

Attosecond delays of high harmonic emissions from isotopes of molecular hydrogen measured by Gouy phase XUV interferometer

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Synopsis We present a precise measurement of HHG phase difference between two isotopes of molecular hydrogen using the advanced extreme-ultraviolet (XUV) Gouy phase interferometer [1]. The measured phase difference is about 200 mrad, corresponding to ~ 3 attoseconds time delay which is nearly independent of harmonic order. The measurements agree very well with numerical calculations of a four-dimensional time-dependent Schrödinger equation. Numerical simulations also reveal the effects of molecular orientation and intra-molecular two-centre interference on the measured phase differences. This technique enables the observation of subtle effects of molecular structures and nuclear motion on electron dynamics in strong laser fields.

High harmonic spectroscopy can access structural and dynamical information on molecular systems encoded in amplitude and phase of high harmonic generation (HHG) signals⁴. However, measurement of the harmonic phase is a daunting task. Building an interferometer in the XUV region is quite challenging for two reasons: firstly, it is challenging to control the delay of the XUV pulses precisely between the two arms with sub-cycle precision; secondly, the highly reflective XUV optics is yet to be developed. Our passively stabilized Gouy phase interferometer [1,2], on the other hand, is an all-optical direct XUV interferometric technique. It does not require calibration of gas pressures to ensure the same number densities. Additionally, it does not require any XUV optics. The technique provides an elegant way to generate two coherent high harmonic pulses without splitting the driving laser and XUV beams. The two mutually coherent XUV pulses are generated by exploiting the inherent properties (Gouy phase) of a single Gaussian focused laser beam. Its unprecedented resolution of 300 μrad (~ 100 zeptoseconds) is a result of any instability in the distance between the two arms (the gas jets in this case) of the interferometer being determined relative to the Rayleigh length, z_R of the fundamental laser beam as opposed to the XUV wavelength in a conventional optical interferometer.

Here, we apply the technique to investigate the effect of nuclear dynamics on the electron motion in molecular hydrogen by precise measurement of high harmonic phase difference (and corresponding HHG phase delays) produced in H_2 and D_2 . Since the ionization potentials of H_2 ($I_p = 15.43$ eV) and D_2 ($I_p =$

15.46 eV) are almost identical, the difference in the phase accumulated by electron in the continuum is considered to be negligible. However, due to the nuclear mass difference, the evolution of nuclear wave packet while electron propagates in the continuum and then recombines to the ground state may differ substantially. The harmonic intensity from the heavy isotope was shown to be higher compared to the lighter molecule as harmonic emission is sensitive to the nuclear motion, though for D_2 the ionization probability is less than for H_2 . The aim of this work is to measure a small phase difference of HHG signals and to gain an insight into the correlated electron–nuclear dynamics for the two isotopes of molecular hydrogen.

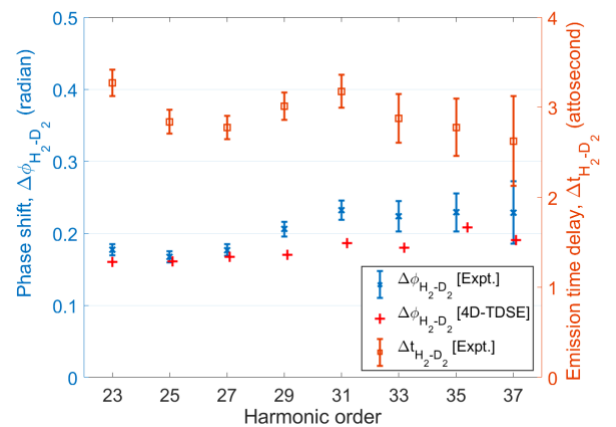


Figure 1. Phase differences and delays between H_2 and D_2 .

References

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