



Brazilian Journal of Physics

ISSN: 0103-9733

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Sociedade Brasileira de Física  
Brasil

B. de Castro, A. Rubens; Moeller, Thomas; Wabnitz, H.; Laarmann, T.  
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Brazilian Journal of Physics, vol. 35, núm. 3A, september, 2005, pp. 632-635  
Sociedade Brasileira de Física  
São Paulo, Brasil

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## Dependence of Transition Probabilities for Non-Linear Photo-Ionization of He Atoms on the Structure of the Exciting Radiation Pulses

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Received on 10 February, 2005

We discuss the expected dependence of the probability transitions for 2-photon and 3-photon absorption in Helium gas on the spatial and temporal structure of the exciting radiation pulses. Regarding spatial structure, we assumed a Gaussian radial intensity distribution; we find, as expected, that the 2-photon and 3-photon processes become negligible at distances  $D$  away from the focus, where  $D$  is of the order of the beam waist FWHM. Regarding temporal structure, we compared transition probabilities for square, Gaussian and cosine squared temporal profiles; we find that for the same FWHM, Gaussian and cosine squared pulses give essentially the same transition probabilities, but the square pulses are about twice as efficient. We finally studied the effect of sharp versus smooth rise and fall edges in the light pulse; we find negligible correlation with the shape of the pulse edges, and strong correlation with the pulse FWHM, i.e., with pulse total energy, as might be expected.

### I. INTRODUCTION

The interaction of matter with intense pulses in the soft X-ray spectral range is very complicated and not well described by semi-classical concepts like field ionization and ponderomotive force. The new physics unearthed by such experiments is just now being analyzed and understood [1,2,3].

We have recently proposed a reasonably simple approach for the interpretation of the photo-electron spectra of Helium gas recorded under excitation with the soft X-ray pulses produced by the DESY FEL at  $h\nu=13$  eV (in the so-called “Phase 1” closed in March 2002) [4,5]. It is believed our approach could be generalized for more complicated systems illuminated with radiation of arbitrarily high intensity and time-dependence.

Our approach consists in the direct numerical solution of the time-dependent Schrödinger equation for the quantized radiation field in the presence of matter, using a limited basis set. Details such as the pulse temporal and spatial profile can be easily included in the formalism. In brief, our calculation starts from the Hamiltonian  $H_{\text{Helium}}+H_{\text{inter}}+H_{\text{Radiation}}$ , where  $H_{\text{inter}}$  is the electron-radiation interaction term, complete with  $\mathbf{A}\cdot\mathbf{p}$  and  $\mathbf{A}\cdot\mathbf{A}$  contributions. Here  $\mathbf{p}$  is the electronic linear momentum and  $\mathbf{A}$  is the operator for the potential vector, which, in the Schroedinger picture used here, is independent of time; all time-dependence is vested on the system quantum states, without loss of generality. As basis we take direct products of  $H_{\text{Helium}}$  and  $H_{\text{Radiation}}$  eigenstates. Define the system ket as a linear combination of the basis states with time-dependent coefficients. Go into the Interaction representation, and solve the resulting set of differential equations using implicit Runge-Kutta algorithms of high order, optimized for “stiff” systems. As initial condition we set the population of the ground state as unity and all others zero. It was checked that the results did not depend on the phase of the

initial state, and that the total probability was independent of time (to within better than one part in  $10^6$ ) during the full length of the numerical integration.

In the discussion of our Helium gas data, the simplest possible assumptions were made, since there is as yet only very little experimental knowledge on the structure of the DESY FEL pulses, beyond the evidence that the time envelopes, total energy and spectral composition change from pulse to pulse. The scarcity of data is a consequence of the novelty of these FEL facilities and of the severe experimental difficulties of dealing with extremely intense and fast pulses in the soft X-ray spectral range in an environment of ultra high vacuum. However, it is expected that a soft X-ray auto-correlator [6] will be available for the coming experiments in the upgraded (“Phase 2”) DESY FEL. This would allow much more detailed knowledge of the pulse temporal profile and the question of transition probability dependence on the pulse shape of the exciting radiation becomes very relevant. We have therefore studied how the transition probabilities for non-linear photo-ionization of Helium depend on the spatial and temporal profile of the exciting radiation.

Regarding spatial structure, we assumed a Gaussian radial intensity distribution; we find that the 2-photon and 3-photon processes become negligible at distances  $D$  away from the focus, where  $D$  is of the order of the beam waist FWHM.

Regarding temporal structure, we compared transition probabilities for square, Gaussian and cosine-squared temporal profiles; we find that for the same FWHM, Gaussian and cosine-squared pulses give essentially the same transition probabilities, but the square pulses are about twice as efficient. We finally studied the effect of *sharp* versus *smooth* rise and fall edges in the light pulse; we find negligible correlation with the shape of the pulse edges, and strong correlation with the pulse FWHM.

The results reported here were obtained using the codes described in [5], with parameters chosen arbitrarily for con-

venience of simulation as suggested by previous calculations made for He [7].

## II. EFFECT OF SPATIAL NON-HOMOGENEITY

In our experiments we collect in a time-of-flight (TOF) spectrometer, photo-electrons generated in a conical region of length  $L$  and half-angle  $\xi$ . The He gas density  $\rho_{He}$  is assumed to be uniform, but the light beam has a radial Gaussian profile with width  $\sigma(z)$ , where  $z$  is the axis of propagation of the radiation. The TOF can be moved, and in general  $L = z_2 - z_1$  while  $\sigma(z) = \sigma_o + \xi z$  if we place the origin at the focus. The intensity is therefore a function of  $r$  and  $z$ :

$$I(r, z) = I(z) \exp(-r^2/\sigma(z)^2) \quad (1)$$

$$\sigma(z) = \sigma_o + \xi z$$

$$I(z) = [\sigma_o/\sigma(z)]^2 I_{\text{focus}}$$

The number of photo-electrons generated inside a ring-like volume element is

$$dN_e = P(I(r, z)) \rho_{He} 2\pi r dr dz \quad (2)$$

where  $P$  is the probability for non-linear photo-ionization. The total number of photo-electrons collected is

$$N_e = \int_0^\infty \int_{z_1}^{z_2} P(I(r, z)) \rho_{He} 2\pi r dr dz \quad (3)$$

For a 3-photon absorption process,  $P(I) \approx \Pi^{(3-ph\ 13)}(I(r, z))^3$ , where  $\Pi^{(3-ph\ 13)}$  is independent of  $r, z$  but depends on  $I_{\text{focus}}$ . The superscript (n-ph 13) in  $\Pi$  is intended to remind the reader that it refers to n-photon absorption at  $h\nu = 13$  eV. We now can make the integrations to get

$$N_e = \Pi^{(3-ph\ 13)} I_{\text{focus}}^3 \rho_{He} (\sigma_o/\xi) (\pi/15) f_5(z_1, z_2) \quad (4)$$

$$f_n(z_1, z_2) = [1 + \xi z_1/\sigma_o]^{-n} - [1 + \xi z_2/\sigma_o]^{-n}$$

$$0 < z_1 < z_2 \quad n = 1, 2, 3, \dots$$

Table I (L=2 mm)

$z_1$ (mm)	$f_5$	$f_3$
-1	2.0	1.9
0	1.0	1.0
1	$6.1 \cdot 10^{-3}$	$4.3 \cdot 10^{-2}$
2	$4.9 \cdot 10^{-4}$	$8.8 \cdot 10^{-3}$
3	$9.0 \cdot 10^{-5}$	$2.9 \cdot 10^{-3}$

The maximum  $N_e$  occurs when  $-z_1 = z_2 = L/2$  and falls off very fast away from the focus, as shown in Table I, where we assumed  $L = 2$  mm.

Similarly, using  $P(I) \approx \Pi^{(2-ph\ 13)}(I(r, z))^2$  we get for the 2-photon absorption

$$N_e = \Pi^{(2-ph\ 13)} (I_{\text{focus}}/\sigma_o^2)^2 \rho_{He} (\sigma_o/\xi) (\pi/6) f_3(z_1, z_2) \quad (5)$$

and for single-photon absorption (of, say, the FEL 3<sup>rd</sup> harmonic at 39 eV) the result is, as expected, independent of  $z_1, z_2$ :

$$N_e = \Pi^{(single-ph\ 39)} (I_{\text{focus}} \sigma_o^2 L) \rho_{He} \pi \quad (6)$$

These results make clear the difficulty of even adjusting the equipment to measure photo-electrons from specific high-order non-linear processes; they are almost always masked by the presence of large backgrounds coming from linear processes, as demonstrated by our TOF data on the positive He ions generated in these linear and non-linear processes [4].

## III. EFFECT OF DIFFERENT TIME-DEPENDENT ENVELOPES

We compared the time evolution of the probability for 2-photon absorption at a peak intensity of  $3.0 \cdot 10^{14}$  W/cm<sup>2</sup> for three different envelopes. All the envelopes had the same FWHM duration  $T_{FWHM} = 100$  atomic units of time, unit maximum, and almost the same integrated area  $\approx T_{FWHM}$ . The pulse shapes were: (i) a quasi-rectangular pulse with smooth rise and fall edges; (ii) a Gaussian pulse  $\exp[-t^2/(T_{FWHM}/1.665)^2]$ ; (iii) a cosine-squared pulse  $\cos^2[\pi t/2T_{FWHM}]$ ,  $(-T_{FWHM} < t < T_{FWHM})$ .

It is necessary to specify just how these envelopes act, whether on the *intensity* itself or in the *vector potential field*.

Figure 1 shows the results. The quasi-rectangular pulse is clearly more efficient for the 2-photon process, because it maximizes the size of the coefficients in the coupled differential equations for the longest possible time. Modulation of the *field* with the Gaussian or cosine squared envelopes is less efficient in allowing 2-photon absorption than modulation of the *intensity*: since  $1 \geq$  envelope and it enters in the equations *squared*, its effective FWHM is less. For the quasi-rectangular pulse, on the contrary, because the envelope  $\approx 1$  most of the time, it does not matter whether this quantity is squared or not.

## IV. EFFECT OF THE WINGS OF THE FEL PULSE

There are indications in the literature [8] that in non-linear optical processes the transition probabilities are strongly dependent on whether the radiation field is turned on adiabatically or suddenly. In order to investigate this issue, we progressively changed from a sharp quasi-rectangular pulse to a smooth envelope of approximate cosine-squared profile, as given by

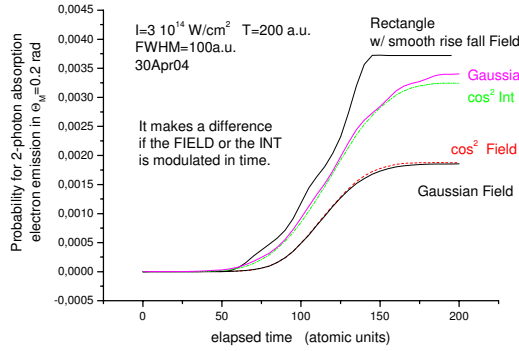


FIG. 1: Time evolution of the probability for 2-photon absorption in Helium for field intensity  $I=3 \cdot 10^{14} \text{ W/cm}^2$ , photon energy  $h\nu=13 \text{ eV}$ , total pulse length 200 atomic units, FWHM 100 atomic units. The results shown are for three different pulse shapes: Gaussian, cosine squared and rectangular with smooth rise and fall edges. Also, we show the effect of modulating the *vector potential field*, or of modulating the *field intensity*.

$$\begin{aligned}
 \text{envelope}(t) &= 1 - \cos^2[\pi t/t_1] & 0 < t < t_1 \\
 &= 1 & t_1 < t < t_1 + t_2 \\
 &= \cos^2[\pi(t - t_1 - t_2)/t_2] & t_1 + t_2 < t < 2t_1 + t_2 \\
 &= 0 & \text{elsewhere}
 \end{aligned} \quad (7)$$

The time duration  $t_2$  of the flat center of the pulse was kept constant at 100 atomic time units. The peak intensity was set to  $3.0 \cdot 10^{14} \text{ W/cm}^2$ . We modulated the *field*, not the intensity, in keeping with ref [8].

The full circles in Fig. 2 show the probability for 2-photon absorption as a function of the wing length  $t_1$ . Fig. 3 shows the same for the 3-photon absorption.

We also show (open squares) the probabilities for 2- and 3-photon absorption for He illuminated with rectangular pulses of duration equal to the FWHM of the smooth pulses. It is clear from the figures that most of the dependence on wing length  $t_1$  is actually explained by the attending increase in FWHM ( $t_{FWHM} = t_1 + t_2$ ), reinforcing the idea that the main effect comes from the integrated intensity, not from the particular shape of the light pulse.

## V. DISCUSSION AND CONCLUSIONS

We have used an approach based on the direct numerical solution of the Schrodinger equation to study the dependence of transition probabilities for Helium non-linear photo-ionization as a function of temporal pulse shape. We find, by comparing results for various pulse time-envelopes, that most of the dependence is described by just the FWHM of the pulses, not their shape. In particular, there is not any clear difference between adiabatic and sudden turn-on or turn-off of the excitation pulses.

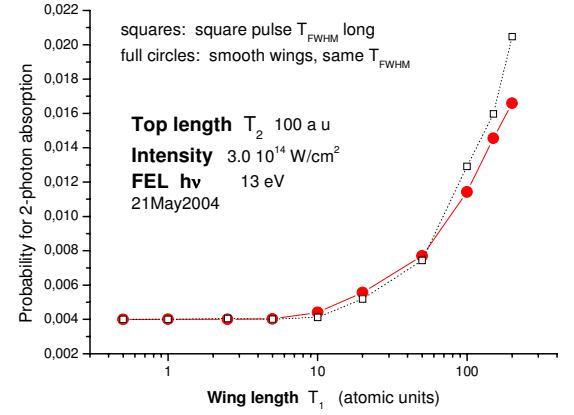


FIG. 2: Effect of variable wing length  $t_1$  on the probability for 2-photon absorption in Helium, same field conditions as in figure 1, except that, while  $t_2$  (the duration of the flat part of the pulse) is kept constant at 100 atomic units, the FWHM given by  $t_1 + t_2$  is, of course, not the same for each point. The squares show the effect of rectangular pulses with time duration  $t_1 + t_2$ .

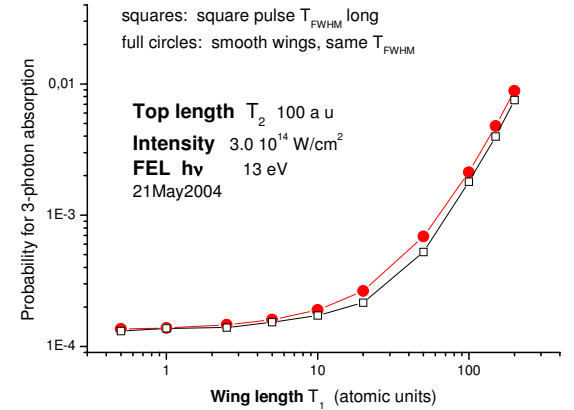


FIG. 3: Same as figure 2, but for 3-photon absorption in Helium.

As regards our method, let us observe that the direct numerical solution of the Schrodinger equation is a safe choice for new situations where the physics is still not altogether clear, but it requires a truncation of the infinite set of equations obtained for all systems of physical interest. In the present study, we included in the basis set 20 states which might be accessed, as allowed by symmetry and energy-momentum conservation. For low field amplitudes, the transition probabilities for non-linear 2-photon and 3-photon absorption obtained in our calculation scale as the square and cube of the field intensity, as expected, but depart from this behavior at higher intensities.

The near independence of the transition probabilities on the nature of the field turn-on and turn-off, as found in our simulations, is quite consistent with a very general approach recently proposed by Uiterwaal et al [9], where the effective cross-sections for non-linear processes induced by fast and extremely intense pulses are made to depend on *time-integrals* of

the excitation rather than on details of turn-on and turn-off.

The detailed dependence of multi-photon absorption on pulse shape seems at present to be an open question. Santra and Greene, in a recent publication [10], address the problem of Xenon multiple ionization described in one of our previous reports [3], with VUV radiation field intensities of up to  $10^{13}$  W/cm<sup>2</sup>. They used a technique similar to the one used by ourselves to explain our photo-emission data in Helium gas [4,5], although the treatment of the isolated atomic problem,

of the interaction radiation-matter, and of the quasi-free states is somewhat different. They solved rate equations to make estimates of Xe multiple ionization assisted by multi-photon absorption, assuming 10 different simulated FEL pulse shapes (all with the same total energy) but reported just the average result. It is therefore not clear which statements might be made about specific dependence of multi-photon absorption on pulse shape.

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