Unveiling unique ultrafast nonlinearities in liquid-phase high-order harmonic generation: supplementary information

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1 Propagation effects in liquid high-order harmonic generation experiments

To evaluate the influence of propagation effects on liquid high-order harmonic generation, we performed two sets of measurements. First, we measured the incident and transmitted spectra of the 800 nm driving laser before and after it propagated through the liquid flat-sheet. As shown in Figure S1(a) and (b) for ethanol and water, respectively, the transmitted spectra remained nearly identical to the incident spectra across different laser intensities, exhibiting no appreciable spectral broadening or frequency shift. This indicates that the driving laser is not significantly altered by propagation through the liquid flat-sheet.

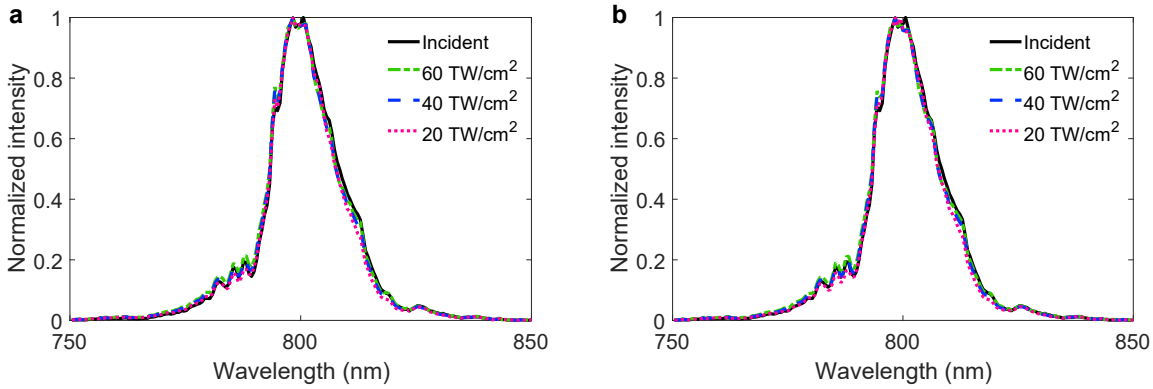

Figure S1: **Spectra of the fundamental driving laser before and after propagation through liquid sheet.** **a**, Results for ethanol. **b**, Results for water. The solid lines represent the incident spectra before propagation, whereas other lines correspond to spectra post-passing through the liquid sheet at different intensities. The in-sample intensities are labeled in the legend. The near-identical profiles suggest that the propagation effects are negligible under the experimental conditions.

On the other hand, we have also measured HHG spectra at different vertical positions d of the laser focus relative to the top edge of the liquid flat-sheet under a fixed laser intensity of 30 TW/cm^2. For the liquid flat-sheet, the thickness of the sheet varies with the vertical position d. If the propagation effect did play an important role in liquid HHG, the generated harmonic spectrum would exhibit a strong sensitivity to the sheet thickness—and hence to the vertical position d. As shown in Figure S2, the measured harmonic spectra for both water and ethanol remain essentially unchanged as the vertical position d varies. All the above results suggest that the spectral redshift and broadening observed in liquid HHG originate not from macroscopic propagation effects but from intrinsic electronic dynamics within the liquid medium.

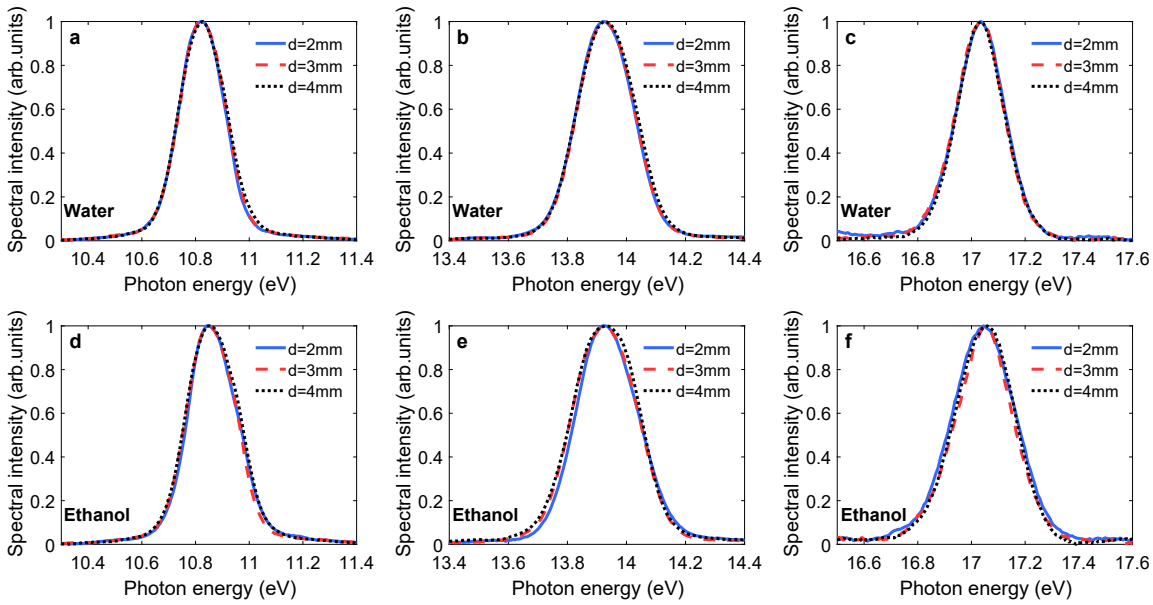

Figure S2: **Measured spectra of HHG in liquids at different vertical position of the laser focus. a-c**, Normalized harmonic spectra of H7-H11 measured for water. **d-f**, Same as **a-c**, but for ethanol.

2 Measured and simulated spectral redshift and broadening for H7 and H9 in liquids

While the main text focuses on H11 as the exemplar, the characteristic spectral redshift and broadening are universal features of liquid-phase HHG. This phenomenon also persists across other harmonic orders like H7 and H9. In this section, we present the measured and simulated results for H7 and H9 to demonstrate this generality. Figure S3 displays the experimentally measured and theoretically simulated harmonic spectra of H7 from liquid water (left panels) and ethanol (right panels) at some specific laser intensities. Corresponding results for H9 are presented in Figure S4 . The results of argon gas are presented for comparison (dashed lines). As shown, the spectra of H7 and H9 in liquids exhibit obvious intensity-dependent redshift and broadening, similar to the results of H11 in the main text.

Besides, we have also quantitatively examined the intensity-dependent frequency redshift of H7 and H9 in liquids. HHG from argon gas serves as a benchmark to evaluate the frequency shift of HHG from the liquidis. Figure S5 (a-b) show the measured redshift of H7 and H9 from liquid water (squares) and ethanol (circles) as a function of the laser intensity. Corresponding simulation

results are shown in Figure S5 (c-d). As the laser intensity increases, both liquids exhibit harmonic redshift, with water showing a greater shift than ethanol. All the experimental measurements are in reasonable agreement with the simulation results.

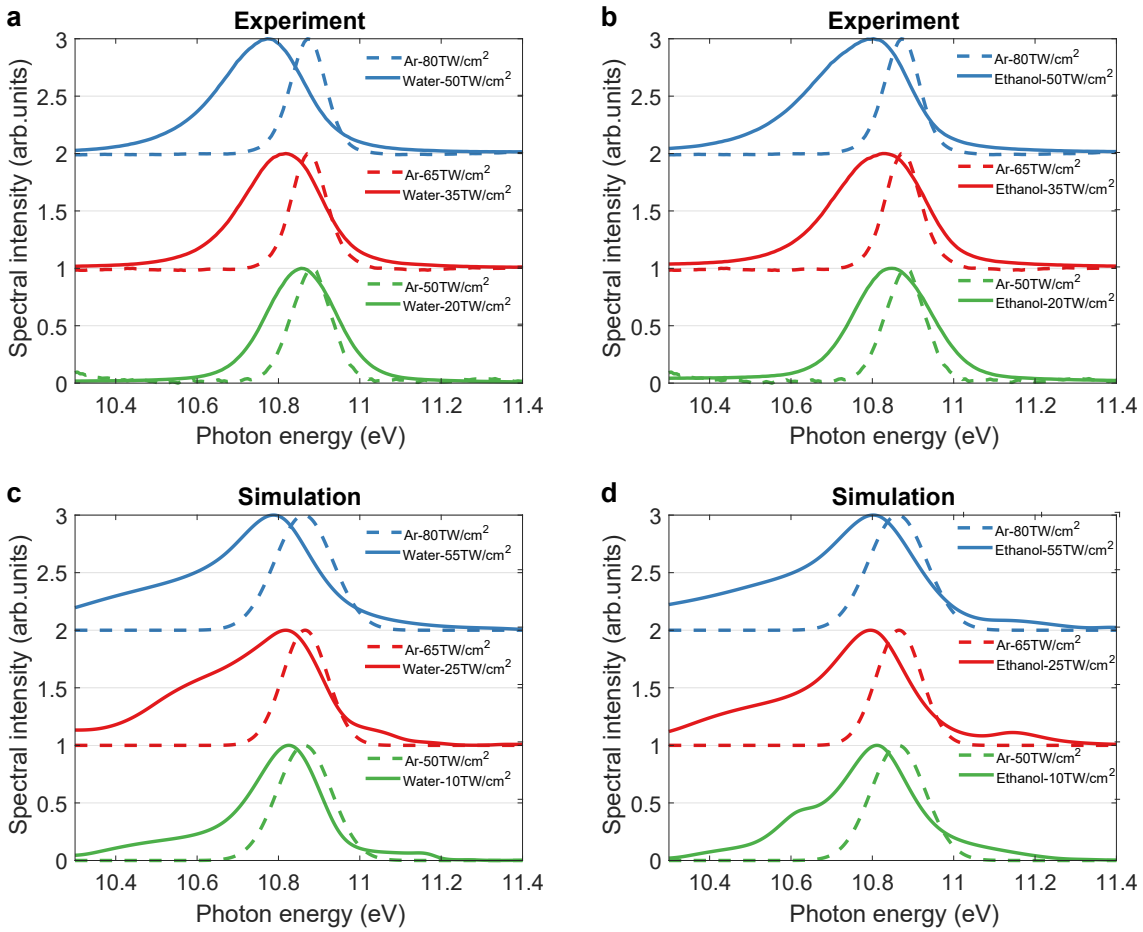

Figure S3: **Measured and simulated spectra of H7 from liquids. a**, Experimentally measured spectra (solid lines) of H7 from liquid water at some specific laser intensities. For comparison, the results of Ar gas are also presented (dashed line). **b**, Same as **a**, but for the liquid ethanol. **c-d**, Same as **a-b**, but for the simulation results. In the simulations, the laser intensities are slightly different from the experiment.

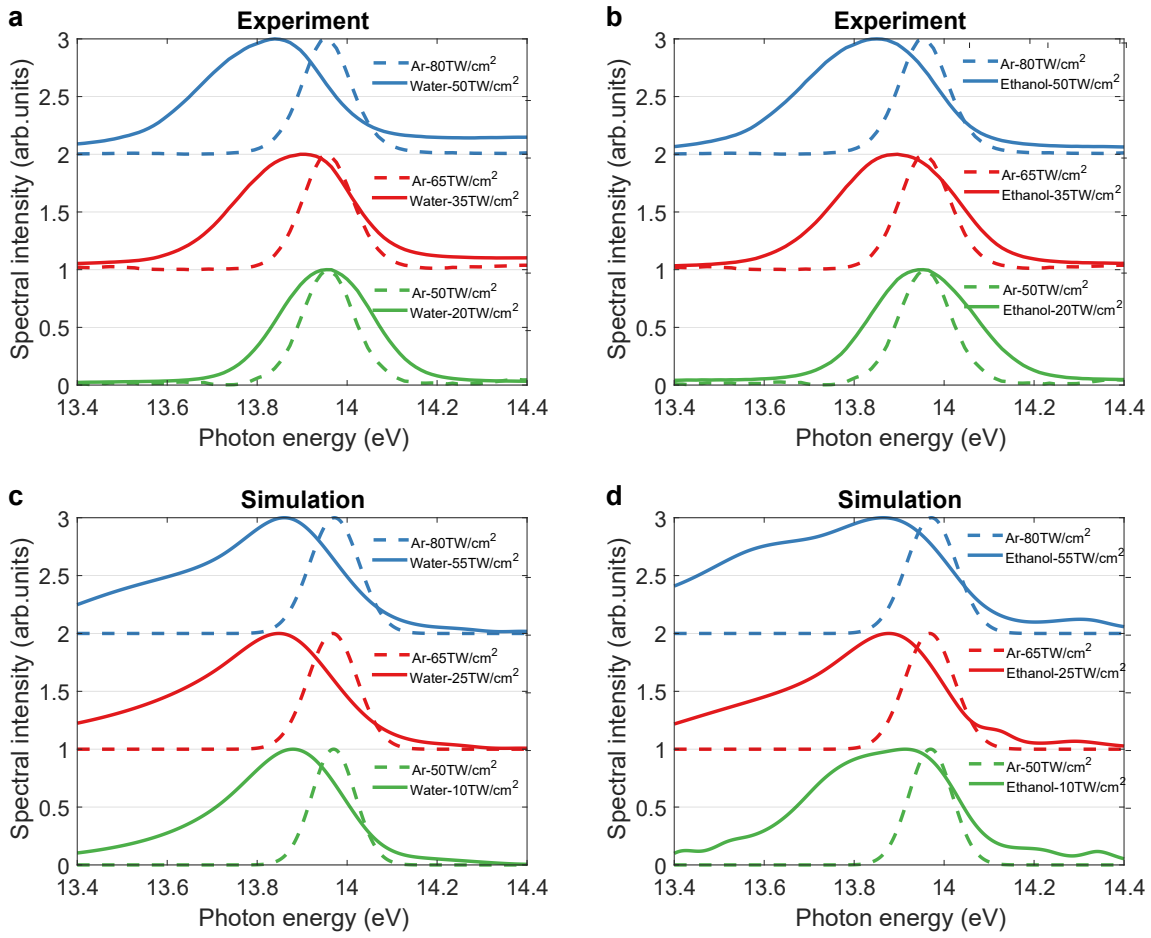

Figure S4: **Measured and simulated spectra of H9 from liquids.** **a**, Experimentally measured spectra (solid lines) of H9 from liquid water at some specific laser intensities. For comparison, the results of Ar gas are also presented (dashed line). **b**, Same as **a**, but for the liquid ethanol. **c-d**, Same as **a-b**, but for the simulation results. In the simulations, the laser intensities are slightly different from the experiment.

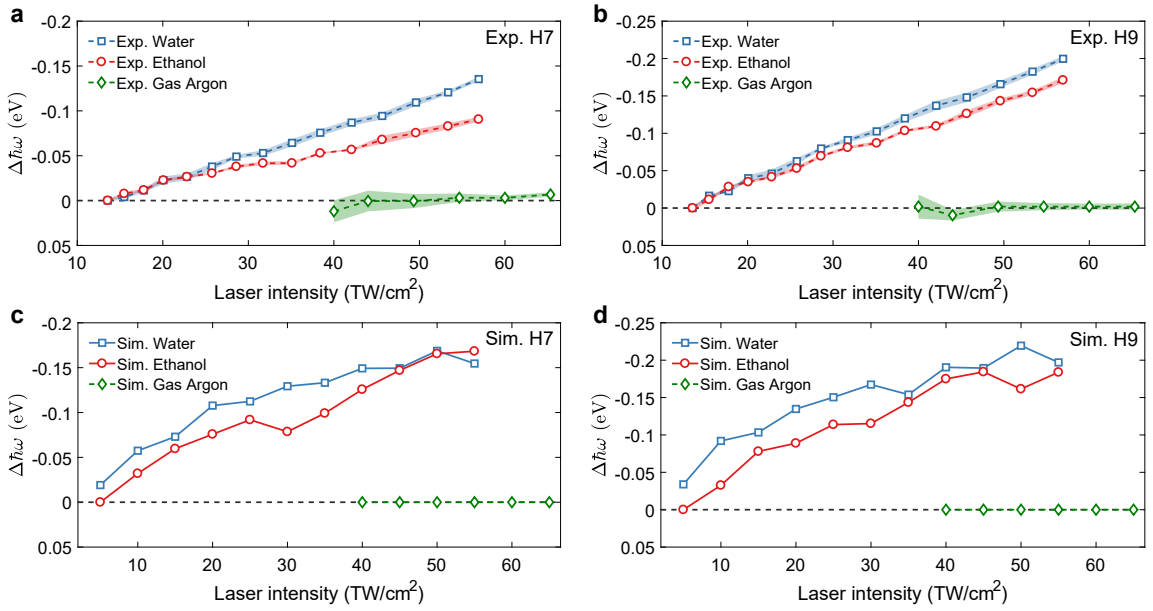

Figure S5: **Laser-intensity-dependent spectral redshift of HHG from liquids and gas. a,** Experimentally measured spectral redshift of H7 from liquid water (squares) , ethanol (circles), and gas Ar (diamonds) as a function of the laser intensity. **b,** Same as **a**, but for the experiment results of H9. **c-d,** Same as **a-b**, but for the simulation results of H7 and H9.