An Introduction to High-Harmonic Generation: Towards High-Harmonic Spectroscopy

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1 What is high-harmonic generation

High-harmonic generation (HHG) in the context of this project refers to the generation of coherent extreme ultraviolet (XUV) or vacuum ultraviolet (VUV) radiation from laser light in the visible or infrared (IR). The frequency of the generated radiation is $q$ times larger than the frequency of the driving laser, whereas $q$ can be up to a hundred or more. Experimentally there are different methods of creating such harmonics. Common is HHG in noble gases, where the light is interacting directly with atoms and a more complicated scheme of HHG from surfaces, where the light is interacting with dielectric or metallic surfaces. We will focus here on the first approach.

Figure 1: Phenomenological picture of HHG. An atom within a sufficiently strong electric light field emits radiation of higher frequencies. Experimentally the light field driving the process is usually a short light pulse.

For HHG from noble gases typical laser intensities of the order of $10^{14} \text{W/cm}^2$ are required. This is explained by the onset of tunnel ionization. How this is related and what it is will become clear later. Compared to an average nuclear power plant with a continuous output power of giga ($10^9$) watts, these intensities are enormous. Experimentally they are supplied by short pulse lasers with durations in the order of femtoseconds ($10^{-15}$s, from the Swedish word for fifteen: femton), yielding sufficiently high peak intensities while having manageable average output powers. Figure 1 depicts the process as explained above.

Figure 2: HHG in a free focusing geometry, the gas is supplied via a pulsed nozzle injecting the sample into high vacuum. This has various advantages, mainly the possibility of cooling the individual atoms or molecules in a supersonic gas expansion and keeping the gas load in the vacuum system low.

An important point is that the absorption length of the harmonics at atmospheric pressure is in the order of millimeters. Therefore the whole setup, from generation to interaction to
detection must be placed in vacuum. This complicates the task of making laser pulses and gas interact, as the handling of a gas source in vacuum can be delicate. In general, there are different possibilities for the laser-gas interaction. For the scope of this project it is sufficient to consider the simplest case of a laser freely focused into a pulsed gas jet as depicted in figure 2.

2 The three step model

There is a very simple and straightforward model to explain HHG. It is called the semi classical three step model and was initially suggested by Paul Corkum [1]. Corkum stated that the process of creating high-order harmonics can be divided in three distinct steps:

1. The initially bound electron is ionized and appears with zero velocity in the light field.
2. The electron is accelerated by the light field.
3. Re-colliding with the atom it creates an oscillating electric dipole that emits radiation.

The first step is here the most complicated one to explain in an understandable manner. It would be necessary to consider the electron in a quantum mechanical context and distinguish between several regimes depending on how strong the field is compared to the atomic potential. It is sufficient to know at this point that the electron can appear at several different times with respect to the external light field and that the probability of this ionization process scales strongly nonlinearly with the intensity. Once it is subjected to the external field, the motion of the electron can be described classically by the forces acting on it due to the electromagnetic field of the laser. Under certain circumstances the electron will then recombine with the atom, emitting its kinetic energy plus the binding energy in the form of electromagnetic radiation. An immediate deduction would be that the maximal photon energy can not exceed the electron energy at impact plus the ionization potential, due to the conservation of energy. Based on the three step model, very simple calculations reproducing characteristic features of HHG are possible. Figure 3 depicts the complete process.

![Figure 3: a) The electron is ejected into the continuum. b) The electron propagates in the continuum on a trajectory defined by the electromagnetic field. c) The wave packet comes back to its initial position at a later time and recombines with the atom or molecule.](image)

3 Simulations based on the three step model

Figure 4 depicts simulations of the concept described above, assuming a cosine shaped linearly polarized electric field of the laser. The different black lines represent one dimensional trajectories of electrons that have appeared at different times in the electric field. They were given
the properties \( y(t_0) = 0 \), meaning that the atoms appear directly at the position of the atom and \( v(t_0) = 0 \), meaning that they have no initial velocity. Some of those electrons re-collide with the atom, in other words they come back to \( y = 0 \), while others wiggle away from the atom.

Figure 4: Different one-dimensional trajectories (black lines) for electrons appearing at different times in the external field (red line). The time is given as the argument of the cosine function.

Above we deduced that the maximal photon energy cannot exceed the kinetic energy at impact plus the ionization potential. Plotting the kinetic energy of the simulated electrons at the re-impact on the atom (see figure 5), we find that this model predicts that

\[
E_{\text{photon-max}} \leq 3.17U_p + I_p, \tag{1}
\]

where \( U_p = \frac{e^2 E_0^2}{4m_0\omega^2} \) is the ponderomotive potential, which is in this case just a convenient way to collect factors like the electron charge, the electron mass, the light frequency etc. and \( I_p \) being the ionization potential.\(^1\) This result agrees surprisingly well, with what has been measured in experiments. Even for expressions obtained from more sophisticated models, the deviation from the limit above is small.

Figure 5: Kinetic energy at the re-impact on the atom for different electron emission times.

The emission time is given in terms of multiples of \( \pi \), representing the phase of the driving electric field \( E \propto \cos(t) \) - see figure 4.

\(^{1}\)However it can be shown in a straightforward manner that \( U_p \) represents the average kinetic energy of a free electron in a monochromatic light field.
4 Classical predictions on the HHG spectrum I

Considering a realistic laser pulse, it is clear that the process of electrons recombining with the atom or molecule repeats itself every half cycle for the duration of the pulse (sketched in figure 6). This property can be used to predict some important spectral properties of the generated radiation.

![Phenomenological picture of a train of HHG light bursts.](Image)

Figure 6: Phenomenological picture of a train of HHG light bursts. It is important, that the bursts are separated by one laser half cycle. This is due to the ionization probability scaling strongly nonlinearly, therefore being highest at the peaks of the electric field.

Any periodic function \( g(t) \) consisting of equally spaced identical functions \( f(t) \) can be expressed mathematically as a convolution of the corresponding function \( f(t) \) with a comb \( f_{\text{comb}}(t) \) of Dirac functions. The definition of this Dirac comb is

\[
f_{\text{comb}}(t) = \sum_{n=-\infty}^{+\infty} \delta(t - n\Delta t).
\]

In this expression \( \Delta t \) is the temporal spacing between the individual events. This describes the behavior in figure 6 already fairly well, \( g(t) \) is some sort of function describing the system and \( f(t) \) is the function corresponding to the individual light bursts. However, the Dirac comb as defined above is infinitely long. This also implies that \( g(t) \) would be infinitely long, therefore an additional envelope \( f_{\text{envelope}} \) is introduced to confine \( g(t) \) temporally. Thus we have

\[
g(t) = [f(t) \otimes f_{\text{comb}}(t)] f_{\text{envelope}}(t).
\]

Therefore the Fourier transform is given as

\[
g(\omega) = \mathcal{F}(g(t)) = [f(\omega) f_{\text{comb}}(\omega)] \otimes f_{\text{envelope}}(\omega).
\]

It can be shown that the Fourier transform of the Dirac comb for positive frequencies is

\[
f_{\text{comb}}(\omega) = \frac{2\pi}{\Delta t} \sum_{n=0}^{+\infty} \delta(\omega - n\frac{2\pi}{\Delta t}).
\]

If we take \( \omega_0 = \frac{2\pi}{\Delta t} \) this allows us to write

\[
g(\omega) = \left[ \omega_0 \sum_{n=0}^{+\infty} f(n\omega_0) \delta(\omega - n\omega_0) \right] \otimes f_{\text{envelope}}(\omega).
\]

or

\[
g(\omega) = \omega_0 \sum_{n=0}^{+\infty} f(n\omega_0) f_{\text{envelope}}(\omega - n\omega_0).
\]

Thus, we expect to see in the spectrum is a set of peaks at multiple frequencies of the fundamental, where each of the harmonic peaks has a width corresponding to the envelope function.
Typically it is a good approximation to take the envelope function of the Gaussian laser pulse as approximation for the envelope function above. The total extent of the spectrum is given by $f(\omega)$.

## 5 Classical predictions on the HHG spectrum II

Even though $f(\omega)$ and $f(t)$ can not be discussed thoroughly within the scope of this project, we can still deduce some of its properties with simple symmetry considerations. Here it has to remain a postulate, but $f(t)$ is related to the polarization $P(t)$. Polarization refers in this context to the displacement of the electron cloud from the equilibrium position by the electromagnetic field. In nonlinear optics the polarization is often written in a power expansion of the electric field

$$P(t) = \epsilon_0 \left[ \chi^{(1)} \otimes E(t) + \chi^{(2)} \otimes E(t) \otimes E(t) + (\ldots) \right].$$

(8)

The $\chi^{(n)}$ occurring in this expression are the $n$-th order dielectric susceptibilities. In case $E(t)$ is monochromatic equation 8 can be rewritten as

$$P(t) = \epsilon_0 \left[ \chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + (\ldots) \right].$$

(9)

In a spherical symmetric system we can require $P(E) = -P(-E)$, meaning that the electron cloud displacement is independent of the direction of the electric field (up to a sign change). This implies automatically that for all $m \in \mathbb{N}$, $\chi^{2m}$ has to be zero. If we take an initial electrical field of the form

$$E(t) = E_0 \exp(i\omega t),$$

(10)

it follows directly that the absence of even order $\chi$ also implies an absence of even harmonics. Therefore in the overall spectrum that was predicted in the previous section no even harmonic peaks will be seen as long as a spherical symmetry is given.

## 6 Towards high harmonic spectroscopy

In the previous sections, it has been stated on a qualitative level how high-harmonic generation is explained within the three step model. Anyhow, the narration was chosen such that it is intuitive and simple. For instance, instead of saying the incident electromagnetic field distorts the Coulomb potential of the atom or molecule thereby allowing parts of the electron wave function to couple to the continuum through multiphoton-, tunnel- or above threshold ionization, it was said an electron appears in the continuum. This section should now sketch a model on a fully quantum mechanical basis. It can then be shown that in a special case this model reduces to the three step model.

The starting point here is something that has been used already, namely that the polarization can be identified as a source term in the calculation of electric field propagation. Therefore what needs to be found is a quantum mechanical description of the polarization of the single atom or molecule. It is convenient to start with the very simple model of a single electron in a neutral host. The polarization is then just given by

$$\vec{P}(t) = e\vec{x}(t).$$

(11)
Where $\vec{x}(t)$ is the displacement from the equilibrium position. Quantum mechanically $\vec{x}(t)$ is given by the expectation value of the $\hat{x}$ operator, or in Dirac notation

$$x(t) = \langle \Psi, t | \hat{x} | \Psi, t \rangle \quad (12)$$

This basically only leaves us with the task of calculating $|\Psi, t \rangle$, or more explicitly finding the solutions of the time-dependent Schrödinger equation for the given Hamiltonian of the system

$$i\hbar \frac{\partial}{\partial t} |\Psi, t \rangle = \hat{H} |\Psi, t \rangle . \quad (13)$$

The interaction Hamiltonian is here $\hat{H} = \hat{H}_0 + \hat{V}_{\text{light}}(x,t)$, where $\hat{H}_0$ denotes the Hamiltonian without the electromagnetic field. Solving the time-dependent Schrödinger equation (eq. 13) (TDSE) exactly is a very tedious undertaking as for a real atom with a multi electron Hamiltonian. It can be almost arbitrarily complicated. Therefore, as of today, exact solutions are only available in a very limited range of cases.

Within a set of approximations that have been dubbed as strong field approximation (SFA) [2], $P(t)$, for a linear polarized electric field with a slowly-varying envelope, is given by

$$\vec{P}(t) = ei \int_{-\infty}^{+\infty} \left[ \int_0^t \frac{eE_0}{\hbar} \cos(\omega t') dx \left( \Pi - e\vec{A}(t') \right) \times \right.$$}

$$\left. d_x^* \left( \Pi - e\vec{A}(t) \right) \exp \left( -iS(\Pi, t, t') \right) dt' + c.c. \right] d\Pi^3. \quad (14)$$

In the expression above, the term $d_x(a)$ and its complex conjugate represent only a short form of the dipole matrix element $\langle a|x|\Psi_0 \rangle$ that describes the coupling from the ground state to the continuum state $|a\rangle$. The different terms can now be interpreted according to reference [2]. The first term in the integral $\frac{eE_0}{\hbar} \cos(\omega t') dx \left( \Pi - e\vec{A}(t') \right)$ is the part of an electron wave packet that makes the transition to the continuum at time $t'$ (see figure 7) with canonical momentum $\Pi$. $\exp \left( -iS(\Pi, t, t') \right)$ can be identified as the free space propagator, governing the phase gain of that part of the wave packet during propagation. Finally $d_x^* \left( \Pi - e\vec{A}(t) \right)$ describes the recombination at time $t$. This resembles the semi classical three step model described earlier with the difference that we are considering and calculating all possible pathways for the electron wave packet. The possible interferences between these quantum paths distinguishes the two models, as for the semi classical case only whole electrons are propagated.

Within this formalism we expect informations about $|\Psi_0 \rangle$ to be encoded in the general radiation properties. This is why HHG is interesting from a spectroscopical point of view.[3]
Figure 7: Tunneling of the initially bound electron into the continuum. The electron confined by the atomic potential can tunnel out of the effective potential (red) that is created by superimposing the electric field potential (red, dashed). After it has appeared in the continuum the electron will move on the potential surface created by the electric field.
7 Experimental spectra

In this section a set of experimental HHG spectra are collected. These spectra are required for the tasks given in the next section.

Figure 8: HHG spectrum recorded from argon at $3 \times 10^{13} \text{ W cm}^{-2}$ [4].
Figure 9: HHG spectrum recorded from oriented OCS [5]. The dashed blue line corresponds to a random distribution of orientations of the molecules in space.

Figure 10: HHG spectrum recorded from Argon at $3.5 \times 10^{14}$ [6]. There are two distinct features in this spectrum: a) a pronounced dip between 50 and 60 eV and b) the lack of the harmonic peak structure for high photon energies.
8 Tasks

1. Show that the average kinetic energy of an electron in a monochromatic electromagnetic field is given as $U_P = \frac{e^2 E^2}{4m_0 c^2}$.

2. Write a small program that reproduces the results displayed in figures 4 and 5.

3. Show that the Fourier transform of a Dirac comb is again a Dirac comb.

4. Use the knowledge you have obtained to interpret the spectrum given in figure 8. Find the approximate wavelength of the driving laser field and give a lower boundary for the pulse duration from information in the figure. Calculate $U_P$ and interpret the cutoff. Hint: The ionization potential of argon is at 15.8 eV.

5. Figure 9 shows a HHG spectrum from oriented OCS. Oriented means in this context, that the molecules have a fixed orientation of their main axes relative to the electric field. You can clearly observe even harmonics which we have excluded above. Can you reason where the train of thought above fails and why? What can be said about the difference of the driving laser pulse length from figure 9 and 8.

6. Explain briefly what the term canonical means in this context and sketch how the canonical momentum for a free electron in an electromagnetic field can be obtained.

7. Some spectra show additional structures to the ones you have learned about. Take a look at figure 10. Consider equation 14 can you explain in which of the terms you expect effect a) to be contained in. Sketch how you would prove your hypothesis. Explain effect b) and sketch the electric field as a function of time of the driving laser pulse and indicate at which point in time the individual contributions are generated.

References


