Feynman’s path integral analysis of high harmonic phases

In the spirit of Feynman’s path integrals, each harmonic is characterised by a complex amplitude $D_q = A_q e^{i\phi(q)}$ which is a sum over all quantum paths (orbits) that can contribute to the harmonic emission: $D_q = \sum_n a_n e^{iS[r_n(t_i,t_r,\vec{p})]}$. The $n^{th}$ quantum orbit is defined by an amplitude $a_n$ and a phase given by the action along the path $r_n(t_i,t_r,\vec{p})$:

$$S[r_n(t_i,t_r,\vec{p})] = q\omega t + \int_{t_i}^{t_r} \left[ \frac{(\vec{p} + \vec{A}(t))^2}{2} + I_p \right] dt, \quad (1)$$

where $I_p$ is the atom ionisation potential, $\vec{p}$ the electron drift momentum and $\vec{A}(t)$ the vector potential associated with the electric field. An infinite number of quantum orbits contribute to $D_q$. However, in the strong field regime ($S1$, $S2$), the sum reduces to a few closed quantum orbits which render the phase stationary, meaning that they satisfy the classical least action principle $\delta S[r_n(t_i,t_r,\vec{p})] = 0$. Partial differentiation of the action with respect to $t_i$, $t_r$ and $\vec{p}$ gives the following saddle point equations:

ionisation: $\left. \frac{(\vec{p} + \vec{A}(t_i))^2}{2} + I_p \right| = 0, \quad (2)$
The physical content of these equations coincides with the three-step model, as labelled above. In order to identify the quantum orbits associated with the emission of a given harmonic order, the nonlinear set of equations (2)–(4) must be solved. As $I_p$ is positive, the pair of solution $(t_i, t_r)$ is complex. The imaginary part of the time has been invoked as a trace of the tunnelling process responsible for the ejection of the electron. It becomes all the more important as the electron kinetic energy (i.e. the harmonic order) increases ($S3$). Once $t_i$ and $t_r$ are determined, we evaluate the harmonic phase $\varphi$ given by the real part of Eq. 1.

Recognising that $t_i$, $t_r$ and $\vec{p}$ are functions of the order $q$, and thus of the frequency, we can derive the expression:

$$\frac{\partial \varphi}{\partial \omega} = \frac{\partial t_i}{\partial \omega} \frac{\partial \varphi}{\partial t_i} + \frac{\partial \varphi}{\partial \omega} \frac{\partial t_i}{\partial \varphi} + \frac{\partial \varphi}{\partial \omega} \frac{\partial \varphi}{\partial \vec{p}} \frac{\partial \varphi}{\partial \vec{p}}.$$ 

Thus, with the help of the saddle point equations (2)–(4) and the expression of the harmonic phase (1), we can show that:

$$t_e = \frac{\partial \varphi}{\partial \omega} = \Re(t_i).$$

We checked that these calculations are in good agreement with the emission times extracted from numerical solution of the time-dependent Schrödinger equation. Note that pure classical trajectory simulations do not provide correct emission times.

**Relative phase measurements through two-photon two-colour photoionisation**

When an atom interacts with an XUV field, it can be ionised, producing electrons of energy
The photoelectron spectrum given by a high order harmonic beam focused in a detecting gas jet produces peaks corresponding to harmonics whose energies are above the ionisation potential of the detecting gas. If one adds a weak IR field to the harmonic field, then a two-photon, two-colour ionisation can occur. New peaks, called sidebands, appear on the photoelectron spectrum exactly halfway between the harmonics. The key point is that two main quantum paths can lead to the same sideband: (i) absorption of a photon of harmonic \( q \) (H\(_q\)) plus one infrared photon, and (ii) absorption of an H\(_{q+2}\) photon and emission of one IR photon. These two paths interfere, and the sideband amplitude contains an oscillatory term which can be written as

\[
S_{q+1} = S_{q+1}^0 \cos(2\phi_{\text{IR}} + \phi_q - \phi_{q+2} + \Delta\phi_{q+1}^{\text{at}}).
\]

Note that the phases involved in this formula are evaluated in the detecting gas jet, and not at the point of generation. Propagation from the generation to the detection point could in principle affect them (S4). However, in our case, additional phase shifts due to geometrical dispersion and atomic dispersion by the residual gas have a negligible effect. The atomic phases \( \Delta\phi_{q+1}^{\text{at}} \) are calculated from the theory (S4). By delaying the IR beam with respect to the harmonic beam, one sets \( \varphi_{\text{IR}} = \omega_0 \tau \), and the sideband intensity oscillates at twice the laser frequency as the delay increases. The phase of its oscillations, corrected by the atomic term, provides the phase difference between two consecutive harmonics and thus the harmonic emission time:

\[
t_c(\omega_{q+1}) = \frac{1}{2\omega_0} (\Phi_{\text{SB}}(\omega_{q+1}) + \Delta\phi_{q+1}^{\text{at}}).
\]

The atomic phase contribution has only a small effect on these calculations (Fig. S1).
Experimentally, one must determine precisely $\varphi_{ir}$ in order to access the absolute value of these phase differences, and thus, the absolute timing of the attosecond pulse train with respect to the fundamental laser field. In our setup, the generating and dressing beams are superposed in the generating gas jet and can thus optically interfere, resulting in modulations of the total harmonic signal at the laser frequency as the delay varies. The contrast of the infrared intensity oscillations is small because the probe beam is much less intense than the generating one. However, the high nonlinearity of the HHG process enhances it and allows clear detection of the total signal modulation ($S5$). By calculating the total number of detected electrons as a function of delay, one can access this oscillation and set its phase as the origin for the sidebands phase measurements.

The photoelectron spectrometer resolution decreases with increasing electron energy, making simultaneous measurements of more than ten harmonics difficult. By setting a retardation potential in the spectrometer flight tube, it is possible to slow down the fastest electrons, bringing their energy down to a range where the resolution is good. The slowest electrons are then not detected. Broadband analysis results from consecutive phase measurements of groups of ten sidebands with increasing retardation potential by steps of $10\omega_0$. Each sideband phase is thus measured twice, allowing the reproducibility to be checked.

**Experimental details**

The diameter of the dressing beam was typically 2 mm, but could be adjusted up to 8 mm
by use of a diaphragm placed in the beam-splitting delay line. A half-wave plate and a polariser, also set in the dressing beam arm, allow an intensity control of the dressing beam, independent of the total intensity.

The diameter, energy, and focusing position of the generating beam depend on the generating gas. The measurements in argon were performed with 14 mm diameter, 1.3 mJ pulse energy, and a beam focused 5 mm ahead of the gas jet, resulting in an intensity of $1.2 \times 10^{14}$ W/cm² in the jet (the beam is two times diffraction limited). In the case of neon, the beam diameter was 19 mm, the pulse energy 3.6 mJ and the laser was focused 10 mm in front of the jet, resulting in an intensity of $3.8 \times 10^{14}$ W/cm². In both cases, the intensity value is consistent with the cutoff position observed in the harmonic spectra, according to the $I_p + 3U_p$ cutoff law.

The Pt-coated toroidal mirror is set at grazing angle of 3°. Its focal length is 70 cm. The diaphragm that blocks the infrared annular beam has a 4 mm diameter. Its alignment is checked by a camera placed after the detecting gas jet, ensuring that no infrared light from the generating beam reaches the spectrometer gas jet. The dressing beam on the delay-line can also be blocked and a check made that no sidebands are generated. However, at high pressure in the generating jet and high laser intensity, some of the generating infrared can be scattered on axis and is thus not blocked by the diaphragm. This scattered light contributes to the sidebands generation and induces a modulation at the laser frequency ($S4$). However, the phase of the oscillatory part at $2\omega$, that we extract by FFT, is not affected by this process.

The spectra are averaged on a digital oscilloscope at the laser repetition rate, and then
transferred to a computer for recording and analysis.

References


Fig. S1: Atomic phase contribution to the harmonic emission time calculated in argon (squares) and helium (circles).