Two-color photoionization in XUV-FEL and Laser fields

Richard Taïeb¹, Alfred Maquet¹ and Michael Meyer²
¹UPMC, Université Paris 06, CNRS, UMR 7614, Laboratoire de Chimie Physique-Matière et Rayonnement, 11, rue Pierre et Marie Curie, 75231 Paris, Cedex 05, France.
²LIXAM, UMR 8624, Université Paris Sud, Bâtiment 350, 91405 Orsay Cedex, France
E-mail: richard.taieb@upmc.fr

Abstract. We address several questions related to the nonlinear ionization processes observed when atoms are in the simultaneous presence of intense and coherent infrared (IR) and extreme ultraviolet (XUV) laser pulses. This topic is of much interest in the context of the current development of new XUV coherent sources, either from high-order harmonics or from free-electron laser (FEL) devices, like the one available at FLASH in Hamburg, that can be synchronized with IR lasers. In this paper, we discuss the advantages and limitations of the so-called soft-photon approximation, which, we will show here, provides most useful insights in the analysis of different processes involving these two fields.

1. Introduction
Multiphoton single-color ionization in intense optical or infrared laser fields has been the subject of multiple experimental and theoretical studies for more than two decades and is by now a very well understood process (e.g. [1]). The extension of these studies to multiphoton absorption in the extreme ultraviolet (XUV) regime becomes possible with the advent of new intense radiation sources like High order Harmonics Generation (HHG) of an IR field or Free electron LASer in Hamburg (FLASH). The latter, based on the process of Self-Amplified Spontaneous Emission (SASE), is a powerful source of extremely bright and ultrashort coherent laser light. Therefore, facilities that feature synchronized infrared (IR) and extreme ultraviolet laser pulses, make feasible the development of new spectroscopy tools designed to study the properties of the continuous spectrum of atomic and molecular species.

In this paper, we will argue that, within this context, the so-called soft-photon approximation provides a very convenient conceptual framework to address the challenges raised by the results obtained from such new set of experiments. We will present different schemes which will provide useful informations on the dynamics of these two-color processes.

2. Theory
The development of strong IR laser fields in the last 40 years provided experimental and theoretical physicists with a powerful tool in order to explore different multiphoton processes in atoms or small molecules. In fig. 1, we sumarize these processes going from multiphoton ionization first observed in the late 60s, to HHG through "Above Threshold Ionization". The advent of intense XUV sources arises the question of whether we can transpose some of these
Atomic processes in intense (IR) laser fields

Ground state

Moderate Intensities

Multiphoton Ionization

«Above Threshold Ionization» (ATI)

Higher Intensities

+ Harmonic Generation

Ionization limit

\( I_p \)

\( \omega_L \)

\( E_{\text{kin}} \approx n\omega_L - I_p - U_p \)

\( \omega_H = (2n+1)\omega_L \)

\( o_H = (2n+1) o_L \)

**Figure 1.** Processes observable for a IR laser as a function of the field intensity. \( I_p \) is the ionization potential, \( \omega_L \) the laser frequency and \( U_p \) the ponderomotive energy (see text)

processes to higher photon energies. In order to answer this, one can consider the motion of a "classical" electron within an electric field polarized along the \( x \) axis with frequency \( \omega_L \), i.e. \( \vec{F}(t) = F_0 \vec{\epsilon} \sin(\omega_L t + \phi) \). The Newton equation then writes as:

\[
m \frac{d^2 \vec{x}}{dt^2} = qF_0 \vec{\epsilon} \sin(\omega_L t + \phi)
\]

Integrating this equation, we obtain the expressions for the velocity and the position of the electron as:

\[
\vec{v}(t) = \frac{d\vec{x}}{dt} = -\frac{qF_0 \vec{\epsilon}}{m\omega_L} \left[ \cos(\omega_L t + \phi) - \cos(\phi) \right] + \vec{v}(0)
\]

\[
\vec{x}(t) = -\alpha_0 \left[ \sin(\omega_L t + \phi) - \sin(\phi) \right] + \left[ \vec{v}(0) + \alpha_0 \omega_L \cos(\phi) \right] t + \vec{x}(0).
\]

\[
\alpha_0 = \frac{qF_0 \vec{\epsilon}}{m\omega_L^2}
\]

Here, \( \vec{v}(0) \), \( \vec{x}(0) \) are the initial velocity and position, respectively and \( \alpha_0 \) the excursion length of the oscillatory motion. In the case of tunneling ionization these quantities are equal to 0. However, when we consider that the ionization is due to the absorption of an XUV photon \( \omega_X \), we have \( v^2(0)/2 = \omega_X - I_p \), where \( I_p \) is the ionization energy.

When averaging over time the kinetic energy of the oscillatory motion, one recognizes the ponderomotive energy \( U_p \) given by:

\[
\frac{1}{2m} \langle v^2(t) \rangle = U_p = \frac{q^2 F_0^2}{4m \omega_L^2}.
\]
Such similar treatment leads to the position of the "cut-off" in the harmonic generation [2]. One notes the \(\omega_L^2\) for both \(\alpha_0\) and \(U_p\). Therefore these two quantities, which become comparable either to atomic quantities (\(a_0 \approx 0.529\) Å, the Bohr radius, and \(I_P \approx 10\) eV, the ionization potential) for an intensity \(\approx 10^{14}\) W cm\(^{-2}\) of an IR (Ti:Sapphire) laser of frequency \(\omega_L \approx 1.55\) eV, become negligible for an XUV field of frequency \(\approx 92\) eV, like in FLASH. This leads to the conclusion that multiphoton processes will be much more difficult to observe in the X-UV than in IR regime.

To treat the case of the photoionization of He resulting from the absorption of one XUV photon in the presence of the IR field, one has to write the corresponding S-matrix transition amplitude, starting from the He ground state and ending in a dressed continuum state, [3]:

\[
S = -i \int_{-\infty}^{+\infty} d\tau \langle \chi_{\vec{k}}(\vec{r},\tau) | \frac{1}{c} \vec{A}_X, \vec{p} | \psi_{1s2}(\vec{r},\tau) \rangle.
\]  

where \(\vec{A}\) is the vector potential associated to the XUV electric field, given by \(\vec{F}_X(t) = -\frac{1}{c} \frac{\partial \vec{A}_X}{\partial t}(t)\).

\[
\chi_{\vec{k}}(\vec{r}, t) = (2\pi)^{-3/2} \exp[i(\vec{k} \cdot \vec{r} - \frac{k^2}{2} t - \alpha_0 \vec{k} \sin(\omega_L t + \phi))],
\]

is the so-called Volkov wavefunction representing the electron in the continuum dressed by the laser field, [4]. Integrating over time and using properties of the Bessel function and of their generating function [5], we obtain:

\[
S = \sum_{n=-\infty}^{+\infty} S^{(n)} \delta \left( \frac{k^2}{2} - (E_{1s2} + \omega_X + n\omega_L) \right),
\]

where the components \(S^{(n)}\) are associated with the net exchange of \(n\) laser photons in addition to the absorption of one XUV photon. The corresponding differential cross section is:

\[
\frac{d\sigma^{(n)}}{d\Omega} = J_n^2(\alpha_0 \cdot \vec{k}_n) \frac{d\tilde{\sigma}}{d\Omega},
\]

where \(k_n = \sqrt{2(E_{1s2} + \omega_X + n\omega_L)}\) is the shifted wave number of the ejected electron and \((d\tilde{\sigma}/d\Omega)\) the laser free cross section corresponding to the absorption of one \(\omega_X\) photon. The cross section \(\sigma^{(n)}\) is obtained by integrating over the electron solid angle \(\Omega \equiv (\eta, \phi)\). For a 1s He electron, and after integrating over \(\phi\) we obtain

\[
\sigma^{(n)} \propto a_n + b_n \cos^2 \theta,
\]

where the coefficients \(a_n\) and \(b_n\) are:

\[
\begin{cases}
  a_n = a_n = \int_0^\pi d\eta \sin \eta J_n^2(\alpha_0 k_n \cos \eta) \left\{ \frac{\sin^2 \eta}{2 - 3 \sin^2 \eta} \right\},
  b_n = b_n = \int_0^\pi d\eta \sin \eta J_n^2(\alpha_0 k_n \cos \eta) \left\{ \frac{\sin^2 \eta}{2 - 3 \sin^2 \eta} \right\},
\end{cases}
\]

and \(\theta\) denotes the angle between the polarization directions of the two fields.

In the case of parallel polarization between the two fields (\(\theta = 0\)), we have performed full resolution of the Time-Dependent Schrödinger Equation (TDSE) in order to prove the validity of the "soft-photon" approximation.
3. Results
Two typical two-colour, angle-integrated, photoionization spectra of He are shown in figure 1. In these simulations, we have considered the case of linearly polarized XUV and IR pulses, with parallel polarizations, photon energies $\omega_X = 92$ eV $\omega_L = 1.55$ eV, respectively, and same duration. The respective peak intensities of the fields are $I_X = 10^{12}$ Wcm$^{-2}$ and $I_L = 5 \times 10^{11}$ Wcm$^{-2}$ (figure 1(a)) and $I_L = 2 \times 10^{12}$ Wcm$^{-2}$ (figure 1(b)). These parameters are representative of those that are currently used in experiments performed at the Hamburg FLASH facility. In the absence of the laser, one observes a single photoelectron peak located at $k_0^2/2 \approx 68.0$ eV, see figure 1(a). We mention that two-photon ionization from the 92 eV XUV field is completely negligible at this intensity.

![Figure 2](image-url)

**Figure 2.** TDSE treatment versus soft-photon approximation for two-colour IR-XUV photoionization of He. Angle-integrated spectra for $\omega_X = 92$ eV $\omega_L = 1.55$ eV at different IR laser intensities, see text. Thin black line: TDSE calculation; red squares: soft-photon approximation. (a) $I_L = 5 \times 10^{11}$ Wcm$^{-2}$. The thick green line is the XUV single-photon absorption spectrum, without IR field, from the TDSE treatment. (b) $I_L = 2 \times 10^{12}$ Wcm$^{-2}$. The two fields have parallel polarizations. (Note the change of scale.)

In a typical two-colour IR-XUV experiment, the photoelectron energy spectrum consists
of a main line associated with the absorption of one XUV photon accompanied by sideband lines, located symmetrically on each side, see figure 1. The equally spaced sidebands, that are separated from each other by $\Delta E = \omega_L$, are associated with the additional exchange of laser photons with frequency $\omega_L$, through absorption and/or stimulated emission processes. One notes the excellent agreement between our two theoretical approach, giving confidence to the "soft-photon" approximation in providing qualitative and quantitative results.

We see that the number and the height of the sidebands strongly depends on the intensity of the IR field and therefore on both the spatial and temporal overlap between the two fields. We have used this fact to measure the temporal jitter of the FEL [7, 8].

In addition, we have recently performed experiments at FLASH [9], where we varied the angle between the polarizations. Briefly, the XUV FEL and the optical laser beams were introduced into the vacuum chamber in a collinear geometry and intersect an effusive gas jet within the acceptance volume of a magnetic bottle electron spectrometer. In order to ensure perfect time overlap between the pulses and to eliminate effects arising from the inherent time jitter of the FEL of about 500 fs [7], the experiments were performed using picosecond optical lasers synchronized to the 10-20 fs FEL pulses [8]. The polarization of both lasers was almost perfectly linear and the relative orientation between the polarization vectors could be changed by a rotatable half-wave plate in the optical path.

**Figure 3.** Variation of the upper sidebands yield for the two-photon ionization in the 1s shell of He at 13.7 nm as a function of the relative angle between the linear polarization vectors of the FEL and the optical laser light. Left panel: Low field regime ($I_L = 8 \times 10^{10}$ Wcm$^{-2}$). Right panel: Moderate field regime ($I_L = 6 \times 10^{11}$ Wcm$^{-2}$). The solid lines denote the fit to the experimental data (circles and diamonds for the first and second sideband, respectively). The dotted lines represent the results of the "soft-photon" approximation.

At low laser intensity, i.e. $z = \alpha_0 k_n \cos \eta << 1$, cf. Fig.(3) left panel, the Bessel function in Eq. (9) is proportional to $(\alpha_0 k_n \cos \eta)^n$, which gives $b_n/a_n = 2n$, leading to a contrast of $\sigma_{\max}^{(n)}/\sigma_{\min}^{(n)} = 2n + 1$ in the $\theta$ dependence of $\sigma^{(n)}$. This coincides with the result at low intensity where we obtain an experimental contrast of $3.25 \pm 0.25$ very close to the value of 3 ($n = 1$).

However, at higher intensity, cf. Fig.(3) right panel, when $z \approx 1$, one gets for the first
sideband \((n = 1)\):

\[
a_1 \approx \frac{z^2}{15} \left[ 1 - \frac{3z^2}{28} + \frac{25z^4}{4032} \right]
\]

\[
b_1 \approx \frac{2z^2}{15} \left[ 1 - \frac{6z^2}{28} + \frac{75z^4}{4032} \right].
\]  

This shows that the ratio \(b_1/a_1\) is reduced when increasing the laser intensity, reducing the contrast in the angular distribution. This is reminiscent to what was observed when combining XUV harmonics with an IR laser field: the contribution of higher angular momenta resulted in the decrease of the relative maxima in the angular distributions [10].

This reduced contrast in the angular distribution can be explained when performing a development of the Bessel function for larger value of its argument \(z\). We can, therefore, show that the ratio \(b_1/a_1\) becomes smaller with increasing laser intensity. This comes from that, the inclusion of different pathways, with higher angular momenta for the outgoing electron, leading to the formation of the first sideband results in destructive interference when the two fields are parallel. Moreover, the contrast for the second sideband is, as predicted by the ”soft-photon” approximation, larger (3.3 in the experiment and 3.5 in theory), although it does not reach the limiting value of \(2n + 1 = 5\).

4. Conclusion

We have investigated the validity of the soft-photon approximation to describe two-colour IR-XUV photoionization spectra in He, in the strong field regime. To this end, we have compared the results of the numerical resolution of the TDSE for a (3D) single-active model He atom to the predictions of the ”soft-photon” approximation, based on the use of Volkov wavefunctions to describe the ejected electron. The results presented here clearly demonstrate the usefulness of the soft-photon approximation in the perspective of interpreting the data for IR-XUV ionization processes in the strong field regime.

Acknowledgments

Parts of the computations have been performed at the Institut du Developpement et des Ressources en Informatique Scientifique (IDRIS).

References

[1] L. F. Di Mauro and P. Agostini, Adv. At.Mol. Opt. Phys. 35, 79 (1995).
[2] P. B. Corkum, Phys. Rev. Lett. 71, 1994 (1993).
[3] A. Maquet and R. Taïeb, J. Mod. Opt. 54, 1847 (2007).
[4] F.V. Bunkin and M.V. Fedorov, Sov. Phys.-JETP 22 844 (1966).
[5] A. Abramowitz and I.A. Stegun, Handbook of Mathematical Functions (NBS, Washington, USA, 1964).
[6] See for example V. Véniard, R. Taïeb, and A. Maquet, Phys. Rev. A 54, 721 (1996) and references therein.
[7] M. Meyer et al., Phys. Phys. A 74, 011401(R) (2006).
[8] P. Radcliffe et al., Appl. Phys. Lett. 90, 131108 (2007).
[9] M. Meyer et al., Phys. Rev. Lett. submitted.
[10] O. Guyétand et al., J. Phys. B 41, 051002 (2008).