

Supplemental Material: Analytical derivation for the phase of the below-threshold harmonics in a two-level model system

Samuel Schöpa¹, Lina Bielke¹, Falk-Erik Wiechmann^{1,2}, Franziska Fennel^{1,2}, and Dieter Bauer^{1,2}

¹*Institute of Physics, University of Rostock, 18051 Rostock, Germany.*

²*Department Life, Light & Matter, University of Rostock, 18051 Rostock, Germany.*

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The succeeding derivation follows the manuscript of L. Plaja and L. Roso [1] very closely. Our derivation slightly differs in the treatment of the electric field and the phase after (13).

We start with the Hamiltonian

$$\hat{H}_0 = \begin{pmatrix} 0 & 0 \\ 0 & \omega_{10} \end{pmatrix}, \quad (1)$$

that is the same as in our letter, just with shifted energies. The interaction part is

$$\hat{H}_1(t) = \begin{pmatrix} 0 & E(t)d \\ E(t)d & 0 \end{pmatrix}, \quad (2)$$

with the electric field $E(t)$ and the transition dipole matrix element $d = \langle \Psi_0(0) | x | \Psi_1(0) \rangle$. We express the wavefunction

$$|\Psi(t)\rangle = C_0(t) |\Psi_0(0)\rangle + C_1(t) |\Psi_1(0)\rangle \quad (3)$$

using time-dependent populations $C_0(t)$ and $C_1(t)$. The time-dependent Schrödinger equation for the populations follows with

$$\frac{d}{dt}C_0(t) = -iE(t)dC_1(t) \quad (4)$$

$$\frac{d}{dt}C_1(t) = -i\omega_{10}C_1(t) - iE(t)dC_0(t). \quad (5)$$

From the populations we can calculate the dipole

$$\begin{aligned} D(t) &= \langle \Psi(t) | x | \Psi(t) \rangle \\ &= d(C_0^*(t)C_1(t) + C_1^*(t)C_0(t)). \end{aligned} \quad (6)$$

For the dipole velocity follows

$$\begin{aligned} \frac{d}{dt}D(t) &= d[\dot{C}_0^*(t)C_1(t) + C_0^*(t)\dot{C}_1(t) \\ &\quad + \dot{C}_1^*(t)C_0(t) + C_1^*(t)\dot{C}_0(t)], \end{aligned} \quad (7)$$

and by plugging in (4) and (5)

$$\begin{aligned} \frac{d}{dt}D(t) &= d[iE(t)dC_1^*(t)C_1(t) \\ &\quad + C_0^*(t)(-i\omega_{10}C_1(t) - iE(t)dC_0(t)) \\ &\quad + (i\omega_{10}C_1^*(t) + iE(t)dC_0^*(t))C_0(t) \\ &\quad - C_1^*(t)iE(t)dC_1(t)] \\ &= -i\omega_{10}d[C_0^*(t)C_1(t) - C_1^*(t)C_0(t)]. \end{aligned} \quad (8)$$

For the dipole acceleration follows

$$\begin{aligned}
\frac{d^2}{dt^2}D(t) &= -i\omega_{10}d[\dot{C}_0^*(t)C_1(t) + C_0^*(t)\dot{C}_1(t) \\
&\quad - \dot{C}_1^*(t)C_0(t) - C_1^*(t)\dot{C}_0(t)] \\
&= -i\omega_{10}d[iE(t)dC_1^*(t)C_1(t) \\
&\quad + C_0^*(t)(-i\omega_{10}C_1(t) - iE(t)dC_0(t)) \\
&\quad - (i\omega_{10}C_1^*(t) + iE(t)dC_0^*(t))C_0(t) \\
&\quad + C_1^*(t)iE(t)dC_1(t)] \\
&= -\omega_{10}^2 \overbrace{d[C_0^*(t)C_1(t) + C_1^*(t)C_0(t)]}^{D(t)} \\
&\quad + 2\omega_{10}E(t)d^2[C_1^*(t)C_1(t) - C_0^*(t)C_0(t)],
\end{aligned} \tag{9}$$

and with the introduction of the population inversion $w(t) = |C_1(t)|^2 - |C_0(t)|^2$

$$\frac{d^2}{dt^2}D(t) + \omega_{10}^2 D(t) = 2\omega_{10}E(t)d^2w(t). \tag{10}$$

Using (4) and (5) we can also derive an expression for the time-derivative of the population inversion by

$$\begin{aligned}
\frac{d}{dt}w(t) &= \frac{d}{dt}[C_1^*(t)C_1(t) - C_0^*(t)C_0(t)] \\
&= \dot{C}_1^*(t)C_1(t) + C_1^*(t)\dot{C}_1(t) \\
&\quad - \dot{C}_0^*(t)C_0(t) - C_0^*(t)\dot{C}_0(t) \\
&= [i\omega_{10}C_1(t)^* + iE(t)dC_0^*(t)]C_1(t) \\
&\quad + C_1^*(t)[-i\omega_{10}C_1(t) - iE(t)dC_0(t)] \\
&\quad - iE(t)dC_1^*(t)C_0(t) - C_0^*(t)[-iE(t)dC_1(t)] \\
&= 2iE(t)d[C_0^*(t)C_1(t) - C_1^*(t)C_0(t)].
\end{aligned} \tag{11}$$

With (8) follows

$$\frac{d}{dt}w(t) = -2\frac{E(t)}{\omega_{10}}\frac{d}{dt}D(t). \tag{12}$$

We consider a periodic electric field

$$E(t) = E_0 \cos(\omega_L t - \varphi). \tag{13}$$

If our system is only populating a single Floquet state of the interacting system, all dynamics are adiabatic, and the dipole and the population inversion will be periodic with the laser period [2]. Therefore, they can be expressed in terms of multiples of the laser frequency by the Fourier expansion

$$\begin{aligned}
w(t) &= \sum_n w_n e^{in\omega_L t} \\
D(t) &= \sum_n d_n e^{in\omega_L t},
\end{aligned} \tag{14}$$

with the time-independent coefficients w_n and d_n , and the index n running over all integer numbers. Plugging (14)

into (12) yields

$$\begin{aligned}
\frac{d}{dt} \sum_n w_n e^{in\omega_L t} &= \sum_n -2 \frac{E_0}{\omega_{10}} \cos(\omega_L t - \varphi) \frac{d}{dt} d_n e^{in\omega_L t} \\
\sum_n n w_n e^{in\omega_L t} &= \sum_n -\frac{E_0}{\omega_{10}} n d_n e^{in\omega_L t} (e^{i\varphi} e^{-i\omega_L t} + e^{-i\varphi} e^{i\omega_L t}) \\
&= \sum_n -\frac{E_0}{\omega_{10}} n d_n (e^{i\varphi} e^{i(n-1)\omega_L t} + e^{-i\varphi} e^{i(n+1)\omega_L t}) \\
&= \sum_n -\frac{E_0}{\omega_{10}} [(n+1)d_{n+1}e^{i\varphi} + (n-1)d_{n-1}e^{-i\varphi}] e^{in\omega_L t},
\end{aligned} \tag{15}$$

and by comparing the coefficients we obtain

$$n w_n = -\frac{E_0}{\omega_{10}} [(n+1)d_{n+1}e^{i\varphi} + (n-1)d_{n-1}e^{-i\varphi}]. \tag{16}$$

Analogously, by inserting the Fourier expansions (14) into (10),

$$\begin{aligned}
\sum_n [(in\omega_L)^2 d_n e^{in\omega_L t} + \omega_{10}^2 d_n e^{in\omega_L t}] &= \sum_n 2\omega_{10} E_0 \cos(\omega_L t - \varphi) d^2 w_n e^{in\omega_L t} \\
&= \sum_n \omega_{10} E_0 d^2 w_n e^{in\omega_L t} (e^{i\varphi} e^{-i\omega_L t} + e^{-i\varphi} e^{i\omega_L t}) \\
&= \sum_n \omega_{10} E_0 d^2 (w_{n+1} e^{i\varphi} + w_{n-1} e^{-i\varphi}) e^{in\omega_L t},
\end{aligned} \tag{17}$$

and again comparing the coefficients,

$$(\omega_{10}^2 - n^2 w_L^2) d_n = \omega_{10} E_0 d^2 (w_{n+1} e^{i\varphi} + w_{n-1} e^{-i\varphi}) \tag{18}$$

follows. We can utilize (16) to write

$$w_{n+1} = -\frac{1}{n+1} \frac{E_0}{\omega_{10}} [(n+2)d_{n+2}e^{i\varphi} + n d_n e^{-i\varphi}], \quad \text{and} \tag{19}$$

$$w_{n-1} = -\frac{1}{n-1} \frac{E_0}{\omega_{10}} [n d_n e^{i\varphi} + (n-2)d_{n-2}e^{-i\varphi}], \tag{20}$$

and insert these expressions into (18), to obtain a continued-fractions expression for the dipole coefficients

$$\begin{aligned}
(\omega_{10}^2 - n^2 w_L^2) d_n &= -\omega_{10} E_0 d^2 \left(\frac{1}{n+1} \frac{E_0}{\omega_{10}} [(n+2)d_{n+2}e^{i\varphi} + n d_n e^{-i\varphi}] e^{i\varphi} \right. \\
&\quad \left. + \frac{1}{n-1} \frac{E_0}{\omega_{10}} [n d_n e^{i\varphi} + (n-2)d_{n-2}e^{-i\varphi}] e^{-i\varphi} \right) \\
\frac{\omega_{10}^2 - n^2 w_L^2}{E_0^2 d^2} d_n &= - \left[\frac{n+2}{n+1} d_{n+2} e^{2i\varphi} + \left(\frac{n}{n+1} + \frac{n}{n-1} \right) d_n + \frac{n-2}{n-1} d_{n-2} e^{-2i\varphi} \right] \\
\left(\frac{2n^2}{n^2-1} + \frac{\omega_{10}^2 - n^2 w_L^2}{E_0^2 d^2} \right) d_n &= - \left(\frac{n+2}{n+1} d_{n+2} e^{2i\varphi} + \frac{n-2}{n-1} d_{n-2} e^{-2i\varphi} \right) \\
d_n &= - \left(\frac{n+2}{n+1} d_{n+2} e^{2i\varphi} + \frac{n-2}{n-1} d_{n-2} e^{-2i\varphi} \right) \left(\frac{2n^2}{n^2-1} + \frac{\omega_{10}^2 - n^2 w_L^2}{E_0^2 d^2} \right)^{-1}.
\end{aligned} \tag{21}$$

For a parameter regime, where each successive harmonic order is substantially weaker than the previous one, we can neglect the d_{n+2} -term in (21), resulting in the expression

$$d_n = -e^{-2i\varphi} \frac{n-2}{n-1} d_{n-2} \left(\frac{2n^2}{n^2-1} + \frac{\omega_{10}^2 - n^2 w_L^2}{E_0^2 d^2} \right)^{-1}. \tag{22}$$

Due to the nature of the Fourier expansion, the complex coefficient $d_n = \tilde{D}(n\omega_L)$ is equivalent to the Fourier transform of the dipole at the frequency $n\omega_L$. Therefore, the phase of the n -th harmonic can be extracted by $\varphi_n = \arg(d_n)$.

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- [1] L. Plaja and L. Roso, High-order harmonic generation in a two-level atom, J. Mod. Opt. **40**, 793 (1993).
 - [2] V. Kapoor, M. Ruggenthaler, and D. Bauer, Periodicity of the time-dependent kohn-sham equation and the floquet theorem, Phys. Rev. A **87**, 042521 (2013).