Two-photon above-threshold ionization of hydrogen over the photon energy range from 15 eV to 50 keV

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We investigate the absorption of two identical photons from the ground state of hydrogen-like atoms over an energy range that extends beyond that explored up to now. Our approach is based on a hybrid formula, valid in second-order perturbation theory, in which the $\mathbf{A}^2$ contribution from the nonrelativistic Hamiltonian is treated exactly, while the $\mathbf{A}\cdot\mathbf{P}$ contribution is calculated in dipole approximation. We find that, at least up to 50 keV, the nonrelativistic dipole approximation, based only on the $\mathbf{A}\cdot\mathbf{P}$ contribution, determines the values of the total cross section. Our numerical results, covering photon energies from 90 nm (13.7 eV) to 0.0248 nm (50 keV) are in very good agreement with most previous theoretical works. Differences with recent results are discussed.

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I. INTRODUCTION

The interest in the ionization of atoms through simultaneous absorption of two identical infrared photons started well before its experimental detection in 1962 [1], and it is still a challenging problem at shorter wavelengths. Well-documented studies of the nonlinear processes from the microwave to the ultraviolet frequencies do exist, and they led to numerous applications. With the emergence of x-ray free-electron lasers (XFEL) these studies have been extended to the extreme ultraviolet (euv) and x-ray photon energy range [2,3]. A recent experiment illustrates the application of a femtosecond kilovolt x-ray beam (at the SLAC National Accelerator Laboratory): with energy near the ionizing threshold of the 1*s* electrons of neon (1196 eV), Doumy *et al.* [4] have found evidence of the two-photon direct ionization of Ne^{8+}.

It is expected that new experimental results, such as those cited above, will stimulate the development of theoretical approaches able to describe nonlinear processes with intense and short pulses in the x-ray domain, which has not been thoroughly investigated up to now. Furthermore it will be necessary to study the case of complex (many-electron) atoms, which is far from being well understood in the context of multiphoton absorption. Keeping this objective in mind, we first focus on the case of one-active-electron atoms and ions. The purpose of the present theoretical study is to reconsider the case of the hydrogenic atom (fixed nucleus with charge Z) interacting with a monochromatic radiation. Our calculation is extended to photon energies as high as 50 keV; results above this energy, shown in order to clarify properties of the various contributions to the cross section, have to be reconsidered with relevant relativistic equations. The nonrelativistic analytic formulas we use are valid in second-order perturbation theory and are adequate for the case of most xuv and x-ray sources under construction, which are, in general, not too intense. Regarding the latter point, it is worth recalling that the ponderomotive energy of a quasifree electron in a focused x-ray beam (of the order of keV), with a peak intensity of $\sim 10^{17}$ W cm^{-2} produced at the Atomic Molecular and Optical Science (AMOS) end station at Linac Coherent Light Source (LCLS) [4], is totally negligible compared to the binding energy of the electron. Therefore nonperturbative effects, like recollision processes, are absent. This is the chief difference between nonlinear ionization in the x-ray field and its counterpart under a long wavelength at similar intensity. The immediate consequence is that lowest-order perturbation theory (LOPT) applies in the present case.

The hydrogen case has been extensively discussed and investigated *in the low photon energy range*, for energies close to (or smaller than) the photoeffect threshold at 91.18 nm; to our knowledge the shortest wavelength considered is 2 nm [5]. For details on the existing LOPT calculations, see the review of Maquet *et al.* [6] and the paper of Karule and Moine [7] (and references therein). An interesting analytic result is the derivation of the ratio between the maximum values of the cross sections for circular and linear polarization in the case of two- and three-photon absorption [8] and then for N-photon absorption [9,10]. Karule has obtained accurate values for generalized cross sections of two-photon ionization of the hydrogen ground state [5,11] and hydrogen excited states with principal quantum number n ranging from 2 to 8 [12]. Recently, Karule also investigated above-threshold ionization of *ns* hydrogen states with the absorption of up to four excess photons [13] in an energy range that extends from 90 to 10 nm.

In a recent paper, Varma *et al.* [14] explore two-photon ionization of H with photon energies up to 8 keV. LOPT is used and retardation effects are partially included in the nonrelativistic transition amplitude. It is expressed with two terms: the first one, $\mathbf{A}^2$ (where $\mathbf{A}$ is the vector potential in the

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Coulomb gauge), vanishes exactly in the dipole approximation (DA); the second one, due to the second-order contribution of the interaction term $\mathbf{A}\cdot\mathbf{P}$ (where $\mathbf{P}$ is the momentum operator), is treated within the DA. We adopt this approach in the present paper; we will put emphasis on the relative role of the contributions coming from $\mathbf{A}^2$ and $\mathbf{A}\cdot\mathbf{P}$ terms, using analytic formulas. As we will see, our conclusions differ significantly from those of Varma *et al.* For the $\mathbf{A}^2$ term, we also discuss the approximation where the final continuum state is treated like a free wave (neglecting the Coulomb effect).

Regarding analytic results based on Coulomb's Green's functions, a brief description of the situation of the problem is the following: *nonrelativistic* closed-form analytic formulas for the two-photon transition amplitude have been derived by Klarsfeld, both with retardation included [15] and in DA approximation [16]. The same expression of the transition amplitude can be easily obtained from Gavrila's analytic formulas describing K-shell Compton scattering [17,18]. In the nonrelativistic context the retardation effects related to the term $\mathbf{A}\cdot\mathbf{P}$ have never been evaluated. On the basis of a *relativistic* treatment, Koval *et al.* [19] and [20] have used the partial-wave expansions of the relativistic Coulomb Green's matrix and the continuum-state bispinor in order to obtain exact analytic expressions but not in closed form as in the nonrelativistic case, which is not tractable analytically. They were primarily interested in the case of medium- and high-Z hydrogen-like ions at two photon energies: 5% and 40% above the two-photon threshold. The effects of the nuclear charge Z on the cross section and electron angular distributions have been studied by the same authors [20], but not on their energy dependence.

This paper is organized as follows: the equations and analytic formulas are briefly presented in Sec. II; most of them are derived from previous works, and details are presented in Appendixes A and B. In Sec. III we present and discuss our results; they are compared with other calculations, and we also show cross sections at photon energies for which there are not available data in the literature. The figures and Table I cover both the cases of linearly and circularly polarized photons. When extending our calculation above 10 keV, we are guided by the peculiar features of other fundamental processes [21] in this energy domain. In particular, cancellation of relativistic and retardation effects is known to occur in one-photon absorption and Compton and Rayleigh scattering. The situation is analyzed in Appendix B for the $\mathbf{A}^2$ contribution to the total cross section, for which simple expressions based on algebraic transformations are given.

II. THEORETICAL APPROACH

We consider here the case of a hydrogen-like atom with a fixed nucleus of charge $-Ze$ (with the electron charge $e<0$). As explained in the Introduction, LOPT is used, which, for two-photon ionization, is of second order in the atom-photon interaction. This leads to the well-known expression for the *nonrelativistic* transition amplitude associated with two-photon ionization from a bound electron, *including retardation*,

$$\mathcal{M}_{\rm NR}^{\rm ret} = \langle\, E\mathbf{n}-\mid e^{\frac{2i}{\hbar}\boldsymbol{\kappa}\cdot\mathbf{r}}\mathbf{s}^2 - \frac{2}{m}e^{\frac{i}{\hbar}\boldsymbol{\kappa}\cdot\mathbf{r}}\mathbf{s}\cdot\mathbf{P} \times G(E_1+\hbar\omega+i\epsilon)\,\mathbf{s}\cdot\mathbf{P}\,e^{\frac{i}{\hbar}\boldsymbol{\kappa}\cdot\mathbf{r}}\mid E_1\rangle, \quad \epsilon\to 0^+, \tag{1}$$

$$G(\Omega) = \sum_n \frac{\mid n\,\rangle\langle\, n\mid}{E_n-\Omega}, \tag{2}$$

where E_1 is the initial electron energy, $\boldsymbol{\kappa}$ is the photon momentum, and $\hbar\omega$ is its energy. The emitted electron has an energy E and an asymptotic direction characterized by the unity vector $\mathbf{n}$; the corresponding energy eigenfunction $\langle\,\mathbf{r}\mid E\mathbf{n}-\,\rangle$ is normalized on the energy and solid-angle scales, and it has the ingoing asymptotic behavior. This leads to a matrix element with the same dimension as $\sqrt{1/E}$. The vector $\mathbf{s}$ is the polarization vector of the photon, normalized as $\mathbf{s}^*\cdot\mathbf{s}=1$; it is real for linear polarization, and otherwise, it is complex. As mentioned in the Introduction, in the case of ground-state hydrogen-like atoms, analytic expressions for the two-photon transition amplitude have been already established.

The final energy of the electron is

$$E = E_1 + 2\hbar\omega\,, \qquad \omega \geqslant \frac{\mid E_1\mid}{2\hbar}\,, \qquad E_1 = -\frac{\lambda^2}{2m}\,, \qquad \lambda = \alpha Zmc, \tag{3}$$

where m is the electron mass, c is the velocity of light, and α is the fine-structure constant. The two-photon absorption threshold is half of the photoelectric threshold. For low-Z elements this nonrelativistic threshold is low (it reaches 1.06 keV for $Z=13$), so the absorption of two photons in the low energy range is well described in DA. Given that the contribution of the first term in the matrix element given in Eq. (1) (i.e., the $\mathbf{A}^2$ contribution, called the sea-gull term in the literature of Compton scattering) vanishes in DA, the absorption of low-energy photons has been described in the literature by using the second term in Eq. (1), taken in DA.

For photon energies ranging from the two-photon to the one-photon (photoelectric) ionization thresholds, ionization is energetically possible only through the absorption of at least two photons. Above the second threshold one-photon and two-photon ionization are both possible.

As mentioned in the Introduction, there are no published results for photon energies above 2 nm (620 eV). In order to extend the calculations to higher energies, we use a hybrid approach where we consider the contribution of both terms in Eq. (1), but retardation is included only in the first term. Under the latter assumption the transition amplitude reads

$$\mathcal{M}_{\rm NR}^{\rm hyb} = \langle\, E\mathbf{n}-\mid e^{\frac{2i}{\hbar}\boldsymbol{\kappa}\cdot\mathbf{r}}\mathbf{s}^2 - \frac{2}{m}\mathbf{s}\cdot\mathbf{P} \times G(E_1+\hbar\omega+i\epsilon)\mathbf{s}\cdot\mathbf{P}\mid E_1\,\rangle\,, \qquad \epsilon\to 0^+. \tag{4}$$

As already mentioned, our approach is close to the one adopted by Varma *et al.* [14]; the difference is that we do not use approximations to evaluate the different terms in Eq. (4). A similar approach has been recently used by Drukarev *et al.* [22] in a study of Compton scattering in the low-energy domain.

We reproduce here the structure of the matrix element (4), using notations close to [17] and [18],

$$\mathcal{M}_{\rm NR}^{\rm hyb} = \mathcal{M}_{\mathbf{A}^2} + \mathcal{M}_{\mathbf{A}\cdot\mathbf{P}}^{\rm DA}, \tag{5}$$

$$\mathcal{M}_{\mathbf{A}^2} = \mathcal{O}^{\rm abs-two}\,\mathbf{s}^2\,, \qquad \mathcal{M}_{\mathbf{A}\cdot\mathbf{P}}^{\rm DA} = -2\,[\,\mathcal{P}^{\rm DA}\,\mathbf{s}^2 + \mathcal{T}^{\rm DA}\,(\mathbf{s}\cdot\mathbf{n})^2\,]\,. \tag{6}$$

The analytic expressions used for the invariant amplitudes $\mathcal{O}^{\text{abs-two}}$, $\mathcal{P}^{\text{AD}}$, and $\mathcal{T}^{\text{AD}}$ are obtained from Eqs. (23), (81), and (82) of [17], respectively, by using $\boldsymbol{\kappa}_2 = -\boldsymbol{\kappa}_1 \equiv -\boldsymbol{\kappa}$ (in the case of radiation scattering $\boldsymbol{\kappa}_1$ is the absorbed photon momentum and $\boldsymbol{\kappa}_2$ is the emitted photon momentum). The invariant amplitudes $\mathcal{P}^{\text{DA}}$ and $\mathcal{T}^{\text{DA}}$ are given in Appendix A; see Eq. (A1). They are expressed in terms of three Appell's functions F_1 in which appropriate expressions are given to the parameters and variables.

Appendix B gives the analytic expression of $\mathcal{O}^{\text{abs-two}}$ in Eq. (B2). We define first the matrix element (B1), which becomes $\mathcal{O}^{\text{abs-two}}$ if the explicit expression of the parameters λ and η (related to the nuclear charge) appearing in the initial (ground state) and final (continuum) wave functions, respectively, are used together with the specification of $\mathbf{K}$ and the use of the conservation energy law. This allows us to do two different operations : (i) to distinguish easily the contributions of the plane wave and first-order Born approximations and (ii) to follow the effect of the energy conservation on $\sigma_{\mathbf{A}^2}$, the contribution of $\mathbf{A}^2$ to the total cross section. As a result, we show that the zero-order (plane wave) and first-order Born approximations are of the same order in αZ. Then we derive simple formulas for $\sigma_{\mathbf{A}^2}$: Eq. (B15) in the nonrelativistic case and Eq. (B18) based on the relativistic energy conservation. The nonrelativistic formula has spurious poles, which are not present in (B18). This situation is connected with the more general problem of cancellation between relativistic and retardation effects [21]. In Sec. III the particularities of $\sigma_{\mathbf{A}^2}$, one connected with the Born approximation and the other with the energy conservation, are illustrated (see Fig. 3).

The transition rate $d\Gamma$ for two-photon ionization is proportional to the second power of the photon flux J, defined as the number of photons crossing the unity surface in the unity of time. In the literature one can find two definitions for a "generalized cross section." Dividing $d\Gamma$ by J leads to a quantity $\widetilde{d\sigma}$ with the dimension of a surface. Some authors divide it once more by J, getting a cross section independent of the photon flux,

$$d\sigma = \frac{\widetilde{d\sigma}}{J} = \frac{d\Gamma}{J^2}, \tag{7}$$

leading to a generalized cross sections in cm^4 s. Other authors divide $\widetilde{d\sigma}$ by the radiation intensity $I = \hbar\omega J$ and get

$$d\sigma_{\text{gen}} = \frac{\widetilde{d\sigma}}{I} = \frac{d\Gamma}{\hbar\omega J^2}. \tag{8}$$

They express their results in $\text{cm}^4\ \text{W}^{-1}$. We adopt definition (7), which leads to

$$d\sigma^{\text{hyb}} = \frac{2\pi^3\alpha^2}{\omega^2}\frac{\hbar^3}{m^2}\left|\mathcal{M}_{\text{NR}}^{\text{hyb}}\right|^2 d\Omega_{\mathbf{n}}, \quad E = E_1 + 2\hbar\omega. \tag{9}$$

For angular distributions, we use a Cartesian system of reference with the z axis taken along the initial photon momentum $\boldsymbol{\kappa}$. In the case of linear polarization the x axis is taken along the polarization vector. The polar angles of $\mathbf{p}$ are denoted by θ and ϕ. The total cross section σ^{hyb} is obtained by integrating over the direction of the emitted electron. The amplitude $\mathcal{O}^{\text{abs-two}}$ depends on the electron angle θ but not on the azimuthal angle ϕ. The amplitudes $\mathcal{P}^{\text{DA}}$ and $\mathcal{T}^{\text{DA}}$ do not depend on angles.

Following the work of Varma *et al.*, one of our objectives is to establish under which conditions the term $\mathcal{M}_{\mathbf{A}^2}$ comes into play and to identify the energy region where the interference with the term $\mathcal{M}_{\mathbf{A}\cdot\mathbf{P}}^{\text{DA}}$ becomes non-negligible. To this purpose, we write the differential cross section (DCS) as the sum of three terms:

$$d\sigma^{\text{hyb}} = d\sigma_{\mathbf{A}^2} + d\sigma_{\mathbf{A}\cdot\mathbf{P}}^{\text{DA}} + d\sigma_{\text{interf}}. \tag{10}$$

The same splitting is done for the total cross section

$$\sigma^{\text{hyb}} = \sigma_{\mathbf{A}^2} + \sigma_{\mathbf{A}\cdot\mathbf{P}}^{\text{DA}} + \sigma_{\text{interf}}. \tag{11}$$

The angular integration is performed analytically for the three separate terms, but it is not straightforward to extract an analytic expression for the interference term.

In the case of circular polarization, as $\mathbf{s}^2 = 0$, the DCS reduces to

$$d\sigma_{\text{circ}}^{\text{hyb}} = \frac{2\pi^3\alpha^2}{\omega^2}\frac{\hbar^3}{m^2} \mid \mathcal{T}^{\text{DA}} \mid^2 \sin^4\theta\, d\Omega. \tag{12}$$

For the cross section $\sigma_{\mathbf{A}\cdot\mathbf{P}}^{\text{DA}}$, which dominates (at least up to photon energies of 50 keV, as we shall see below), a simple scaling low is valid: $Z^6\sigma_{\mathbf{A}\cdot\mathbf{P}}^{\text{DA}}(Z,\omega) = \sigma_{\mathbf{A}\cdot\mathbf{P}}^{\text{DA}}(1,\omega/Z^2)$ [23].

III. DISCUSSION AND NUMERICAL RESULTS

The evaluation of $\mathcal{M}_{\mathbf{A}^2}$ is straightforward (see Appendix B); for $\mathcal{M}_{\mathbf{A}\cdot\mathbf{P}}^{\text{DA}}$ one needs to evaluate the three Appell functions described in Appendix A; see Eq. (A2). We calculate them by using their standard integral representation (an integral on the real axis extended from 0 to 1 [24]), a method we already tested in the past [25]. Note that the procedure cannot be applied for $\tau \geqslant 2$ [τ is defined in Eq. (A6)] due to the presence of a singularity at the origin in the integrand. This limit corresponds to a photon energy of $3/8 \mid E_1 \mid$ ($\mid E_1 \mid$ is the photoeffect threshold).

Figure 1 refers to the low-energy region, ranging from 13.7 to 800 eV. The contributions $\sigma_{\mathbf{A}^2}$ and $\sigma_{\mathbf{A}\cdot\mathbf{P}}^{\text{DA}}$ are shown separately. The first quantity is smaller than the second one by orders of magnitude, so it is shown multiplied by the factor 2×10^3. It is evident that *the nonrelativistic dipole approximation is valid* within this photon energy range. For circularly polarized radiation there is no contribution from the $\mathbf{A}^2$ term of the interaction. The comparison with other calculations for the total cross section $\sigma_{\mathbf{A}\cdot\mathbf{P}}^{\text{DA}}$ shows an excellent agreement, both for linearly and circularly polarized photons. At the photon energy of 620 eV (2 nm), our result for σ_{gen}, defined in Eq. (8), coincides with that published by Jayadevan and Thayyullathil [26] with four significant digits.

Our calculations disagree with the results presented by Varma *et al.* [14] for the case of linear polarization. Our cross sections are much larger than the ones presented in Fig. 1 of [14], and the disagreement increases with the photon energy: at 600 eV our results are two orders of magnitude higher than theirs.

We present in Fig. 2 our results for photon energies ranging from 0.8 to 50 keV, with the two contributions shown separately since the interference term is negligible. Here we compare with Fig. 2 of [14], which refers to linear polarization and photon energies below 10 keV. Now the $\mathbf{A}^2$ term is in agreement with the curve labeled $\mathbf{A}^2$ (Coulomb wave) in

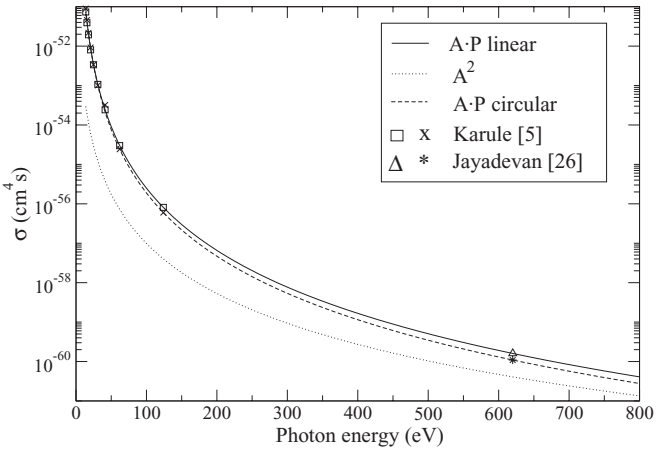

FIG. 1. The contribution $\sigma^{\mathrm{DA}}_{\mathbf{A}\cdot\mathbf{P}}$ to the total cross section [see Eq. (11)] for linearly (solid line) and circularly (dashed line) polarized photons vs the photon energy in the range 13.7–800 eV. Other calculations are also presented; see the legend. The much smaller contribution $\sigma_{\mathbf{A}^2}$ is also shown (magnified by a factor of 2×10^3).

Fig. 2 of [14], but the results for $\sigma^{\mathrm{DA}}_{\mathbf{A}\cdot\mathbf{P}}$ disagree again. Figure 2 shows that the nonrelativistic dipole term continues to give the dominant contribution; the crossing displayed in Fig. 2 of [14] is not observed in our data, with the two curves being almost parallel. We illustrate the differences between our results for $\sigma_{\mathbf{A}\cdot\mathbf{P}}$ (expressed in cm^4 s) and those given in [14] at 5 and 8 keV, where our calculations give 2×10^{-65} for 5 keV and 1.5×10^{-66} for 8 keV. These two latter values have to be compared with the cross sections extracted from Fig. 2 of [14], which are close to 4.2×10^{-68} for 5 keV and 2×10^{-69} for 8 keV. The origin of such a large discrepancy is unclear; nevertheless, we have to remark that the calculation of $\mathbf{A} \cdot \mathbf{P}$ contribution in [14] is based on an approximation [see Eqs. (11) and (12) of [14]], which requires a better justification, while we use exact analytic expressions.

For the range 5–50 keV the difference between $\sigma_{\mathbf{A}^2}$, expressed with the plane-wave approximation [see the discussion in Appendix B and Eq. (B19)], and the exact result is shown in Fig. 3.

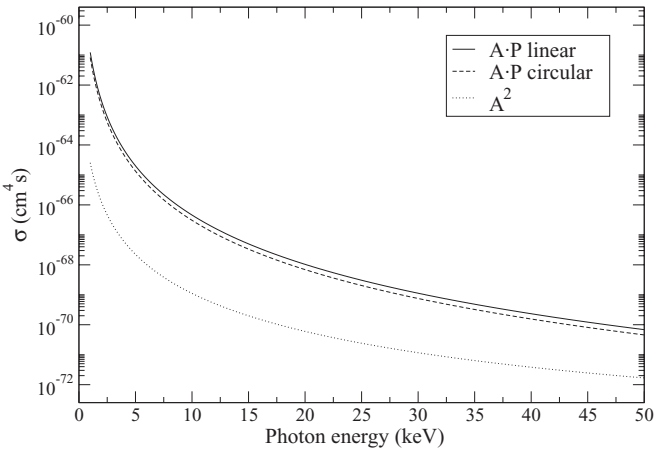

FIG. 2. The term $\sigma^{\mathrm{DA}}_{\mathbf{A}\cdot\mathbf{P}}$ in the total cross section [see Eq. (11)] for linearly (solid line) and circularly (dashed line) polarized photons vs the photon energy in the range 0.8–50 keV. The quantity $\sigma_{\mathbf{A}^2}$ is also shown (dotted line).

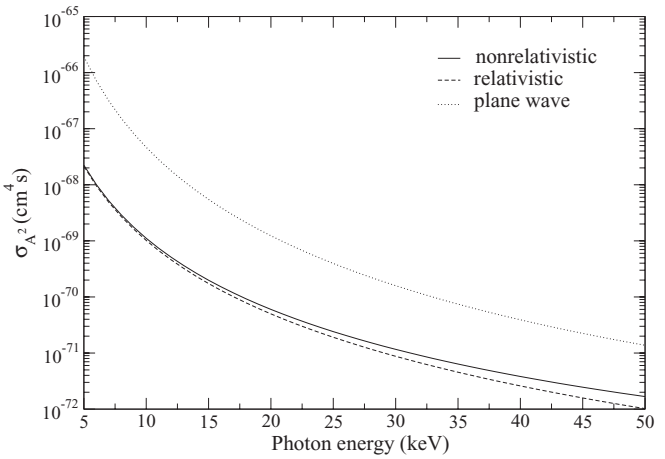

FIG. 3. The term $\sigma_{\mathbf{A}^2}$ of the total cross section [see Eq. (11)] vs the photon energy in the range 5–50 keV and linear polarization. Solid line: calculation with the nonrelativistic energy conservation law [see Eq. (3)]; dashed line: calculation with the relativistic law [see Eq. (B16)]. The dotted line is based on the plane-wave approximation; see Eq. (B19).

Table I gives, at fixed photon energies, the total cross section for circularly polarized photons (third column). For linear polarization, the fourth column gives the contribution $\sigma^{\mathrm{DA}}_{\mathbf{A}\cdot\mathbf{P}}$, which up to 10 keV is practically identical to σ^{hyb}, which is shown in the fifth column (only when they are different). For larger photon energies differences of the order of a few percent appear.

Above 10 keV we enter a domain where both retardation and relativistic effects have to be included, but we expect these

TABLE I. The total cross sections $\sigma^{\mathrm{circ}}_{\mathbf{A}\cdot\mathbf{P}}$, $\sigma^{\mathrm{lin}}_{\mathbf{A}\cdot\mathbf{P}}$, and σ^{hyb} given in cm^4 s for photon energy in the 0.6–50 keV range.

E (keV)	λ (Å)	$\sigma^{\mathrm{circ}}_{\mathbf{A}\cdot\mathbf{P}}$	$\sigma^{\mathrm{lin}}_{\mathbf{A}\cdot\mathbf{P}}$	σ^{hyb}
0.6	20.66	1.30×10^{-60}	1.91×10^{-60}	
0.7	17.71	5.47×10^{-61}	8.36×10^{-61}	
0.8	15.49	2.76×10^{-61}	4.08×10^{-61}	
0.9	13.77	1.46×10^{-61}	2.16×10^{-61}	
1	12.40	8.31×10^{-62}	1.23×10^{-61}	
1.25	9.91	2.49×10^{-62}	3.69×10^{-62}	
1.5	8.26	9.28×10^{-63}	1.38×10^{-62}	
1.75	7.08	4.03×10^{-63}	5.99×10^{-63}	
2	6.19	1.95×10^{-63}	2.91×10^{-63}	
3	4.13	2.16×10^{-64}	3.23×10^{-64}	
4	3.10	4.52×10^{-65}	6.76×10^{-65}	
5	2.47	1.34×10^{-65}	2.00×10^{-65}	
6	2.06	4.97×10^{-66}	7.43×10^{-66}	
8	1.55	1.03×10^{-66}	1.54×10^{-66}	
10	1.24	3.06×10^{-67}	4.58×10^{-67}	4.61×10^{-67}
15	0.83	3.33×10^{-68}	5.00×10^{-68}	5.04×10^{-68}
20	0.62	6.91×10^{-69}	1.03×10^{-68}	1.04×10^{-68}
25	0.49	2.03×10^{-69}	3.05×10^{-69}	3.10×10^{-69}
30	0.41	7.51×10^{-70}	1.12×10^{-69}	1.15×10^{-69}
40	0.31	1.55×10^{-70}	2.32×10^{-70}	2.40×10^{-70}
50	0.25	4.57×10^{-71}	6.85×10^{-71}	7.15×10^{-71}

effects to be small below 40–50 keV. The interaction of low-Z hydrogen-like atoms with electromagnetic radiation up to 50-keV photon energy is usually treated within *nonrelativistic quantum theory* and, for fields of moderate intensities, in perturbation theory. The most studied case is that of one-photon absorption (atomic photoeffect). For low energies, the dipole approximation is usually adopted, with retardation corrections being small. Relativistic calculations indicate a cancellation between relativistic and retardation corrections [21,27]. In the Coulomb case this cancellation was analyzed by Costescu and Spanulescu [28] for K-shell Compton scattering and by Costescu *et al.* [29] for K-shell Rayleigh scattering, and the consequences on photoeffect were also discussed. When handled directly, the cancellation occurring in the photoeffect becomes very transparent. In the latter processes it appears that the nonrelativistic total cross sections calculated within DA provide a better approximation than the total nonrelativistic cross sections including retardation. Angular distributions are much more sensitive to retardation effects at photon energies where relativistic effects are negligible.

A complete analysis of the case of two-photon absorption would require working with the expression [Eq. (1)] of the transition amplitude, which includes retardation in the $\mathbf{A}\cdot\mathbf{P}$ contribution, leading to a more complicated analytic expression than in the DA version [15]. Nevertheless, as in the cases mentioned before (K-shell Compton and Rayleigh scattering), we have to also consider relativistic effects, in particular the differences coming from the use of the relativistic instead of nonrelativistic energy conservation formulas. In order to illustrate this aspect, we have considered the simpler case of $\sigma_{\mathbf{A}^2}$. As already mentioned in Sec. II, using relation (B16), which expresses the energy conservation for the relativistic energies of the bound and final electrons, we end up with Eq. (B18) instead of Eq. (B15), based on the nonrelativistic Eq. (3). The comparison between the two expressions shows that the differences become visible at a photon energy of 10 keV, as shown in Fig. 3, covering the photon energy range 5–50 keV. These differences increase with the photon energy in the photon energy range 50–200 keV; see Fig. 4.

Figure 4 covers, for linearly polarized photons, energies above 50 keV, beyond the expected region of validity of nonrelativistic equations. The purpose is to show the behavior with increasing energy of the three contributions to the total cross section (11). We have considered the two possibilities for the energy conservation (relativistic or nonrelativistic energy conservation law) to calculate the first term on the right-hand side of Eq. (5). They practically do not affect the interference term in Eq. (11); therefore we show $\sigma_{\mathbf{A}^2}^{\mathrm{NR}}$ and $\sigma_{\mathbf{A}^2}^{\mathrm{rel}}$, which cause the differences in the total cross section. The dominant contribution continues to be given by $\sigma_{\mathbf{A}\cdot\mathbf{P}}^{\mathrm{DA}}$, so the two curves for the total cross section are close. We note that for the nonrelativistic quantity $\sigma_{\mathbf{A}^2}^{\mathrm{NR}}$ tends to increase, leading to a crossing at 179 keV with the curve representing $\sigma_{\mathbf{A}\cdot\mathbf{P}}^{\mathrm{DA}}$. The increase is related to the presence of spurious poles at $mc^2(1\pm i\alpha Z)$, as explained in Appendix B. For the same reason, at shorter wavelengths this term would show a maximum if the nonrelativistic conservation energy law is applied, while it decreases monotonically if relativistic kinematics is considered. We expect a similar modification in the second term of (1), which was treated here in DA.

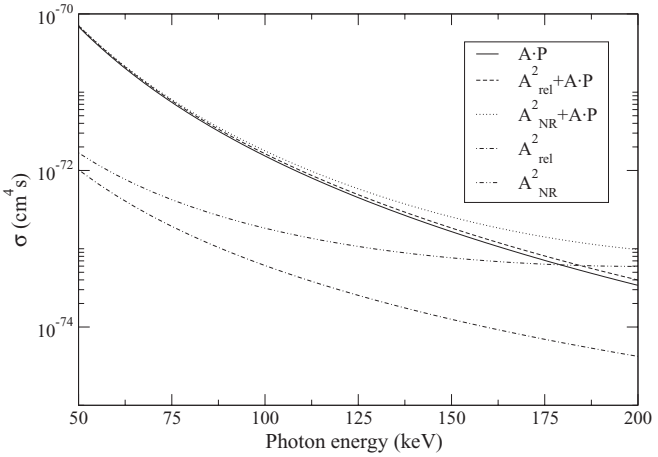

FIG. 4. The total cross section σ^{hyb} and the term $\sigma_{\mathbf{A}^2}$ [see Eq. (11)] using the relativistic and nonrelativistic conservation laws expressed in Eqs. (B16) and (3), respectively. The photon energies vary from 50 to 200 keV, and linear polarization is considered.

Nevertheless, as below 50 keV the deviations from DA should not be significant, at least for low Z, we expect that our results for $Z=1$, displayed in Figs. 1 and 2 and in Table I, should be very close to the exact fully relativistic ones. Our next objective is to investigate the case of hydrogen-like ions and the retardation effects on the $\mathbf{A}\cdot\mathbf{P}$ contribution.

IV. CONCLUSIONS

Our study of two-photon ionization of hydrogen in a fundamental state covers a photon energy range starting from the infrared region, which has been extensively investigated in the past, to a maximum value of 200 keV. The two-photon amplitude has been expressed in analytic form, using the nondipole approximation for the term $\mathbf{A}^2$. Although we have found differences with the recent calculations of Varma *et al.* [14], our values agree very well with the other calculations. The main conclusion is that, at least up to 50 keV, the nonrelativistic dipole approximation formula, based on the second-order contribution of the $\mathbf{A}\cdot\mathbf{P}$ of the electron-photon interaction, is valid. The $\mathbf{A}^2$ term of the interaction, which was suspected to bring a retardation correction in the keV range [14], was found to give a small contribution. Nevertheless, if the difference between calculations based on dipole approximation and full calculation is small at 50 keV, it is not negligible. We have thoroughly investigated various approximations used to calculate $\mathbf{A}^2$ in order to clarify the discrepancy with previous results and to present its sensitivity to relativistic corrections.

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APPENDIX A: THE DIPOLE APPROXIMATION ANALYTIC RESULTS

The dipole approximation result, coming from the term $\mathbf{A}\cdot\mathbf{P}$ in the Hamiltonian, considered in second-order perturbation theory, is determined by the two invariant amplitudes $\mathcal{P}^{\mathrm{DA}}$ and $\mathcal{T}^{\mathrm{DA}}$ in (6). Their compact analytic expressions, obtained using [17], as described after Eq. (6), are

$$\mathcal{P}^{\mathrm{DA}}(\Omega)=N(X-ip)^{-i\eta-3}(X+ip)^{i\eta-1}\frac{\tau}{(1+\tau)^4}\frac{f_1}{2-\tau},$$
$$\mathcal{T}^{\mathrm{DA}}(\Omega)=2N(X-ip)^{-i\eta-3}(X+ip)^{i\eta-3}\frac{p^2(1-i\eta)(2-i\eta)}{(1+\tau)^4}\times\left[\frac{f_2}{2-\tau}-\left(\frac{1-\tau}{1+\tau}\right)^4\frac{f_3}{4-\tau}\right], \tag{A1}$$

with f_1, f_2, and f_3 being the three Appell functions,

$$f_1=F_1(2-\tau;3+i\eta,1-i\eta;3-\tau;x,y), \tag{A2}$$

$$f_2=F_1(2-\tau;3+i\eta,3-i\eta;3-\tau;x,y),$$
$$f_3=F_1(4-\tau;3+i\eta,3-i\eta;5-\tau;x,y), \tag{A3}$$

depending on the variables

$$x=\frac{X-\lambda}{X+\lambda}\frac{X+ip}{X-ip},\qquad y=\frac{X-\lambda}{X+\lambda}\frac{X-ip}{X+ip}. \tag{A4}$$

The argument Ω of the amplitudes $\mathcal{P}^{\mathrm{DA}}$ and $\mathcal{T}^{\mathrm{DA}}$ comes from the Green's function in (4) and has the expression

$$\Omega=E_1+\hbar\omega+i\epsilon,\qquad \epsilon\to 0^+. \tag{A5}$$

It determines the two quantities X and τ in the previous expressions,

$$X^2=-2m\Omega,\quad \mathrm{Re}X\geqslant 0,\qquad \tau=\frac{\lambda}{X}. \tag{A6}$$

The quantity η is introduced by the continuum wave function (B5), and the constant factor N is

$$N=(32/\pi)(2\lambda^5pm)^{1/2}\Gamma(1-i\eta)\exp\left(\frac{\pi}{2}\eta\right),$$
$$\eta=\lambda/p,\qquad \lambda=\alpha Zmc. \tag{A7}$$

Between the two-photon and one-photon absorption thresholds, one has $\Omega<0$, and consequently, X is real. Above the photoelectric threshold X is purely imaginary, namely, $X=-i\mid X\mid$.

APPENDIX B: ON THE CONTRIBUTION OF THE $\mathbf{A}^2$ TERM

The contribution of the $\mathbf{A}^2$ term comes from the first term in (4). In order to discuss it in more detail, we consider the integral

$$\mathcal{O}=\langle E\mathbf{n}-\mid e^{\frac{i}{\hbar}\mathbf{K}\cdot\mathbf{r}}\mid E_1\rangle,\qquad E=p^2/2m,\qquad \mathbf{n}=\mathbf{p}/p, \tag{B1}$$

with the continuum energy eigenfunction normalized in the energy and solid-angle scale. Due to its behavior toward the rotation, $\mathcal{O}$ does not depend on $\mathbf{p}$ and $\mathbf{K}$ separately but only on their magnitudes and the angle between them.

The analytic expression of $\mathcal{O}$ was established almost 80 years ago [30] and has been used and rederived many times in the literature. In the case of the absorption of two identical photons, we have to replace $\mathbf{K}$ by $2\boldsymbol{\kappa}$. Then Eq. (23) of [17] becomes

$$\mathcal{O}^{\mathrm{abs-two}}=\frac{N}{8}\mathcal{O}_1\mathcal{O}_2\mathcal{O}_3,\qquad \mathcal{O}_1=\mathbf{K}^2-(1+i\eta)\mathbf{p}\cdot\mathbf{K}, \tag{B2}$$

$$\mathcal{O}_2=[\mathbf{K}^2+(\lambda-ip)^2]^{-1-i\eta},$$
$$\mathcal{O}_3=[(\mathbf{K}-\mathbf{p})^2+\lambda^2]^{-2+i\eta},\qquad \mathbf{K}=2\boldsymbol{\kappa}, \tag{B3}$$

with N in (A7).

For reasons explained further we have redone the analytic calculations of (B1). During the calculation we have considered λ, brought by the ground-state wave function,

$$\langle\mathbf{r}\mid 1s\rangle=\frac{1}{\sqrt{\pi}}\left(\frac{\lambda}{\hbar}\right)^{3/2}e^{-\frac{\lambda}{\hbar}r}, \tag{B4}$$

and η coming from the continuum-state eigenfunction,

$$\langle\mathbf{r}\mid E\mathbf{n}-\rangle=\sqrt{\frac{mp}{(2\pi\hbar)^3}}e^{\pi\eta}\Gamma(1+i\eta)\times e_1^{\frac{i}{\hbar}\mathbf{p}\cdot\mathbf{r}}F_1(-i\eta,1;-\frac{i}{\hbar}(pr+\mathbf{p}\cdot\mathbf{r})), \tag{B5}$$

as *independent parameters*. The difference in our result and that written in (B2) appears only in the factor $\mathcal{O}_1$; namely, we find instead of it

$$\widetilde{\mathcal{O}}_1=\frac{1}{2\lambda}[(\lambda-\eta p)(\lambda-ip)^2+(\lambda+\eta p)K^2-2\eta(p+i\lambda)\mathbf{p}\cdot\mathbf{K}]. \tag{B6}$$

As a matter of fact, if one uses of the relation $\eta=\lambda/p$, $\widetilde{\mathcal{O}}_1$ and $\mathcal{O}_1$ coincide.

The expansion in η of the matrix element $\mathcal{O}$ gives the successive terms of the Born approximation for the continuum wave function. In the exact result (B2) η appears in the factor N, in the exponents of $\mathcal{O}_2$ and $\mathcal{O}_3$, and in the quantity $\mathcal{O}_1$ we are analyzing now.

By setting $\eta=0$ in $\widetilde{\mathcal{O}}_1$ we neglect the Coulomb effect on the ejected electron, getting the contribution of the zeroth-order term in the Born series, i.e., the contribution of the plane wave:

$$\widetilde{\mathcal{O}}_1|_{\eta=0}=\tfrac{1}{2}[(\lambda-ip)^2+K^2]. \tag{B7}$$

Indeed, based on it, with $\eta=0$ in all terms, we get

$$\mathcal{O}\Big|_{\eta=0}=\frac{\sqrt{8\lambda^5mp}}{\pi}\frac{1}{[(\mathbf{p}-\mathbf{K})^2+\lambda^2]^2}, \tag{B8}$$

which is identical to the result obtained directly from (B1) by using the plane wave (adequately normalized) as an approximation for the final-state wave function $\langle\mathbf{r}\mid E\mathbf{n}-\rangle$. At the same time, taking $\eta=0$ in $\mathcal{O}_1$, we get

$$\mathcal{O}_1|_{\eta=0}=K^2-\mathbf{p}\cdot\mathbf{K}. \tag{B9}$$

Obviously, the expressions $\widetilde{\mathcal{O}}_1|_{\eta=0}$ in (B7) and $\mathcal{O}_1|_{\eta=0}$ here do not coincide. The difference comes from the fact that,

using $\eta = \lambda/p$ in Eq. (B6), the first term is 0 and, inside the parentheses of the second one, the term with η has the same contribution as the other term. One concludes that the zeroth- and first-order Born approximations lead to contributions of the same order αZ. The situation described here is similar to the well-known case of the matrix element of $\mathbf{r}$ between initial and final states similar to the ones used here. This is, in fact, due to the particular behavior of the ground-state eigenfunction, which, in momentum space, has the property (for $\lambda \to 0$)

$$u_0(\mathbf{p}) \to C_0 \frac{\pi^2}{\lambda} \delta(\mathbf{p}), \qquad C_0 = \sqrt{\frac{8\lambda^5}{\pi^2}}. \tag{B10}$$

As shown in Fig. 3, the plane-wave approximation leads to an overestimation of the integral (B1), and it is a poor approximation, in agreement with the data displayed at lower energies in Fig. 2 of Varma *et al.* [14] for the $\mathbf{A}^2$ contribution.

When integrating the differential cross section associated to $\mathbf{A}^2$ over the electron direction, one gets, after some algebraic manipulations, the expression of the cross section,

$$\sigma_{\mathbf{A}^2} = \frac{2^{11}\pi^3}{3} \frac{\alpha^2\hbar^5}{mc^2} \lambda^6 \frac{\exp(2\eta\phi)}{1-\exp(-2\pi\eta)} \frac{12\kappa^2 + p^2 + \lambda^2}{(A^2 - B^2)^3}, \tag{B11}$$

where

$$A = 4\kappa^2 + p^2 + \lambda^2, \qquad B = -4\kappa p \tag{B12}$$

and

$$\cos\phi = \frac{4\kappa^2 + \lambda^2 - p^2}{\sqrt{A^2 - B^2}}, \qquad \sin\phi = \frac{-2\lambda p}{\sqrt{A^2 - B^2}}. \tag{B13}$$

If in the previous expressions the variables p and κ are connected by the conservation law (3), the numerator in (B11) becomes

$$A^2 - B^2 = 16\kappa^2[(mc - \kappa)^2 + \lambda^2]. \tag{B14}$$

The total cross section is then

$$\sigma^{\mathrm{NR}}_{\mathbf{A}^2} = \frac{2\pi^3}{3} \frac{\alpha^2\hbar^5}{mc^2} \lambda^6 \frac{\exp(2\eta\phi)}{1-\exp(-2\pi\eta)} \frac{mc + 3\kappa}{\kappa^5[(mc-\kappa)^2 + \lambda^2]^3}. \tag{B15}$$

One sees that the contribution of the $\mathbf{A}^2$ term to the total cross section has another particularity: the denominator in (B15) presents two poles in the complex plane of κ, located at $mc \pm i\lambda$. Its presence brings a fictitious maximum of the quantity $\sigma^{\mathrm{NR}}_{\mathbf{A}^2}$. It was remarked in the cases of photoeffect [21] and Compton [27,28] and Rayleigh scattering [29] that this pole disappears if the relativistic energy conservation law is used. This is also our case: if p and κ are connected by the relativistic relation

$$\sqrt{m^2c^4 + c^2p^2} = E_0 + 2c\kappa, \qquad E_0 = mc^2\sqrt{1-(\alpha Z)^2}, \tag{B16}$$

then we get an expression free of poles,

$$(A^2 - B^2)_{\mathrm{rel}} = 16\kappa^2 m^2 c^2; \tag{B17}$$

the corresponding term in the total cross section is

$$\sigma^{\mathrm{rel}}_{\mathbf{A}^2} = \frac{2\pi^3}{3} \frac{\alpha^2\hbar^5}{mc^2} (\alpha Z)^6 \frac{\exp(2\eta\phi)}{1-\exp(-2\pi\eta)} \frac{\frac{E_0}{c} + 4\kappa}{\kappa^5}. \tag{B18}$$

The differences between the results for $\sigma_{\mathbf{A}^2}$ obtained with the two versions (nonrelativistic and relativistic) of the energy conservation law are discussed in relation to Figs. 3 and 4.

Although it is a very poor approximation, for the sake of completeness, we include the cross section $\sigma^{\mathrm{NR,p-w}}_{\mathbf{A}^2}$ evaluated with a plane wave, i.e., using (B7):

$$\sigma^{\mathrm{NR,p-w}}_{\mathbf{A}^2} = \frac{\pi^2}{12} \frac{\alpha^2\hbar^5}{mc^2} (\alpha Z)^5 \frac{p}{mc} \frac{3m^2c^2 + 10mc\kappa - \lambda^2 + 3\kappa^2}{\kappa^6[(mc-\kappa)^2 + \lambda^2]^3}. \tag{B19}$$

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