Effect of Nuclear Mass in Strong-Field Ionization of Hydrogen Molecules and Dissociation of Hydrogen Molecular Ions

Igor V. Litvinyuk, Xiaoshan Wang, Han Xu, D. Kiepienski, R.T. Sang

Centre for Quantum Dynamics, Griffith University, Nathan, Queensland 4111, Australia

E-mail: i.litvinyuk@griffith.edu.au

Synopsis

We measure isotope effect in strong-field ionization and dissociation of molecular hydrogen using a mixed-gas target in a Reaction Microscope (REMI). We compare experimental results with theoretical predictions.

In intense-field physics, a quantitative comparison of theoretical predictions with results of experimental measurements – necessary to validate any theory – often remains problematic. The difficulties arise most commonly from imprecise knowledge of laser (intensity, pulse duration, focal size), detector (acceptance angle, efficiency, resolution) and target (density, beam size) parameters. Even when all those parameters are known with sufficient precision, a proper comparison requires also careful accounting for focal and target geometry in combination with a realistic modeling of the detector. Most of those difficulties could be avoided by comparing measurements performed on different isotopes, i.e. light and heavy hydrogen, under exactly the same conditions. The relative isotope effect could be largely insensitive to variations of unknown or unstable parameters and may be then readily compared with predictions of various theoretical models.

Here we present two specific examples of experimental measurement of isotope effects observed in light and heavy hydrogen molecules – one validating a theory and another one revealing inadequacy of a model. To ensure that the measurements were performed under exactly the same conditions for H₂ and D₂, we used a 50:50 gas mixture as a target with REMI detection, which allowed us to mass-select the two isotopes.

In the first example, we measured single ionization yields for H₂ and D₂ for a number of different intensities. It has been recently predicted theoretically by Tolstikhin, Worner and Morishita [1] that tunneling ionization rate for neutral hydrogen molecules should exhibit a significant isotope effect, which becomes more pronounced at lower intensities. Since a significant fraction of singly ionized molecules also undergo dissociation (and also double ionization), which could itself exhibit an isotope effect, proper accounting for all final channels was necessary for a quantitative comparison. We performed the measurements using circularly polarized pulses and coincident REMI detection of protons, deuterons and electrons. We used ion detection to ascertain the relative ionization yields and coincident ion-electron detection to measure angular anisotropy of ionization for both H₂ and D₂. We found excellent agreement with predictions of Tolstikhin et al. with hydrogen more likely to be ionized (up to 1.4 times more likely at the lowest intensity) than deuterium.

In the second example, we measured CEP-dependent asymmetry of proton (deuteron) ejection in dissociation of H₂⁺ (D₂⁺) as a function of the fragment kinetic energy. We measured the highest asymmetry reported to date – 0.5 for H₂ and 0.4 for D₂. We compared our experimental results to calculations of a two-state model. While taken separately, both hydrogen and deuterium show excellent agreement with model calculations up to the unknown absolute CEP calibration. However, a side-by-side comparison reveals a significant discrepancy between the experimentally observed and calculated relative phase of the CEP dependence of asymmetry. Clearly, the theoretical model, while adequately reproducing the magnitude and energy dependence of CEP-dependent asymmetry, fails to properly account for its phase.

References

1. O.I. Tolstikhin, H.J. Warner and T. Morishita 2013 Phys. Rev. A 87 041401(R)

i E-mail: i.litvinyuk@griffith.edu.au