README file for "Two-photon Two-color transition matrix amplitudes"

D.I.R. Boll*[1], L. Martini[1], and O.A. Fojón[1,2]

[1]Instituto de Física Rosario, CONICET-UNR, Blvd. 27 de Febrero 210 bis, 2000 Rosario, Argentina
[2]Escuela de Ciencias Exactas y Naturales, FCEIA, Universidad Nacional de Rosario, Argentina

August 29, 2022

1 SOPT calculations

Author: Lara Martini

1.1 Files

1. Abs_I_SOPT.txt
2. Abs_II_SOPT.txt
3. Emi_I_SOPT.txt
4. Emi_II_SOPT.txt

1.2 Description

Each file contains the real and imaginary parts of the two-photon two-color radial matrix elements $T^0_{L,1}$, for hydrogen atoms with initial state 1s and final (continuum) states with angular momentum $L = 0$ and $L = 2$, respectively. The first column indicates the harmonic order for the XUV photon in units of the angular frequency of the IR assistant laser ($\omega_0 = 0.056954190$ au). Files are named according to:

- the IR photon process leading to final state, absorption (Abs) or emission (Emi) (see Fig. 1 in Ref. [1]), and
- time-ordering for the photon exchange, *i.e.* XUV-IR (I) and IR-XUV (II).

*boll@ifir-conicet.gov.ar

1.3 Method

Direct calculation based on Eqs. (19) to (22) from Ref. [3]. To obtain the radial matrix elements from those equations, we remove the Coulomb phase from them by taking the modulus of the Gamma function factors in Eqs. (19) and (20). Besides, we multiply the results from Eqs. (19) and (20) by

$$\frac{\sqrt{k}}{\sqrt{8\pi^3}}\sqrt{9} \text{ for } L=0$$
$$-\frac{\sqrt{k}}{\sqrt{8\pi^3}}\sqrt{\frac{45}{4}} \text{ for } L=2,$$

to remove the factors from the partial wave expansion and the corresponding angular algebra. The factor in front $\sqrt{k}/\sqrt{8\pi^3}$, with k the photoelectron momentum of the final continuum state, are not relevant. It only changes the overall normalization.

1.4 Observations

1. To obtain the continuum-continuum phases in Fig. (2) and (3) from Ref. [1], the Coulomb phases $\sigma_\lambda(\kappa)$ associated to the XUV ionization step MUST be removed, and the phase factor $(-i)^L \exp(i\sigma_L)$ MUST be added to the transition matrix amplitudes in the datasets listed above. As usual, these phases are given by $\sigma_l(k) = \arg[\Gamma(l+1-i/k)]$, with k being the photoelectron momentum associated to the intermediate or final state.

2. The radial matrix elements ratios showed in Fig. (4) from Ref. [1] are directly obtained from the datasets listed above.

2 Model calculations

Author: Diego I.R. Boll

2.1 Files

1. Model_A_L_I_ON_1.dat
2. Model_A_L_I_ON_2.dat
3. Model_A_L_I_ON_3.dat
4. Model_E_L_I_ON_1.dat
5. Model_E_L_I_ON_2.dat
6. Model_E_L_I_ON_3.dat

2.2 Description

Each file contains the modulus and phase of the two-photon two-color radial matrix elements $T^{l_i}_{L,\lambda}$, for final states with $L = \lambda - 1$ and $L = \lambda + 1$, corresponding to intermediate states with angular momentum λ. The first column indicates the kinetic energy of the final state in electronvolts. Files are named according to Model_$1_$2_$3_$4_$5.dat where:

- $1={A,E} indicates if the IR photon is Absorbed (A) or Emitted (E),
- $2={L} indicates that calculations were performed in the Length gauge (L),
- $3={I} indicates that calculations were performed for Individual (I) matrix elements,
- $4={ON} indicates that calculations were performed using the approximation $\kappa_\pm \simeq k \mp \omega_0/k$
- $5=$\lambda$ angular momentum of intermediate state

2.3 Method

Direct calculation from Eqs (6) and (8) in Ref. [1]. The radial matrix elements for the first step $\langle R_{\varepsilon_\pm,\lambda}|r|R_{\varepsilon_i,l_i}\rangle$ in Eq. (6) and the Coulomb phase factor $\exp[i\sigma_\lambda(\kappa)]$ in Eq. (8) are omitted for the calculations reported in the above files.

2.4 Observations

1. Using (or not) the approximation $\kappa_\pm \simeq k \mp \omega_0/k$ does not change the analysis and/or any of the conclusions in Ref. [1]. This approximation, related to the soft-photon approximation [2], only slightly improves the results. Its physical origin may be related to the fact that it applies to intermediate continuum states described through asymptotic wavefunctions, for which an asymptotic approximation for the photoelectron momentum gives better results. However, a further understanding of this approximation requires direct comparison of the approximate intermediate states with those obtained from the numerical resolution of the Dalgarno-Lewis method, which is beyond the scope of the present study.

2. The results for continuum-continuum phases we show in Fig. (2) from Ref. [1], are obtained from the above files after applying an *ad-hoc* phase-shift to the four phases for the absorption and emission matrix elements with $L = 0$ and $L = 2$, with the same kinetic energy. The *ad-hoc* phase shift is calculated as the difference between SOPT and Model phases for the emission pathway with $L = 2$. Clearly, this procedure has no physical implications as the transformation applied amounts to a global phase factor.

3. Because of the implementation we use for the Gauss hypergeometric function, the results do not change if 0^+ (see Eq. (8) in Ref. [1]) is identical to zero. However, the results obtained from some other implementations MAY change. In our tests, we found that a numerically safe approach would be to consider $0^+ \sim 1\times10^{-10}$ or smaller.

3 TDSE calculations

Author: Diego I.R. Boll

3.1 Files

1. Abs_TDSE.dat
2. Emi_TDSE.dat

3.2 Description

Each file contains the modulus and phase of the two-photon two-color radial matrix elements $T^0_{L,1}$, for hydrogen atoms with initial state 1s and final (continuum) states with angular momentum $L = 0$ and $L = 2$, respectively. The first column indicates the harmonic order for the XUV photon in units of the angular frequency of the IR laser (ω_0). Files are named according to:

- the IR photon process leading to final state, absorption (Abs) or emission (Emi) (see Fig. (1) from Ref. [1])

3.3 Method

Numerical resolution of the Time-dependent Schrodinger Equation (TDSE) with the Qprop code [4]. The relevant parameters to reproduce the calculations are the following (values in atomic units unless otherwise stated):

- Radial grid step: 0.24
- Cutoff radius: 850
- T-surff radius: 1700
- Number of angular momenta in expansion: 4
- Time step for propagation: 0.06
- IR intensity: 0.05 TW/cm^2
- IR angular freq. ω_0: 0.056954190
- IR pulse duration: 10 IR-cycles
- XUV pulse duration: 3 IR-cycles

The radial matrix elements reported in the files listed above correspond to a linear interpolation between adjacent calculations obtained from the Qprop code. Besides, after the interpolation procedure, we remove the angular (algebra) and time-delay $\exp(\pm i\omega\tau)$ factors to obtain the radial matrix elements from the transition amplitudes.

3.4 Observations

1. To obtain the continuum-continuum phases shown in Fig. (2) and (3) from Ref. [1], the Coulomb phases $\sigma_\lambda(\kappa)$ associated with the XUV ionization step MUST be removed from the transition matrix amplitudes in the datasets listed above.

2. The results for continuum-continuum phases we show in Fig. (2) from Ref. [1], are obtained from the above files after applying an *ad-hoc* phase-shift to the four phases for the absorption and emission matrix elements with $L = 0$ and $L = 2$, with the same kinetic energy. The *ad-hoc* phase shift is calculated as the difference between SOPT and TDSE phases for the emission pathway with $L = 2$. Clearly, this procedure has no physical implications as the transformation applied amounts to a global phase factor.

3. Take into account that moduli for radial matrix elements obtained from TDSE and SOPT calculations cannot be directly compared. The TDSE results contain information about the IR and XUV fields' amplitudes, whereas those parameters are not taken into account in the SOPT calculations. On the contrary, the ratios between partial wave radial matrix elements are well defined as the fields' amplitudes cancel.

4 Changelog

- Reference [1] updated. (DB 2022-08-29)

References

[1] D. I. R. Boll, L. Martini, and O. A. Fojón. Analytical model for attosecond time delays and fano's propensity rules in the continuum. *Phys. Rev. A*, 106:023116, Aug 2022.

[2] J. Dahlström, D. Guénot, K. Klünder, M. Gisselbrecht, J. Mauritsson, A. L'Huillier, A. Maquet, and R. Taïeb. Theory of attosecond delays in laser-assisted photoionization. *Chemical Physics*, 414:53–64, 2013.

[3] A. P. Jayadevan and R. B. Thayyullathil. Two-photon ionization of atomic hydrogen above the one-photon ionization threshold. *Journal of Physics B: Atomic, Molecular and Optical Physics*, 34(4):699–703, feb 2001.

[4] V. Tulsky and D. Bauer. Qprop with faster calculation of photoelectron spectra. *Computer Physics Communications*, 251:107098, 2020.