Attosecond time delay in harmonic emissions of H_2 and D_2

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Synopsis We have measured the relative phase of high harmonic generated radiation from hydrogen (H_2) and deuterium (D_2) using Gouy phase interferometer. This measurement may uncover the underlying information about the nuclear vibrational dynamics of these isotopes.

When a high intense laser field interacts with an atom or molecule, it deforms their potential barrier and electron is ionized. The ionized electron then accelerated away from their parent ion. Finally, as soon as the electric field reverses its direction, the electron accelerates back to its parent ion, recombines and emits high-energy extreme ultraviolet (XUV) photons at odd multiples of fundamental laser frequency. This process is known as high harmonic generation (HHG) and it produces attosecond pulses. In the last 12 years imaging of molecule by HHG attracted a lot of interest.

The amplitude and phase of HHG emission provides a snapshot of the structure and dynamics of the recombining system. A recently developed technique analyse nuclear dynamics with a temporal resolution of roughly 100 attoseconds by measuring HHG intensity ratio of hydrogen molecular isotopes [1]. However, to fully characterize a wavefunction we need both the intensity and phase information. Because of the very short wavelength of HHG radiation it is quite difficult to measure its phase.

Gouy phase interferometer produces XUV pulses in two adjacent gas jets with a variable time delay depending on the separation between the jets in a focus of the Gaussian laser beam. It has <100 zeptoseconds (10^{-21}s) temporal resolution [2]. Thus it can detect up to micro-radian phase shift. The Interferometer consists of 2 individual gas jets; bottom one is fixed in position and top one is movable along 3 translational axes. First we pass H_2 in the top jet and D_2 in bottom and then extract the normalized intensity I_{N1}. Then, by passing D_2 in top jet and H_2 in bottom find I_{N2}. The HHG phase difference between H_2 and D_2 is then given by

$$\phi_{H_2D_2} = \frac{\Delta I_N}{\sin(q\phi_{Gouy})} \qquad (1)$$

Where, $\Delta I_N = I_{N2} - I_{N1}$ and $\sin(q\phi_{Gouy})$ is the derivative of normalized intensity for specific separation between the jets when only Gouy phase dominates. Fig.1 shows the phase shift in radian between H_2 and D_2 at two different gas jet separations. That phase shift is nearly constant and equals ~250 mrad for harmonic orders 23 to 33. Reversing the gas order means gas lines are physically altered and this measurement is taken to account for any pressure difference or gas dynamics which could affect our results. It gives a negative phase shift of similar magnitude.

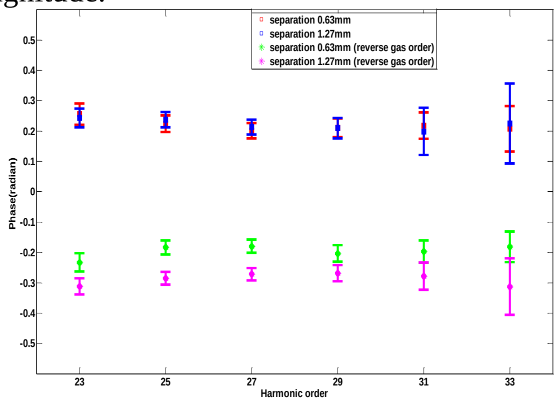

Figure 1. Phase shift between H_2 and D_2

This phase shift corresponds to a time delay of approximately 4 attoseconds between emissions from the two molecular isotopes. We are working on theoretical modeling to explain the nature of this delay.

References

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