Dynamical origin of below- and near-threshold harmonic generation of H_2^+ in an intense NIR laser field

John Heslar[*1] **and Shih-I Chu**[†,‡]

[*,†] Center for Quantum Science and Engineering, Department of Physics, National Taiwan University, Taipei 10617, Taiwan

[‡] Department of Chemistry, University of Kansas, Lawrence, Kansas 66045, USA

Synopsis We perform a fully *ab initio* and high precision 3D quantum study of the below- and near-threshold harmonic generation of H_2^+ molecules in an intense 800-nm near-infrared (NIR) laser field. Combining with a synchrosqueezing transform of the quantum time-frequency spectrum and an extended semiclassical analysis, we explore in-depth the roles of various quantum trajectories, including short- and long trajectories, multiphoton trajectories, resonance-enhanced trajectories, and multiple rescattering trajectories of the below- and near- threshold harmonic generation processes.

High-order-harmonic generation (HHG) is a fundamental atomic and molecular process in strong laser fields that continues to receive considerable attention and it plays a crucial role in the development of ultrafast science and technology. Significant application of the HHG technology includes the generation of ultrashort attosecond pulses and ultrafast molecular imaging, etc., to name only a few. The availability of the attosecond pulse further allows the direct detection and control of the electronic dynamics in atoms, molecules and condensed matter systems.

In the past, major attention was focused on the HHG regime well above the ionization threshold where the semiclassical three-step model and strong field approximation (SFA) are effective to explain the process. More recently considerable attention has been paid to the near- and below-threshold regimes as a potential source of coherent vacuum-ultraviolet radiation. In these lower-energy regimes, the conventional three-step model and the SFA become inadequate, since they neglect the Coulomb potential and the detailed electronic structure of the target atom/molecule. As we have recently demonstrated along with the experimentalists [1], phase-matched below-threshold harmonics can be generated only near the resonance structures of the atomic target.

In this work, we present the first *ab initio* precision study of the below- and near-threshold harmonic generation of the H_2^+ molecule in intense near-infrared laser field by solving the three-dimensional two-centered time-dependent Schrödinger equation (TDSE) accurately and efficiently by means of the time-dependent generalized pseudospectral method (TDGPS) in prolate spheroidal coordinates. In addition, we employ a new synchrosqueezing transform (SST) to analyse the time-frequency spectra of the below-, near- and above-threshold HHG of H_2^+. By comparing the SST time-frequency spectra and the extended semiclassical calculations (Fig. 1), we unravel for the first time the contributions of the short trajectories, long trajectories, multi-scattering trajectories, “resonant" or bounded trajectories, and “continuum" trajectories in the below-, near- and above-threshold harmonic generation, and demonstrate the features of the spectral dynamical phase for the H_2^+ molecule [2].

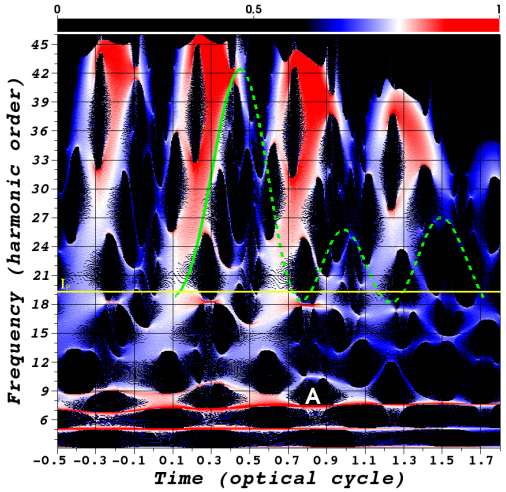

Figure 1. SST time-frequency analysis of the HHG spectra of H_2^+ and semiclassical trajectories [green curves (both solid lines and dashed)].

References

[1] M. Chini, X. Wang, Y. Cheng, H. Wang, Y. Wu, E. Cunningham, P.-C. Li, J. Heslar, D.A. Telnov, S.I. Chu, and Z. Chang, *Nat. Photonics* **8**, 437 (2014).

[2] J. Heslar and S.I. Chu, *Sci. Rep.* **6**, 37774 (2016).

[1]E-mail: john.heslar@gmail.com