Multiphoton bound-bound transition rates

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We point out that recent calculations of multiphoton bound-bound transition rates are incorrect. The error lies in the angular part of the calculation.

In the course of a recent investigation of bound-bound multiphoton transitions in hydrogen-like atoms, we have found serious numerical discrepancies with a previous work by Gontier and Trahin.[1] We would like to suggest here that the source of these discrepancies probably lies in formula Eq. (11b) of that work.

As a matter of fact, in this expression, the angular dependence of the Nth-order transition amplitude, with respect to the polarization direction $\hat{\epsilon}'(\theta', \varphi')$ of the emitted photon is factorized as

$$\mathfrak{M}^{(N)}(\omega, \omega') = \tfrac{1}{3}\hat{\epsilon}\cdot\hat{\epsilon}'\{\cdots\}, \qquad (1)$$

where $\hat{\epsilon}$ is the unit polarization vector of the incident photons of energy ω, and ω' is the energy of the outgoing photon. By factorizing the scalar product $\hat{\epsilon}\cdot\hat{\epsilon}'$, it is implicitly assumed that any second-order bound-bound amplitude corresponding to either graphs A or B in Fig. 1 may be written as

$$M'^{(2)}_{A \text{ or } B}(n', l', m' | n, l, m) = \tfrac{1}{3}\hat{\epsilon}\cdot\hat{\epsilon}' \sum_{\lambda, \mu} \langle l', m' | \hat{r} | \lambda, \mu\rangle \cdot \langle \lambda, \mu | \hat{r} | l, m\rangle \, T^{(2)}_{\lambda}(W_{A \text{ or } B}), \qquad (2)$$

instead of

$$M^{(2)}_{A}(n', l', m' | n, l, m) = \sum_{\lambda, \mu} \langle l', m' | \hat{r}\cdot\hat{\epsilon}' | \lambda, \mu\rangle \langle \lambda, \mu | \hat{r}\cdot\hat{\epsilon} | l, m\rangle \, T^{(2)}_{\lambda}(W_A), \qquad (3a)$$

for graph A in Fig. 1, and

$$M^{(2)}_{B}(n', l', m' | n, l, m) = \sum_{\lambda, \mu} \langle l', m' | \hat{r}\cdot\hat{\epsilon} | \lambda, \mu\rangle \langle \lambda, \mu | \hat{r}\cdot\hat{\epsilon}' | l, m\rangle \, T^{(2)}_{\lambda}(W_B), \qquad (3b)$$

for graph B.

Here the amplitudes correspond to a second-order transition between the initial state $|n, l, m\rangle$ and final state $|n', l', m'\rangle$ of the atomic electron; $\hat{r}$ is the unit vector $\vec{r}/r$; $T^{(2)}_{\lambda}(W_k)$ is the corresponding reduced radial amplitude,

$$T^{(2)}_{\lambda}(W_k) = \langle n', l' | r G_{\lambda}(W_k) r | n, l\rangle, \qquad (4)$$

where

$$G_{\lambda}(W_k) = \sum_{\nu} \frac{|\nu, \lambda\rangle\langle \nu, \lambda|}{W_k - E_{\nu}} \qquad (5)$$

is the partial-wave projection of the atomic propagator, and W_k depends on the graph considered. In our case one has, respectively, $W_A = E_n + \omega$ and $W_B = E_n - \omega'$.

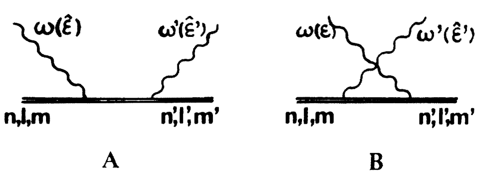

FIG. 1. Diagrams associated to the second-order bound-bound transition involving the absorption of one photon ω of the field, and the emission of one photon $\omega' = E_n + \omega - E_{n'}$, where E_n and $E_{n'}$ are the respective energies of the initial state $|n, l, m\rangle$ and final state $|n', l', m'\rangle$.

For computational convenience we use the $\hat{\epsilon}\cdot\hat{r}$ form of the dipole interaction operator instead of the equivalent $\hat{\epsilon}\cdot\hat{p}$ form used by Gontier and Trahin.[1] It should be noted, however, that this change does not affect the angular part of the calculation.

Then, if one adopts the simplifying hypothesis involved by Eq. (2), it is possible to rewrite the amplitude $M'^{(2)}(n', l', m' | n, l, m)$ as

$$M'^{(2)}_{A\text{ or }B}(n',l',m'|n,l,m)=\tfrac{1}{3}\hat{\epsilon}\cdot\hat{\epsilon}'\sum_{\lambda,\mu}[\langle l',m'|\rho_0|\lambda,\mu\rangle\langle\lambda,\mu|\rho_0|l,m\rangle + \langle l',m'|\rho_+|\lambda,\mu\rangle\langle\lambda,\mu|\rho_-|l,m\rangle + \langle l',m'|\rho_-|\lambda,\mu\rangle\langle\lambda,\mu|\rho_+|l,m\rangle]\,T^{(2)}_{\lambda}(W_{A\text{ or }B}), \qquad (6)$$

where

$$\rho_+=(1/r\sqrt{2})(x+iy)=(\tfrac{4}{3}\pi)^{1/2}Y_{1,1}(\hat{r}),\qquad \rho_-=\rho_+^*=-(\tfrac{4}{3}\pi)^{1/2}Y_{1,-1}(\hat{r}),\qquad \rho_0=z/r=(\tfrac{4}{3}\pi)^{1/2}Y_{1,0}(\hat{r}). \qquad (7)$$

By using the known formulas giving the integral over a product of three spherical harmonics[2] this amplitude may be expressed more symmetrically in terms of the Wigner 3-j symbols

$$M'^{(2)}_{A\text{ or }B}(n',l',m'|n,l,m)=\tfrac{1}{3}(\hat{\epsilon}\cdot\hat{\epsilon}')[(2l+1)(2l'+1)]^{1/2}\sum_{\lambda}T^{(2)}_{\lambda}(W_{A\text{ or }B})(2\lambda+1)\begin{pmatrix} l' & 1 & \lambda\\ 0 & 0 & 0\end{pmatrix}\begin{pmatrix} \lambda & 1 & l\\ 0 & 0 & 0\end{pmatrix} \times\sum_{q,\mu}(-1)^{m'+q+\mu}\begin{pmatrix} l' & 1 & \lambda\\ -m' & q & \mu\end{pmatrix}\begin{pmatrix} \lambda & 1 & l\\ -\mu & -q & m\end{pmatrix}. \qquad (8)$$

This latter expression may be transformed further, and after using the orthogonality properties of the 3-j coefficients, one gets the following result:

$$M'^{(2)}_{A\text{ or }B}(n',l',m'|n,l,m)=\frac{\hat{\epsilon}\cdot\hat{\epsilon}'}{3}\,\frac{1}{2l+1}[(l+1)T^{(2)}_{l+1}(W_{A\text{ or }B})+lT^{(2)}_{l-1}(W_{A\text{ or }B})]\delta_{l',l}\delta_{m',m}, \qquad (9)$$

which means that if this hypothesis were valid, any second-order transition would be forbidden unless $l'=l$ and $m'=m$. Such a result would mean that a second-order $s\rightarrow d$ transition could not be observed, which is clearly nonphysical. Moreover, as we will show, even for transitions with $l'=l$, the expression (9), as it stands is valid only for the particular case $l'=l=0$.

On the other hand, the correct calculation performed from Eq. (3a) or (3b) is more involved. For instance, the general formula corresponding to a second-order transition from an initial state $|n,l,m\rangle$ to a final state $|n',l+2,m'\rangle$ are respectively, if one assumes that the incoming photon ω is linearly polarized along the axis Oz:

$$M^{(2)}_{A}(n',l+2,m'|n,l,m)=[(2l+1)(2l+3)^2(2l+5)]^{-1/2} \times(\cos\theta'\{[(l+1)^2-m^2][(l+2)^2-m^2]\}^{1/2}\delta_{m',m} -\tfrac{1}{2}\sin\theta' e^{i\varphi'}\{(l+2-m)(l+3-m)[(l+1)^2-m^2]\}^{1/2}\delta_{m',m-1} +\tfrac{1}{2}\sin\theta' e^{-i\varphi'}\{(l+2+m)(l+3+m)[(l+1)^2-m^2]\}^{1/2}\delta_{m',m+1})\,T^{(2)}_{l+1}(E_n+\omega) \qquad (10a)$$

for graph A, Fig. 1, and

$$M^{(2)}_{B}(n',l+2,m'|n,l,m)=[(2l+1)(2l+3)^2(2l+5)]^{-1/2} \times(\cos\theta'\{[(l+1)^2-m^2][(l+2)^2-m^2]\}^{1/2}\delta_{m',m} -\tfrac{1}{2}\sin\theta' e^{i\varphi'}\{(l+1-m)(l+2-m)[(l+2)^2-(m-1)^2]\}^{1/2}\delta_{m',m-1} +\tfrac{1}{2}\sin\theta' e^{-i\varphi'}\{(l+1+m)(l+2+m)[(l+2)^2-(m+1)^2]\}^{1/2}\delta_{m',m+1})\,T^{(2)}_{l+1}(E_n-\omega') \qquad (10b)$$

for graph B, Fig. 1.

As another example, the second-order amplitude for a transition between states with the same angular momentum l and magnetic quantum numbers $m=m'=0$, is

$$M^{(2)}_{A\text{ or }B}(n',l,0|n,l,0)=\frac{\hat{\epsilon}\cdot\hat{\epsilon}'}{2l+1}\left(\frac{(l+1)^2}{2l+3}T^{(2)}_{l+1}(W_{A\text{ or }B})+\frac{l^2}{2l-1}T^{(2)}_{l-1}(W_{A\text{ or }B})\right), \qquad (11)$$

whatever the graph considered. It may be easily checked that this formula coincides with the Eq. (9) only in the case $l=0$.

As a consequence of these results it appears that the insertion of Eq. (9) in the expression of a transition amplitude for a N-photon process would lead to incorrect values of the corresponding cross sections. The only exception concerns the

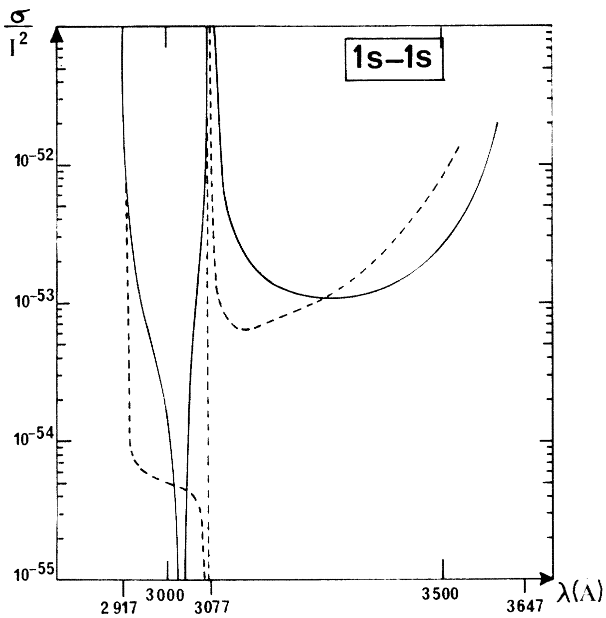

FIG. 2. Transition rate σ/I^2 in $cm^6 W^{-2}$ for the four-photon $1s \rightarrow 1s$ transition. The solid line represents our results. The dashed line corresponds to the results of Ref. 1.

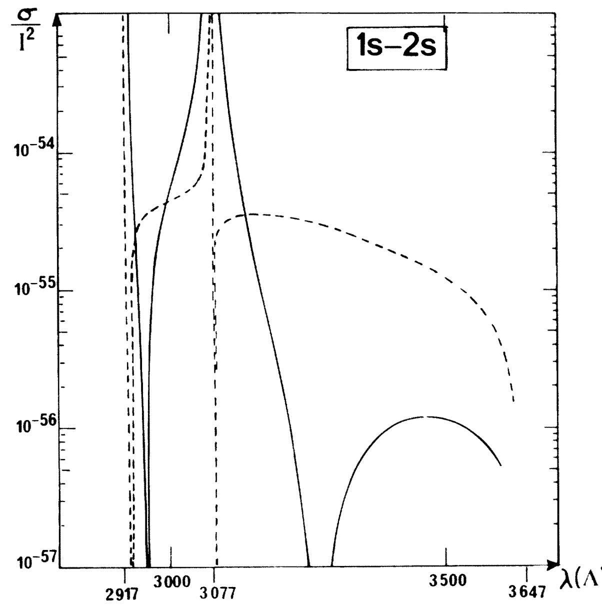

FIG. 3. Transition rate σ/I^2 expressed in $cm^6 W^{-2}$ for the four-photon $1s \rightarrow 2s$ transition. The solid line represents our results. The dashed line corresponds to the results of Ref. 1.

second-order transitions between S states, such as elastic or Raman scattering.

In Figs. 2 and 3 we compare Gontier and Trahin's results[1] with our own data for the four-photon $1s$-$1s$ and $1s$-$2s$ transitions in a hydrogen atom. The numerical evaluation of the corresponding fourth-order reduced radial amplitudes was carried out by using a Sturmian representation of the Coulomb Green's function.[3-5] The comparison shows that the discrepancies are notable, especially for the $1s$-$2s$ transition. More general and detailed results will be published elsewhere.

Finally we should mention that a similar error occurs in a subsequent paper by Biswas, Haque, and Mohan,[6] who studied multiphoton excitation processes involving the emission of two photons.

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