

## Quantum path interference in high harmonic generation

E. Brunetti, R. Isaac, D. A. Jaroszynski

*Department of Physics, University of Strathclyde, 107 Rottenrow, G4 0NG Glasgow, United Kingdom.*

High harmonic generation in gas is a nonlinear process that occurs when intense laser radiation interacts with matters. When atoms are exposed to fields in excess of  $10^{13}\text{W}/\text{cm}^2$ , electrons are removed from the nucleus, accelerated to very high speed and then driven back to the parent ion, where they can recombine. Photons with frequencies corresponding to odd multiples of the laser central frequency are then emitted and up to 300 harmonics have been observed. Therefore, high harmonic generation is a promising method to generate coherent radiation in the UV and soft x-ray regions, as well as to produce attosecond pulses [2].

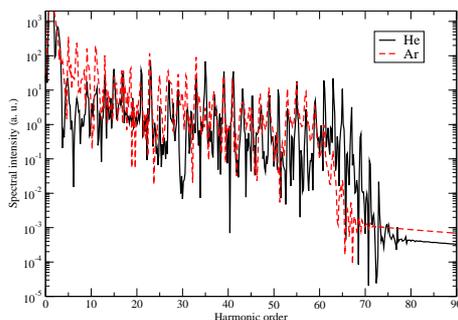


Figure 1: High harmonic spectra for Ar and He obtained by numerically solving the Schrödinger equation at the intensity of  $4 \times 10^{14} \text{ W}/\text{cm}^2$ .

A typical harmonic spectrum is shown in Fig. 1 for two different gases at the intensity of  $4 \times 10^{14} \text{ W}/\text{cm}^2$ . It has been obtained by numerically solving the 1-D Schrödinger equation for an electron subject to a strong electromagnetic field and to the atomic binding potential. It consists of a plateau region, where the harmonic peaks have approximately constant intensity, followed by a cutoff region, where emission ceases abruptly. The existence of the cutoff can be interpreted in term of the re-collision picture as the maximum energy that the electrons can gain, beyond which they do not return to the nucleus. Neglecting trajectories longer than one laser cycle, there is only one electron trajectory generating harmonics in the cutoff region, while two different trajectories exist in the plateau region, as shown in fig. 2.

Recent experimental and theoretical work have however proved that the weight of the different trajectories is not correctly evaluated by this semi-classical model. Fine structure in the angular distribution of above-threshold ionisation (ATI) photoelectrons, the complementary process to harmonic generation, where emitted electrons rather than

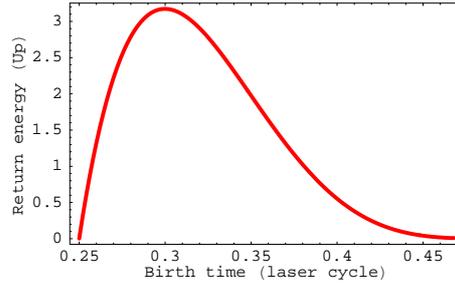


Figure 2: Electron return energy for different birth times. The maximum energy is  $3.2U_p$ , corresponding to the cut-off harmonics.

photons are observed [4], broad harmonic spectral profile [5] and the existence of regions with different coherence times in interference patterns from high harmonic radiation [1], have stimulated the development of a fully quantum mechanical description of laser-atom interaction. In this framework, the electron trajectories are described by means of quantum orbits, with their real part corresponding to the semi-classical electron trajectories and their imaginary part relating to the probability of ionisation.

The phase of the harmonics along the propagation axis follows the intensity profile of the laser beam and is symmetric with respect to the focal position. On the other hand, neglecting ionisation effects, the phase of the laser is dominated by the Guoy shift, which introduces an asymmetry. For a focus after the medium, the shortest trajectory has the slowest phase fluctuations and consequently is selected by phase matching [6]. On the other hand, for a focus behind the medium longer trajectories are phase matched. This has important implications for the production of attosecond pulses, since the selection of one trajectory also locks the relative phase of the harmonics, leading to the production of a train of attosecond pulses after propagation through the medium [3].

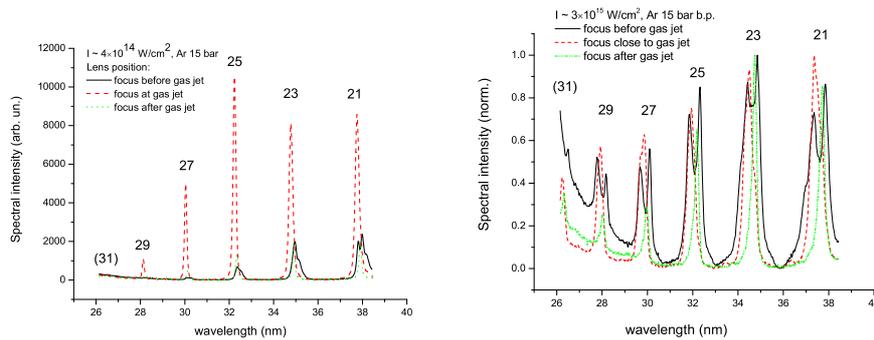


Figure 3: Harmonic spectra measured in an Ar gas jet with a backing pressure of 15 bar for a 60 fs laser pulse with wavelength of 800 nm and an intensity of  $4 \times 10^{14} \text{ W/cm}^2$  (left) and  $3 \times 10^{15} \text{ W/cm}^2$  (right). Different curves correspond to different relative positions between the focus of the laser beam and the gas jet.

We have performed an experiment to observe the contribution of different quantum

paths using a 60 fs Ti:sapphire laser system at the TOPS facility, which delivers pulses at a repetition rate of 10 Hz and a central wavelength of 800 nm. The radiation is focussed with a 50 cm focal length lens into an Ar gas jet, with an interaction length of about 1 mm. Harmonics are detected using a soft x-ray spectrometer<sup>1</sup> consisting of a gold overcoated concave aperiodic grating for resolving XUV radiation between 5 and 105 nm, and a CCD camera. The grating focuses the radiation only along the wavelength axis, thus allowing the angular profile of the harmonics to be observed. A 0.8  $\mu\text{m}$  thick Al filter, which transmits about 15% in the wavelength range 25 – 60 nm, is located before the grating, to prevent radiation from the laser and low harmonics from reaching and saturating the camera.

The spectra in fig 3 show the measured far-field profile of harmonics between the 21<sup>st</sup> and 31<sup>st</sup> order, produced for intensities of  $4 \times 10^{14}$  W/cm<sup>2</sup> and  $3 \times 10^{15}$  W/cm<sup>2</sup>. Spectra for three different lens positions are shown, corresponding to the laser beam focused about 0.5 cm after the gas jet, at the gas jet and 0.5 cm before the gas jet. The shape of the harmonic peaks in the three cases are remarkably different, especially for the lower orders, where the lines are considerably broader and split into a number of components. The bandwidth of the harmonics has a width of a few tens of nanometres when the focus is after the gas jet and is broadened to about 1 nm when the focus is moved to before the gas jet, as expected due to the larger chirp associated with quantum paths that have longer return times.

Selecting the shortest trajectory is important not only because of the better quality and coherence of the generated radiation, but is also essential for the production of phase-locked harmonics, which is required to generate attosecond pulses.

## References

- [1] M Bellini, C Lyngå, A Tozzi, M B Gaarde, T W Hänsch, A L’Huillier, and C-G Wahlström. Temporal coherence of ultrashort high-order harmonic pulses. *Physical Review Letters*, 81(2):297–300, 1998.
- [2] Thomas Brabec and Ferenc Krausz. Intense few-cycle laser fields: Frontiers of nonlinear optics. *Review of Modern Physics*, 72(2):545–591, April 2000.
- [3] P M Paul, E. S. Toma, P Breger, G. Mullot, F. Augé, Ph. Balcou, H. G. Muller, and P. Agostini. Observation of a train of attosecond pulses from high harmonic generation. *Science*, 292:1689–1692, 2001.

---

<sup>1</sup>The spectrometer has been designed by David Neely

- [4] G G Paulus, F Grasbon, A Dreischuh, H Walther, R Kopold, and W Becker. Above-threshold ionization by an elliptically polarized field: interplay between electronic quantum trajectories. *Physical Review Letters*, 84(17):3791–3794, 2000.
- [5] P Salières, B Carré, L Le Déroff, F Grasbon, G G Paulus, H Walther, R Kopold, W Becker, D B Milošević, A Sanpera, and M Lewenstein. Feynman’s path-integral approach for intense-laser-atom interactions. *Science*, 292:902–905, 2001.
- [6] P Salières, A L’Huillier, P Antoine, and M Lewenstein. Study of the spatial and temporal coherence of high-order harmonics. *Advances in atomic, molecular, and optical physics*, 4:83–142, 1999.