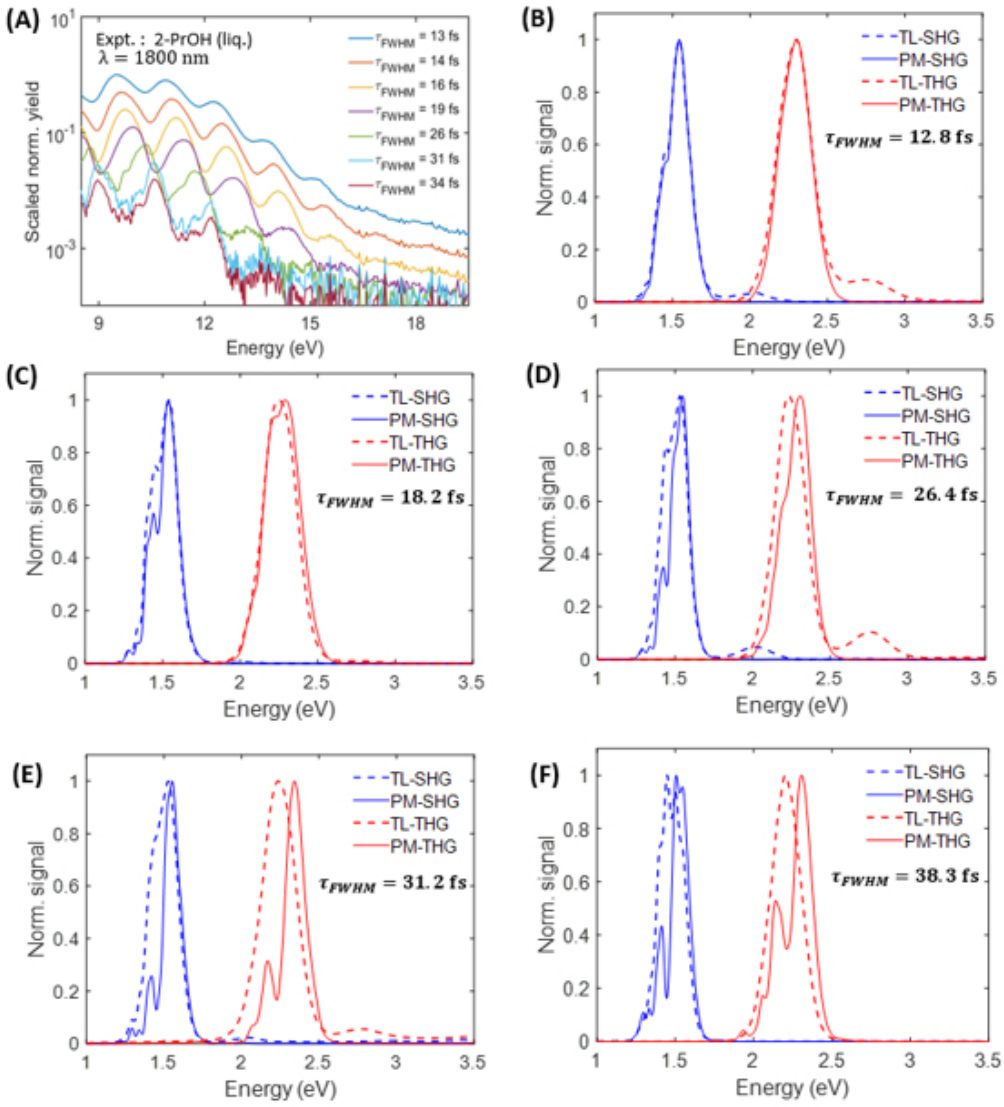

Fig. S3. (A) Shows the pulse duration on liquid 2-PrOH scan presented in Figure 3(B) of the manuscript. Here we observe a red shift of the harmonic peaks with decreasing pulse duration (increasing intensity) indicating that the peak shift effect arises from the slow drift of CEP and not from plasma induced changes. (B)-(F) Calculated spectra of 2nd (SHG) and 3rd (THG) order processes taking into account the instantaneous frequency of the pulse. For comparison, the respective spectra were also calculated assuming a flat spectral phase (no phase modulation) that is denoted as "TL". In the case of 12.8 fs compressed pulses (panel B), the SHG and THG spectra quite closely match the TL calculated spectra thus indicating no significant shift of the central frequency. When an increasing amount of glass is inserted (panels C-F) to stretch the pulses, the SHG and THG spectra exhibit blue-shifting which is consistent with the observations in the HHG spectra (panel A).

Absorption curve of H_2O

Figure S4(A) shows the absorption curve of liquid water. Comparing high harmonic spectra of liquid heavy water shown in Figure S4(B), we do not observe any absorption edge at 14 eV that

can explain the observed cut-off energy.

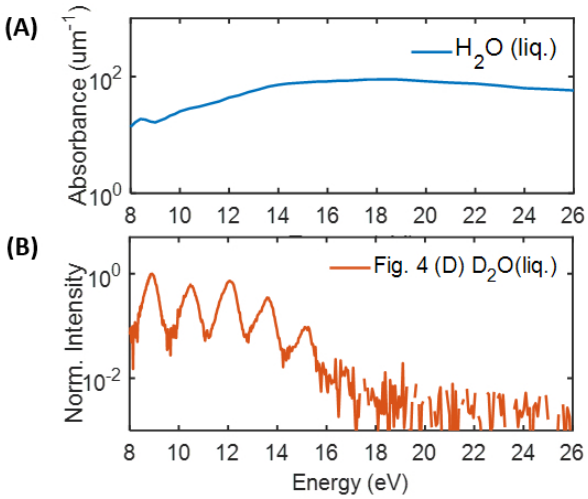

Fig. S4. (A) Absorbance curve of liquid water taken from calculated from $\alpha(E) = 4\pi E$k(E)/(hc), where E is the photon energy, k(E) is the imaginary part of the refractive index of liquid phase water as a function of photon energy, taken from [2] , h is the planck's constant and c is the speed of light. (B) high-harmonic spectrum generated in liquid heavy water by an 1800-nm driving laser (bottom, taken from Figure 4(D), orange line in the present manuscript).

Perturbative regime of harmonic spectrum

Figure S5 shows the high harmonic spectrum of liquid ethanol extending as low as 5 eV. The blue shaded region indicate the pertubative regime.

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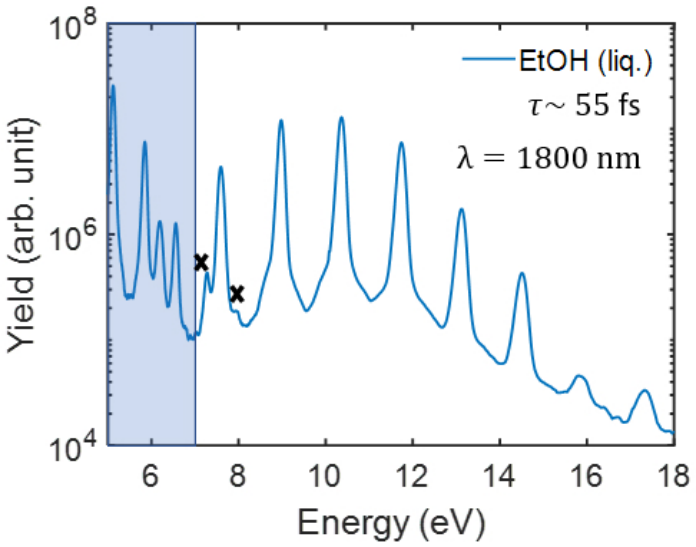

Fig. S5. Shows an experimental spectrum of liquid EtOH where the perturbative region indicated (blue shaded rectangle) is captured in the MCP. The black crosses mark the second order of higher harmonics.