Quantum mechanical approach to probing the birth of attosecond pulses using a two-colour field

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Abstract
We investigate the generation of even and odd harmonics using an intense laser and a weak second harmonic field. Our theoretical approach is based on solving the saddle-point equations within the strong field approximation. The phase of the even harmonic oscillation as a function of the delay between the fundamental and second harmonic field is calculated and its variation with energy is found to be in good agreement with recent experimental results. We also find that the relationship between this phase variation and the group delay of the attosecond pulses depends on the intensity and wavelength of the fundamental field as well as the ionization potential of the atom.

(Some figures in this article are in colour only in the electronic version)

1. Introduction
Attosecond pulses [1, 2] are created through the interaction between intense infrared (IR) laser fields and atoms or molecules in a process known as high-order harmonic generation (HHG). This process is initiated by the creation of an electron wave packet through tunnelling ionization, followed by acceleration in the laser field and recombination with the ion core resulting in extreme ultraviolet (XUV) emission confined to a fraction of the laser cycle. The generation process can be controlled by shaping the driving laser field, e.g. by coherently adding laser fields of different wavelengths to the fundamental laser field [3–6]. Most investigations have been carried out by adding the second harmonic generated in a doubling crystal [7–14], with various goals ranging from optimization of the conversion efficiency, characterization of the emitted attosecond pulses and production of attosecond pulse trains with one pulse per cycle of the fundamental field. Several parameters can be varied in these experiments: the intensity ratio, the phase difference and the relative polarization direction.

When the second harmonic field is a weak perturbation to the fundamental, the generation of odd harmonics is barely changed, but the induced symmetry breaking leads to the appearance of weak even harmonics. High-order harmonic spectra recorded as a function of delay between the two fields show that the intensity of the even harmonics is modulated and that this modulation has an offset depending on the harmonic order, as shown in figure 1(a) [13]. These photon spectrograms are at a first glance similar to the photoelectron spectrograms of the RABITT (reconstruction of attosecond beating by interference of two-photon transitions) method [1] used to characterize attosecond pulses. Dudovich et al [16] suggested that the two-colour HHG spectrograms could be used to determine the emission times of the attosecond pulses in situ, thus ‘probing the birth of attosecond pulses’. This method was then applied by Doumy et al [17], using laser systems of different wavelengths, and by us [18], in a direct experimental comparison with the RABITT method.

In this paper we investigate HHG by a laser field and its second harmonic by solving the Schrödinger equation within the strong field approximation (SFA), using the saddle-point equation method [20]. We assume throughout that the second harmonic is weak and can be considered as a small perturbation to the fundamental field. We calculate the phase variation of the even harmonic oscillation and compare it with the harmonic emission times. Our model also allows us to interpret the rapid variation of this phase at high energy, observed in several experiments [13, 18], as a change of the dominant quasiclassical trajectory from the short to the long...
one (figure 1(b)). The paper is organized as follows. In section 2 we review and compare classical and quasiclassical electron trajectories for one-colour (section 2.1) and two-colour HHG (section 2.2). In section 3 we study the generation of even harmonics close to the cutoff (section 3.1) and we relate the phase of the even harmonic oscillation to the emission times of the attosecond pulses (section 3.2). In section 4 we summarize our results by presenting a more general relationship between the phase of the even harmonic oscillation and the emission times.

2. Quasiclassical trajectories

Our method is based on the stationary phase method (also referred to as the saddle-point method) to solve the Schrödinger equation for an atom exposed to a strong laser field using the SFA [19]. In particular, we consider the interaction of an atom with a strong IR laser field (frequency \(\omega\)) and a weak second harmonic (frequency \(2\omega\)). In the SFA, the electron will (i) tunnel into the continuum, (ii) accelerate in the strong laser field and then (iii) return to the atom and emit a harmonic photon (frequency \(\Omega\)). The phase of the harmonic radiation is related to the quasiclassical action of the electron

\[
S(\vec{p}, t, t_0) = \int_{t_0}^{t} dt' \left( \frac{(\vec{p} - e \vec{A}(t'))^2}{2m} + I_p \right),
\]

where \(\vec{A}(t)\) is the vector potential of the laser field, \(I_p\) is the ionization potential of the atom, and \(\vec{p}, t_0, t, m, e\) are the drift momentum, tunnelling time, return time, mass and charge of the electron, respectively. The high-order harmonic emission will mainly originate from the stationary points of \(S - \hbar \Omega \tau\), with respect to all variables \([\vec{p}, t, t_0]\), which satisfy the following three equations [19]:

\[
\int_{t_0}^{t} dt' e \vec{A}(t') = (t - t_0) \vec{p},
\]

\[
\frac{(\vec{p} - e \vec{A}(t_0))^2}{2m} = -I_p,
\]

\[
\frac{(\vec{p} - e \vec{A}(t))^2}{2m} = \hbar \Omega - I_p.
\]

The electron is thus required to return to the atom at time \(t\) (equation (2)), to undergo complex tunnelling at time \(t_0\) (equation (3)) and to satisfy energy conservation (equation (4)). For any realistic atom, we have \(I_p > 0\) which implies that the electron must tunnel into the continuum. The tunnelling process results in damping of the electron trajectories, i.e. complex stationary points in the harmonic plateau [20]. The electron trajectories beyond the cutoff are always strongly damped (also for \(I_p = 0\)), because they are always classically forbidden, corresponding to large imaginary components of the stationary points. The physical emission time of a given harmonic is given by the real part of the complex emission time [15, 21, 22].

2.1. One-colour case

We first consider the one-colour case with a vector potential, \(\vec{A}(t) = A_1 \sin(\omega t)\), where \(\omega\) is the angular frequency of the fundamental laser light. The complex stationary points \([\vec{p}^{(n)}, t^{(n)}, t_0^{(n)}]\) are calculated as a function of the high-order harmonic photon angular frequency, \(\Omega\). The index \(n\) is used to separate different sets of solutions, where \(n = 1\) corresponds to the short branch and \(n = 2\) corresponds to the long branch. A direct comparison between the stationary points for \(I_p = 15.76\) eV (Ar) and \(I_p = 0\) eV (referred to as the classical case) is shown in figure 2. The detailed behaviour of these stationary points will prove to be important not only for the generation of attosecond pulses.

Figure 1. (a) Experimental two-colour high-order harmonic spectrogram over relative delay [13]. The diamonds (○) indicate the experimental delays corresponding to maxima of the modulation in the even harmonic orders. (b) Calculated delays corresponding to maxima of the even harmonic orders. (c) Calculated delays corresponding to experimental delays corresponding to maxima of the modulation in second harmonic (frequency \(2\omega\)) of an atom with a strong IR laser field (frequency \(\omega\)).

Figure 2. Real and imaginary parts of the stationary points, \([\vec{p}, t, t_0]\). Ionization potentials: argon \(I_p^{(Ar)} = 15.76\) eV (thick blue) and classical \(I_p^{(0)} = 0\) eV (thin red). The classical case is shifted by \(1.3I_p\) to match the quantum mechanical cutoff. The short branch is a line; while the long branch is a dashed line. The intensity is \(2 \times 10^{14}\) W cm\(^{-2}\) and the wavelength is 800 nm.
from one-colour HHG but also in the quantitative analysis of two-colour HHG. The classical case corresponds to solving equations (2)–(4) for \( I_p = 0 \). The stationary points are first real, describing classical trajectories in the continuum. Using classical trajectories to explain the HHG process leads to the approximate cutoff law: \( \hbar \Omega_{\text{max}} \approx 3.2 U_p + I_p \) [23, 24]. Harmonics above this cutoff cannot be generated because there are no electrons returning with sufficient kinetic energy. Quantum mechanically, we see that the stationary points quickly develop large imaginary components above energy. Quantum mechanically, we see that the stationary phase solutions overlap almost perfectly in the main branches. This demonstrates the usefulness of the classical model for understanding the intrinsic chirp of the returning electron wave packet and the resulting chirp of the attosecond light pulses. The real parts of the tunnelling times (figure 2(e)) are, however, quite different. In the lower part of the plateau there is a discrepancy of a factor of 2 for the short branch. The short branch trajectories tunnel at earlier times compared to the classical case due to the ionization potential. This comes from the change of sign of the real part of the tunnelling time (figure 2(f)) where the complex part could, however, also include the effect of the energy-dependent atomic scattering phases, \( \eta_l \), in the stationary phase equations (equations (2)–(4)) (from the recombination matrix element \( d(\vec{p} - e\vec{A}(t))' \)). This would lead to a small change in the stationary points, which would be especially interesting to study in argon due to an unusually strong variation of the scattering phase in the present energy region [25, 26].

Assuming the process to be periodic, the total dipole response, \( \vec{X}_\Omega \), is found by summing the stationary points from one whole period of the fundamental laser field, \( 0 < t < T \), i.e. two adjacent half-periods. There are thus two contributions from each branch,

\[
\vec{X}_\Omega(t + T/2, t_0 + T/2) = -\vec{X}_\Omega(t, t_0) \exp[-i\Omega T/2],
\]

separated by a half-period, \( T/2 \). The overall minus sign comes from the change of sign of \( E(t) \), while the phase factor originates from the Fourier component in the Legendre transformed action. The total dipole response becomes

\[
\vec{X}_\Omega \approx \sum_n \vec{X}_\Omega^{(n)} \left[ 1 - \exp[-i\Omega T/2] \right] = 2 \sum_n \vec{X}_\Omega^{(n)} \left\{ \begin{array}{cl} 1, & \Omega/\omega \text{ is odd} \\ 0, & \Omega/\omega \text{ is even} \end{array} \right.,
\]

where the two contributions add constructively for odd orders and destructively cancel for even orders.

### 2.2. Two-colour case

We now consider a two-colour laser field composed of a fundamental laser field and a weak second harmonic with the same polarization, \( \vec{A}(t) = A_1 \sin(\omega t) + \lambda A_2 \sin(2\omega t + \phi) \), where \( \lambda \) is a perturbation parameter. It is possible to solve the two-colour high-order harmonic emission using the stationary phase equations (equations (2)–(4)) directly, but this requires evaluation of the station at all values of \( \phi \). We will follow a different route where the second harmonic is treated as a perturbation and only the stationary points of one-colour HHG need to be calculated. The two-colour action is expanded in \( \lambda \) as

\[
S \approx \int_{t_0}^{t_f} \left( \frac{[\vec{p} - e\vec{A}_1(t')]^2}{2m} + I_p \right) dt' - \lambda \int_{t_0}^{t_f} \left[ \frac{[\vec{p} - e\vec{A}_1(t')]^2}{2m} + I_p \right] dt' + \frac{\lambda^2}{2} \int_{t_0}^{t_f} \left[ \frac{[\vec{p} - e\vec{A}_1(t')]^2}{2m} + I_p \right] dt' + \frac{\lambda^3}{6} \int_{t_0}^{t_f} \left[ \frac{[\vec{p} - e\vec{A}_1(t')]^2}{2m} + I_p \right] dt'
\]
The first term in equation (10) corresponds to the action in the one-colour case, $S_1$, and the second term is the correction term due to the interaction with the second harmonic field, $\sigma = \sigma(\phi)$. The correction term can be calculated as

$$\sigma = \frac{e}{m} \left[ \frac{p A_2}{2\alpha} \cos(2\omega t' + \phi) + e A_1 A_2 \left( \frac{\sin(3\omega t' + \phi)}{6\omega} - \frac{\sin(\omega t' + \phi)}{2\omega} \right) \right]_0^T,$$

(11)

where $\sigma$ depends on the ionization potential through the stationary points. Note that the derivation of equation (11) using the quantum mechanical stationary points differs from those presented previously [16, 18], because it includes the effect of the ionization potential within the SFA.

The two-colour high-order harmonic dipole can be calculated by using equation (6) and by adding an additional slow factor due to the second harmonic. In analogy with equation (9), the dipole from one period of the fundamental field contains one discrete contribution for each branch and half-period:

$$\tilde{X}_\Omega \approx \sum_n \tilde{x}_\Omega^{(n)} \left[ \exp[i \sigma^{(n)}] - \exp \left[ -i \sigma^{(n)} - i \frac{\Omega T}{\hbar} \right] \right]$$

$$= 2 \sum_n \tilde{x}_\Omega^{(n)} \times \begin{cases} \cos(\sigma^{(n)}/h), & \Omega/\omega \text{ is odd} \\ i \sin(\sigma^{(n)}/h), & \Omega/\omega \text{ is even} \end{cases}$$

(12)

where $\tilde{x}_\Omega^{(n)}$ is the half-period contribution in the one-colour field, and where the property

$$\sigma^{(n)}(t + T/2, t_0 + T/2) = -\sigma^{(n)}(t, t_0)$$

relating the two-colour phase between adjacent half-periods for a given branch $n$ is used. The intensity of the even order harmonic emission from a single atom can be approximated by

$$I_\Omega \propto |\tilde{X}_\Omega|^2 \approx \left| 2 \sum_n \tilde{x}_\Omega^{(n)} \sigma^{(n)}/h \right|^2,$$

(14)

which is valid for $|\sigma^{(n)}| \ll \pi$. We define the intensity from a specific branch, $n$, as

$$I_\Omega^{(n)}(\phi) \propto |\sigma^{(n)}(\phi)|^2.$$

(15)

This (artificial) separation of the branches can be realized in a macroscopic medium either by phase matching in a long gas cell [13] or by spatial separation in the far field [11, 16].

3. Subcycle delay dependence of even harmonics

One fascinating aspect of two-colour HHG is that it depends on the subcycle delay (or relative phase $\phi$) between the laser fields. Dudovich et al [16] have proposed to use the $\phi$-dependence of even-order harmonics in order to estimate the emission times of the attosecond pulses in situ. We will refer to the relative phases that maximize the intensity of the even harmonics as the in situ phases: $\phi_0^{(1)}(\Omega)$ and $\phi_0^{(2)}(\Omega)$, for the short and long branch, respectively. The in situ phases are plotted in figures 3(a) and (d) as a function of harmonic order for argon at two different IR intensities. The corresponding emission times of the high-order harmonics, $\text{Re}(I^{(n)}(\Omega)) = I_k^{(n)}(\Omega)$, are shown in figures 3(b) and (e).

The intensity in (a), (b) is realistic for typical laser pulses of 30 fs in argon, while the intensity in (d), (e) is greater than the saturation intensity and, therefore, not experimentally feasible for multicyle laser pulses. The aim of our analysis is to distinguish between what can be measured experimentally, $\phi_0^{(n)}(\Omega)$, and the desired emission times, $I_k^{(n)}(\Omega)$. The in situ phases from the classical model ($I_p = 0$) are plotted in figures 3(c) and (f) for comparison.

3.1. Behaviour close to cutoff

The similarity between the in situ phases and the emission times is striking, especially for the high intensity (figures 3(d), (e), (f)) where the short and long branch merge in the cutoff at harmonic 63. At lower intensity (figure 3(a)) the in situ phases do not merge in the cutoff. Intuitively, one might think that the short and long branch should merge in the cutoff, but this is not necessary in the quantum mechanical case. The stationary points of the short and long branch do not merge on the imaginary axis in the cutoff (figures 2(b), (d), (f)). The different behaviour of the in situ phases for the short and long branch in the cutoff is, hence, an amplitude effect rather than a pure phase effect. This is verified by inserting only the real part of the stationary points into equation (15) which does indeed yield coincidental cutoff behaviour of both branches, as expected for pure phase effects. Furthermore, it is the long branch that remains physical beyond the cutoff, while the strange behaviour of the short branch arises from...
a set of stationary points that become unphysical beyond the cutoff [27]. In the classical model (figures 3(c), (f)), the in situ phases always merge in the cutoff since there are no amplitude effects (damping) in the plateau (figures 2(b), (d), (f)). It is also worth noting that the in situ phases of the two branches intersect at lower harmonic orders than the cutoff. This ‘intra-plateau crossing’ is marked with a cross (×) in figures 3(a), (d) and it should not be confused as the position of the cutoff. At high intensities it is easy to distinguish between the intra-plateau crossing and the cutoff, while at low intensities they may be separated by a few harmonic orders only. In figure 1 we present an experimental result where \( \phi_0 \) is linear from harmonic 22 to harmonic 28. At higher harmonic orders, a dramatic bend is observed [13, 18]. Using our quantum mechanical model we identify the lower orders as part of the short branch, \( n = 1 \) (full curve), while the higher orders are identified as the long branch, \( n = 2 \) (dashed curve). We stress that the dominance of the long branch close to the cutoff is a new result which already appears at the single-atom level when the short branch becomes unphysical. This new finding illustrates the usefulness of quantitative probing of HHG using a perturbative field. This effect would be very difficult to observe with the RABITT method because the corresponding emission times always merge in the cutoff, see figures 3(b), (e). The classical model fails to reproduce the bend (figure 3(c)) because the corresponding intra-plateau crossing occurs at much lower harmonic orders (not shown). Having discussed the details of the behaviour close to the cutoff, we now turn our attention to the central part of the harmonic plateau.

### 3.2. Ratio of in situ phases and emission times

In the following, we consider the first spectral derivative of the in situ phase, \( \partial \phi_0^{(n)} / \partial \Omega \), which we wish to compare with the group delay dispersion (GDD), \( \partial t_R^{(n)} / \partial \Omega \). Both quantities are evaluated in the central 50% of the harmonic plateau, i.e. in a region set by \( 1 \pm 0.5 \) for the short branch and \( 1 \pm 0.8 \) for the long branch, where both \( \phi_0^{(n)}(\Omega) \) and \( t_R^{(n)} \) are linear to a very good approximation. We avoid fast and nonlinear variations both close to the ionization potential (\( h \Omega = I_p \)) and close to the cutoff (\( h \Omega = 1.3 I_p + 3.2 U_p \)) using this central region. We define the ratio between the two quantities as

\[
\gamma^{(n)} = -\omega \frac{\partial t_R^{(n)}}{\partial \Omega} / \frac{\partial \phi_0^{(n)}}{\partial \Omega} = -\omega \frac{\partial t_R^{(n)}}{\partial \phi_0^{(n)}}. \tag{16}
\]

The GDD can in principle be obtained from the \( \phi \)-dependence of the even-order harmonics as \( \partial t_R^{(n)} / \partial \omega = -\gamma^{(n)} \partial \phi_0^{(n)} / \partial \omega / \omega \). This relation would be very useful if \( \gamma^{(n)} \) was a constant (or at least a constant for each branch \( n \)). Unfortunately we show that \( \gamma^{(n)} \) depends on the ionization potential of the atom, as well as the laser intensity and wavelength.

#### 3.2.1. Role of ionization potential

In this subsection we study how \( \gamma^{(n)} \) varies with laser intensity for a given laser wavelength (800 nm) and different atomic species (helium, argon, sodium and the classical case). In our model of \( \gamma^{(n)} \), atom-specific properties enter only through the value of the ionization potential, \( I_p \). In figure 4, we plot \( \gamma^{(n)} \) for the short branch (a) and the long branch (b). In the classical case \( (I_p = 0) \) we find constant ratios: \( \gamma^{(1)} \approx 1.1 \) for the short branch and \( \gamma^{(2)} \approx 0.84 \) for the long branch, while in all realistic cases, where \( I_p > 0 \), there is a significant change in \( \gamma^{(n)} \) as a function of the fundamental intensity. In the case of argon there is an especially strong variation of the ratio: from 1.6 to 0.5 for the short branch over the given intensity range. This strong intensity dependence is problematic for the experimental determination of two-colour HHG since the exact effective intensity is often unknown in experiments. The variation of the ratios is smaller for the long branch, because the stationary points are more similar to the corresponding classical case, as seen in figure 2. The variation of the ratios is smaller for sodium than for helium because of the smaller ionization potential. Sodium is chosen as an example to show that there is a significant difference between the classical and quantum mechanical cases even for a relatively small ionization potential. Using the two-colour HHG approach for characterization of attosecond pulses would require an accurate determination of the laser intensity, as well as a quantum mechanical calculation of \( \gamma^{(n)} \).

Next we comment on our previous work [18] where we compared the in situ method and the RABITT method for GDD of attosecond pulses. An explanation for the good agreement we found between the two methods may come from the crossing of \( \gamma^{(1)} \) for argon occurring at \( 1.8 \times 10^{14} \) W cm\(^{-2} \) with the classical limit, see figure 4(a). It would be interesting to see more experimental results, carried out on different atoms.

#### 3.2.2. Role of laser wavelength

Finally, we study the dependence of the ratios, \( \gamma^{(n)} \), with laser wavelength. The ratios are calculated for 800 nm, 1.3 \( \mu \)m and 2 \( \mu \)m, corresponding to a titanium–sapphire laser system and two mid-IR laser sources. We use the ionization potential of argon.
Similar to figure 4, all ratios look different when they are plotted as a function of laser intensity, see figure 5(a) for the short branch and (b) for the long branch. The ratios of the short branch show larger variations than those of the long branch. Furthermore, it is seen that the longer wavelengths lead to less variation of the ratios over intensity. The recent experiments of Doumy et al [17] were carried out at these wavelengths. Applying our method improves the scaling law for the GDD from $\lambda^{-0.77}$ to $\lambda^{-1.05}$ which is closer to the expected $\lambda^{-1}$ scaling of the harmonic chirp times intensity. We stress that in order to use the calculated ratios to determine the GDD, the experimental intensity must first be measured as accurately as possible independent of the two-colour HHG scheme.

4. Discussion and conclusions

In our quantum mechanical derivation of two-colour HHG we find that both amplitude and phase effects are important. The ratio of the ponderomotive energy and the ionization potential, $U_p/I_p$, serves as a measure on how ‘classical’ or ‘quantum’ the electron trajectories are. Choosing this ratio as our $x$-axis for the data in figures (4) and (5), we find that all individual ratios $\gamma^{(n)}$ follow a universal curve, as shown in figure 6. Photoelectron emission ranging from the photon picture to the tunnelling picture is described in the theory of Keldysh [28] where the ratio $\sqrt{I_p/2U_p} \ll 1$ (corresponding to $U_p/I_p \gg 1$) implies efficient tunnel ionization. In this limit we find that $\gamma^{(n)}$ slowly converges towards the classical limit, $I_p = 0$. One should look at figure 6 with some caution since the SFA is derived for the long wavelength limit requiring $I_p \gg \hbar \omega$ and $U_p/I_p > 1/2$. It is clear that the behaviour of the HHG process changes dramatically around $U_p/I_p \approx 1$, i.e. when the kinetic energy of the electron is close to the potential energy of the atom.

In conclusion, we have studied the HHG process perturbed by a weak second harmonic field within the SFA. We find that the dependence of the even harmonics on the subcycle delay between the two fields, cannot be understood using classical theory. Our calculations show good agreement with experimental results [13, 18] showing the change of behaviour at high energy, explained as a change of dominant quasiclassical branch of trajectories. We stress that there is an intra-plateau crossing between the short and the long branch which does not coincide with the true cutoff. Furthermore, we calculate the ratio between the GDD of the attosecond pulses and the phase variation of the even-order harmonics, $\gamma^{(n)}$, as a function of intensity, wavelength and ionization potential. The analysis method called in situ probing of the birth of attosecond pulses [16] must be improved by considering the influence of the atomic properties and laser parameters, before it can be applied for quantitative experimental studies. Using the classical analysis will only lead to a qualitative prediction for the GDD of the attosecond pulses with the correct sign. It will be interesting to compare the results of our quantum mechanical approach to probing the birth of attosecond pulses using a two-colour field with more refined calculations.

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