Two-photon absorption of few-electron heavy ions

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The two-photon absorption of few-electron ions has been studied by using second-order perturbation theory and Dirac's relativistic equation. Within this framework, the general expressions for the excitation cross sections and rates are derived including a full account of the higher-order multipole terms in the expansion of the electron-photon interaction. While these expressions can be applied to any ion, independent of its particular shell structure, detailed computations are carried out for the two-photon absorption of hydrogen-, helium-, and berylliumlike ions and are compared with the available theoretical and experimental data. The importance of relativistic and nondipole effects in the analysis and computation of induced two-photon transitions is pointed out. Moreover, we discuss the potential of these transitions for atomic parity-violation studies in the high-Z domain.

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I. INTRODUCTION

The recent advances in the setup of coherent-light sources within the optical, ultraviolet (UV), and even x-ray domains have opened up different avenues to explore two-photon transitions between *bound* atomic states. During recent years, for example, a series of experiments has been performed on the $ns \to n's$ and $ns \to n'd$ two-photon excitation of neutral hydrogen and deuterium atoms [1–4]. These measurements revealed information about quantum electrodynamical (QED) effects in simple atomic systems and, moreover, helped to determine fundamental physical constants with unprecedented accuracy [5]. Apart from hydrogen atom and hydrogenic systems, studies on the outer-shell (two-photon) excitation of low- and medium-Z neutral atoms have been reported extensively in the literature [6–10]. These studies were found to be of particular interest not only for improving our understanding of the electronic structure of complex atoms, but also for the diagnostics of laboratory plasmas.

In contrast to neutral hydrogen and rather light atoms, less attention on studying two-photon absorption processes has been paid in the past to medium- and high-Z atoms and ions. With the help of modern sources of coherent-vacuum UV and x-ray radiation, however, such experiments become feasible today. They may provide valuable insight into relativistic, many-body, and QED phenomena in strong electromagnetic fields and, hence, may serve as a complementary technique to the well-established x-ray absorption spectroscopy [11,12]. Moreover, in recent years, two-photon-induced transitions in heavy highly charged ions have attracted much attention as a promising tool for studying atomic parity-violation (PV) effects [13–15]. In order to understand these effects better, currently, a different generation of experiments on the two-photon absorption of heliumlike ions is being planned at the GSI facility in Darmstadt and the Helmholtz Institute in Jena.

Despite the recent interest in the two-photon spectroscopy of heavy ions, not much theoretical work has been performed in this area that might help in developing and analyzing future experiments. In particular, there is a lack of reliable predictions of the *absorption rates* that account for the relativistic and nondipole effects and allow for detailed investigations of the excitation process in the dependence on the directions and polarizations of the incident photons. Although first steps toward such an analysis have been taken for hydrogenlike [11,12,16,17] and few-electron [13,14] heavy ions, most of these previous studies were restricted to a particular geometry of the two-photon absorption experiment as well as to just some particular atom or ion. Instead, in the present paper, we lay out a unified formalism for the two-photon excitation of many-electron systems, independent of their particular initial- and final-state shell structures as well as the properties of the incident light. This analysis is performed within the framework of second-order perturbation theory whose basic expressions are summarized in Sec. II A. In particular, here, we introduce the transition amplitudes that describe a bound-state transition under the simultaneous absorption of two photons. By making use of the multipole expansion of the electron-photon interaction operator and angular momentum algebra, we are able to factorize these amplitudes into reduced second-order matrix elements, which depend on the electronic structure of a particular ion and a (so-called) angular polarization tensor. The latter contains the complete information about the direction and polarization states of the incident photons and is discussed in detail in Sec. II B. Based on the general formula for the second-order amplitude, in Sec. II C, we then derive the two-photon absorption cross sections and transition rates. While these expressions are general and can be employed to analyze the two-photon excitation of any ion or atom, in Sec. III A, we restrict our

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analysis to hydrogenlike ions. For these ions, we employ the well-established finite-basis-set method to calculate the rates for several $ns \to n's$ and $ns \to n'p$ transitions. The results from these computations indicate the importance of a proper account of relativistic and nondipole effects for the accurate evaluation of the transition rates especially for medium- and high-Z ions. Apart from the hydrogenlike systems, we also use our approach to re-investigate the two-photon excitation of helium- and berylliumlike heavy ions. The (excitation) probabilities for the $1s2p^3P_0 \to 1s2s^1S_0$- and $1s^22s^2\,^1S_0 \to 1s^22s2p\,^3P_0$-induced two-photon transitions in these ions, obtained within the multiconfiguration Dirac-Fock (MCDF) approach, are displayed in Sec. III B and are compared with the previous theoretical predictions. Here, special emphasis is placed on the electric dipole $2E1$ excitations, which may proceed only because of a parity mixing of the ionic states. Finally, a brief summary of these results and outlook is given in Sec. IV.

Atomic units ($\hbar = m_e = e = 1$) are used throughout the paper, unless stated otherwise.

II. THEORETICAL BACKGROUND

A. Evaluation of the transition amplitude

Theoretical studies on two-photon transitions in atomic systems have a long tradition that goes back to the seminal papers by Göppert-Mayer [18] and Breit and Teller [19]. In these papers, the analysis of the decay (or excitation) rates has been traced back usually to the second-order transition amplitudes. For the absorption of two photons with the wave vectors $\boldsymbol{k}_i$ and polarizations $\boldsymbol{\epsilon}_i$ ($i = 1,2$), such amplitudes read as

$$\mathcal{M}_{fi}(M_f,M_i) = \sum_{\alpha_\nu J_\nu M_\nu} \frac{\langle \alpha_f J_f M_f|\hat{\mathcal{R}}(\boldsymbol{k}_1,\boldsymbol{\epsilon}_1)|\alpha_\nu J_\nu M_\nu\rangle\langle \alpha_\nu J_\nu M_\nu|\hat{\mathcal{R}}(\boldsymbol{k}_2,\boldsymbol{\epsilon}_2)|\alpha_i J_i M_i\rangle}{E_\nu - E_i - \omega_2} + \sum_{\alpha_\nu J_\nu M_\nu} \frac{\langle \alpha_f J_f M_f|\hat{\mathcal{R}}(\boldsymbol{k}_2,\boldsymbol{\epsilon}_2)|\alpha_\nu J_\nu M_\nu\rangle\langle \alpha_\nu J_\nu M_\nu|\hat{\mathcal{R}}(\boldsymbol{k}_1,\boldsymbol{\epsilon}_1)|\alpha_i J_i M_i\rangle}{E_\nu - E_i - \omega_1}, \tag{1}$$

where $|\alpha_i J_i M_i\rangle$ and $|\alpha_f J_f M_f\rangle$ denote general (many-electron) initial- and final-ionic states with well-defined total angular momenta $J_{i,f}$ and their projections $M_{i,f}$ and where the $\alpha_{i,f}$ refer to all the additional quantum numbers. Moreover, in Eq. (1), $\omega_{1,2} = k_{1,2}/\alpha$ are the energies of the absorbed photons. In the calculations below, we always assume that the incident light has been tuned into resonance with a two-photon absorption and, hence, that $\omega_{1,2}$ are related to the final- and initial-state energies E_i and E_f by the condition

$$E_f - E_i = \omega_1 + \omega_2. \tag{2}$$

Furthermore, in Eq. (1), $\hat{\mathcal{R}}$ is the transition operator that describes the interaction of the electrons with the electromagnetic radiation and that can be written as a sum of one-particle operators,

$$\hat{\mathcal{R}}(\boldsymbol{k},\boldsymbol{\epsilon}) = \sum_m \boldsymbol{A}_m(\boldsymbol{k},\boldsymbol{\epsilon}) = \sum_m [\boldsymbol{\alpha}_m \cdot (\boldsymbol{\epsilon} + G\hat{\boldsymbol{k}})e^{i\boldsymbol{k}\cdot\boldsymbol{r}_m} - G e^{i\boldsymbol{k}\cdot\boldsymbol{r}_m}]. \tag{3}$$

Here, $\boldsymbol{\alpha}_m = (\alpha_{x,m},\alpha_{y,m},\alpha_{z,m})$ denotes the vector of the Dirac matrices for the mth particle, and G is an arbitrary gauge parameter. In the calculations below, we use two different gauges that are known to lead to well-known nonrelativistic operators. The so-called Coulomb gauge is obtained for $G = 0$ and corresponds to the velocity form of electron-photon interaction operator in the nonrelativistic limit. In the Babushkin gauge, in contrast, one adopts $G = \sqrt{(L+1)/L}$, and this simplifies, for the particular case of $L = 1$, to the dipole length form of the transition operator in the nonrelativistic case.

A further simplification of the two-photon absorption amplitude (1) can be achieved with the help of Racah's algebra if use is made of the radial-angular representation of the Dirac wave functions and if the operator $\hat{\mathcal{R}} = \sum_m \boldsymbol{A}_m(\boldsymbol{k},\boldsymbol{\epsilon})$ is decomposed into spherical tensors. For the emission of the photon in the direction $\hat{\boldsymbol{k}} = (\theta,\phi)$ with respect to the z axis, taken as the quantization axis, such a decomposition reads [20,21]

$$\boldsymbol{A}_m(\boldsymbol{k},\boldsymbol{\epsilon}) = 4\pi \sum_{pLM} i^{L-|p|}[\boldsymbol{\epsilon} \cdot \boldsymbol{Y}^{(p)*}_{LM}(\hat{\boldsymbol{k}})]\boldsymbol{a}^p_{LM,m}(k), \tag{4}$$

where $\boldsymbol{Y}^{(p)}_{LM}(\hat{\boldsymbol{k}})$ is a vector spherical harmonics [22] and the index p describes electric ($p = 1$), magnetic ($p = 0$), and longitudinal ($p = -1$) components of the electromagnetic field. The explicit form of these components has been discussed in several places in the literature; cf. Refs. [20,21].

By inserting Eqs. (3) and (4) into the amplitude (1) and employing the Wigner-Eckart theorem, we obtain

$$\mathcal{M}_{fi}(M_f,M_i) = \sum_{\alpha_\nu J_\nu M_\nu} \sum_{p_1 L_1 M_1} \sum_{p_2 L_2 M_2} \left[\boldsymbol{\epsilon}_1 \cdot \boldsymbol{Y}^{(p_1)*}_{L_1 M_1}(\hat{\boldsymbol{k}}_1)\right]\left[\boldsymbol{\epsilon}_2 \cdot \boldsymbol{Y}^{(p_2)*}_{L_2 M_2}(\hat{\boldsymbol{k}}_2)\right] \frac{(4\pi)^2 i^{L_1+L_2-|p_1|-|p_2|}}{[J_f,J_\nu]^{1/2}} [\langle J_i M_i]L_2 M_2 \mid J_\nu M_\nu\rangle$$
$$\times \langle J_\nu M_\nu L_1 M_1 \mid J_f M_f\rangle S^{J_\nu}_{L_1 p_1, L_2 p_2}(\omega_2) + \langle J_i M_i\, L_1 M_1 \mid J_\nu M_\nu\rangle \langle J_\nu M_\nu L_2 M_2 \mid J_f M_f\rangle S^{J_\nu}_{L_2 p_2, L_1 p_1}(\omega_1)], \tag{5}$$

where the notation $[a,b,\ldots] = (2a+1)(2b+1)\cdots$ is used and the reduced (second-order) matrix element is given by

$$S^{J_v}_{L_1 p_1, L_2 p_2}(\omega_2) = \sum_{\alpha_v} \frac{\left\langle \alpha_f J_f \middle\| \sum_m \hat{a}^{p_1}_{L_1,m}(k_1) \middle\| \alpha_v J_v \right\rangle \left\langle \alpha_v J_v \middle\| \sum_m \hat{a}^{p_2}_{L_2,m}(k_2) \middle\| \alpha_i J_i \right\rangle}{E_v - E_i - \omega_2}. \tag{6}$$

In order to perform the summation over the projections M_1, M_2, and M_v of the angular momenta and, hence, to rewrite Eq. (5) in a more compact form, we introduce the irreducible tensor,

$$\begin{aligned} T^{L_1 p_1, L_2 p_2}_{kq} &= \left\{ \left[\boldsymbol{\epsilon}_1 \cdot \boldsymbol{Y}^{(p_1)*}_{L_1}(\boldsymbol{k}_1) \right] \otimes \left[\boldsymbol{\epsilon}_2 \cdot \boldsymbol{Y}^{(p_2)*}_{L_2}(\boldsymbol{k}_2) \right] \right\}_{km_k} \\ &= \sum_{M_1 M_2} \langle L_1 M_1 L_2 M_2 \mid kq \rangle \left[\boldsymbol{\epsilon}_1 \cdot \boldsymbol{Y}^{(p_1)*}_{L_1 M_1}(\hat{\boldsymbol{k}}_1) \right] \\ &\quad \times \left[\boldsymbol{\epsilon}_2 \cdot \boldsymbol{Y}^{(p_2)*}_{L_2 M_2}(\hat{\boldsymbol{k}}_2) \right], \end{aligned} \tag{7}$$

which completely defines the polarization as well as the angular dependence of the two-photon absorption process. With the help of this tensor, the second-order amplitude then simplifies to

$$\begin{aligned} \mathcal{M}_{fi}(M_f, M_i) = \sum_{kq} \sqrt{2k+1} \langle kq\, J_f M_f \mid J_i M_i \rangle \\ \times U_{kq}(\alpha_f J_f, \alpha_i J_i), \end{aligned} \tag{8}$$

and where the function U_{kq} is defined by

$$\begin{aligned} U_{kq}(\alpha_f J_f, \alpha_i J_i) = \frac{(4\pi)^2}{\sqrt{2J_i+1}} \sum_{L_1 p_1} \sum_{L_2 p_2} i^{L_1+L_2-|p_1|-|p_2|} (-1)^{J_f+J_i+p_1+p_2} T^{L_1 p_1, L_2 p_2}_{kq} \\ \times \sum_{J_v} \left(\begin{Bmatrix} L_2 & L_1 & k \\ J_i & J_f & J_v \end{Bmatrix} S^{J_v}_{L_2 p_2, L_1 p_1}(\omega_1) + (-1)^{L_1+L_2+k} \begin{Bmatrix} L_2 & L_1 & k \\ J_f & J_i & J_v \end{Bmatrix} S^{J_v}_{L_1 p_1, L_2 p_2}(\omega_2) \right). \end{aligned} \tag{9}$$

It follows immediately from the properties of Wigner's $6j$ symbols that the U_{kq} are *nonzero* only if k satisfies the condition $|J_i - J_f| \leqslant k \leqslant J_i + J_f$. Therefore, the characteristics of the two-photon $J_i = 0 \to J_f = 0$ transitions are described by a single U_{00} function, while four components with $k = 0,1$ and $q = -k,\ldots,k$ are required, in general, to analyze the two-photon absorption or emission for some $J_i = 1/2 \to J_f = 1/2$ transition.

B. Properties of the angular-polarization tensor

As seen from formulas (8) and (9), the computation of the amplitude $\mathcal{M}_{fi}(M_f, M_i)$ requires some knowledge about the angular polarization tensor (7) as well as the reduced matrix elements (6). While these matrix elements will be discussed later in Sec. III, here we will briefly recall the basic properties of the T_{kq}. The most general form of this tensor has been derived by Manakov and co-workers in Refs. [21,23]. In the present section, therefore, we do not repeat those (rather elaborate) evaluations but restrict ourselves to the particular case of the linearly polarized photons whose polarization vectors $\boldsymbol{\epsilon}_{1,2}$ are described by the tilt angles χ_1 and χ_2 with respect to the reaction plane, which is spanned by the momenta $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$, respectively. For such a photon pair, the angular polarization tensor reads

$$\begin{aligned} T^{L_1 p_1, L_2 p_2}_{kq} = \frac{[L_1, L_2]^{1/2}}{16\pi} \\ \times \sum_{M_1 M_2} \sum_{\lambda_1, \lambda_2 = -1,1} e^{-i(\lambda_1 \chi_1 + \lambda_2 \chi_2)} (-\lambda_1)^{p_1} (-\lambda_2)^{p_2} \\ \times \langle L_1 M_1 L_2 M_2 \mid kq \rangle\, D^{L_1}_{M_1 \lambda_1}(\hat{k}_1) D^{L_2}_{M_2 \lambda_2}(\hat{k}_2), \end{aligned} \tag{10}$$

with D being the Wigner rotation matrix. This expression can be simplified further by a proper choice of the quantization axis. Since there was no direction initially *preferred* for the excitation of unpolarized (as well as unaligned) ions or atoms, we adopted $\hat{z}$ as the quantization axis along the momentum of the first photon, i.e., $\hat{k}_1 || \hat{z}$. Therefore, a single *opening* angle θ is only required to characterize the emission of γ quanta with respect to each other,

$$\begin{aligned} T^{L_1 p_1, L_2 p_2}_{kq}(\theta) = \frac{[L_1, L_2]^{1/2}}{16\pi} \\ \times \sum_{\lambda_1, \lambda_2 = -1,1} e^{-i(\lambda_1 \chi_1 + \lambda_2 \chi_2)} (-\lambda_1)^{p_1} (-\lambda_2)^{p_2} \\ \times \langle L_1 \lambda_1 L_2 M_2 \mid kq \rangle\, D^{L_2}_{M_2 \lambda_2}(0, \theta, 0), \end{aligned} \tag{11}$$

where $M_2 = q - \lambda_1$, as it follows immediately from the properties of the Clebsch-Gordan coefficients.

With the help of Eq. (11), one can evaluate the tensor T_{kq} for any combination $(L_1 p_1, L_2 p_2)$ of multipoles that is allowed for a particular $|\alpha_i J_i\rangle + \gamma_1 + \gamma_2 \to |\alpha_f J_f\rangle$ transition. Of special interest, however, are the angular and polarization properties of the *leading* term(s) in the multipole expansion, which dominate(s) the two-photon absorption for any given pair of initial- and final-bound states. For example, if we assume a transition to be induced between two $J = 0$ states of equal parity, it will most likely occur due to the absorption of two electric dipole ($2E1$) photons. The characteristics of such a transition are described by a single zero-rank tensor

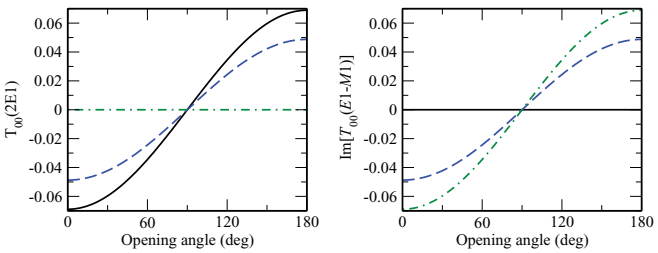

FIG. 1. (Color online) The zero-rank angular polarization tensor T_{00} for the $2E1$ transition (left panel) and the $E1$-$M1$ transition (right panel) as a function of the opening angle θ. The polarization vector of the first photon lies within the reaction plane $\chi_1 = 0$, while the second photon is polarized under the angle $\chi_2 = 0$ (solid line), $\pi/4$ (dashed line), and $\pi/2$ (dashed–dotted line) with respect to this plane.

T_{00}, which is given by

$$T_{00}^{L_1=1,\ p_1=1;\ L_2=1,\ p_2=1}(\theta) = -\frac{\sqrt{3}}{8\pi}(\sin\chi_1 \sin\chi_2 + \cos\chi_1 \cos\chi_2 \cos\theta) = -\frac{\sqrt{3}}{8\pi}(\boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_2). \quad (12)$$

This expression includes the well-known result that any $n\,^1S_0 \to n'\,^1S_0$ transition in heliumlike and $ns \to n's$ transitions in (nonrelativistic low-Z) hydrogenlike ions is forbidden if the polarization vectors of the two photons are orthogonal to each other (see also the right panel of Fig. 1). If, in contrast, we consider a $J_i = 0 \to J_f = 0$ excitation for ionic states of different parity, the tensor T_{00} reads as

$$T_{00}^{L_1=1,\ p_1=1;\ L_2=1,\ p_2=0}(\theta) = \frac{i\sqrt{3}}{8\pi}(\sin\chi_1 \cos\chi_2 - \cos\chi_1 \sin\chi_2 \cos\theta) = -\frac{i\sqrt{3}}{8\pi}\hat{\boldsymbol{k}}_1 \cdot (\boldsymbol{\epsilon}_1 \times \boldsymbol{\epsilon}_2) \quad (13)$$

for the electric dipole magnetic dipole ($E1$-$M1$) channel. Apart from this channel, of course, one has to also account for the $M1$-$E1$ term that corresponds to the combination ($L_1 = 1,\ p_1 = 0;\ L_2 = 1,\ p_2 = 1$). Together, these two pathways give rise to the two-photon amplitude, which is proportional to $(\hat{\boldsymbol{k}}_1 - \hat{\boldsymbol{k}}_2) \cdot (\boldsymbol{\epsilon}_1 \times \boldsymbol{\epsilon}_2)$ and which immediately indicates that such $E1$-$M1$–$M1$-$E1$ transitions *cannot* be induced by a single coherent laser beam [13].

C. Cross sections and transition rates

Equations (8) and (9) display the general form of the second-order matrix element that describes the excitation of the ion (or atom) under the simultaneous absorption of two photons. This matrix element represents the central building block from which all the (two-photon) properties can be calculated. For example, experimental results on the two-photon absorption are often presented in terms of the parameter α_0 that can be calculated as

$$\alpha_0 = \frac{(2\pi)^3}{4\alpha^2\omega^3}\frac{1}{2J_i+1}\sum_{M_i M_f}|\mathcal{M}_{fi}(M_f, M_i)|^2 = \frac{(2\pi)^3}{4\alpha^2\omega^3}\sum_{kq}|U_{kq}(\alpha_f J_f, \alpha_i J_i)|^2. \quad (14)$$

This parameter, which—in *Système International* units—is expressed in $(\mathrm{cm}^2/\mathrm{s})/(\mathrm{W}/\mathrm{cm}^2)$, is directly related to the effective excitation cross section (given in cm^4/W),

$$\sigma = \alpha_0 g(\omega) G^{(2)}, \quad (15)$$

and to the two-photon rate (in s^{-1}):

$$W = \frac{I^2}{\omega}\alpha_0 g(\omega) G^{(2)}. \quad (16)$$

In these expressions, I is the intensity of the incident light, $G^{(2)}$ is the two-photon statistical factor, which takes the value $G^{(2)} = 1$ for a single-mode laser and $G^{(2)} = 2$ for an incoherent source [24,25], and $g(\omega)$ is the line-shape function. By choosing the resonance condition (2) and the Gaussian profile of the laser beam, we can write this function as

$$g(\omega) = \left(\frac{4\ln(2)}{\pi}\right)^{1/2}\frac{1}{\left(\Delta\omega_D^2 + 2\Delta\omega_L^2\right)^{1/2}}, \quad (17)$$

where $\Delta\omega_D$ and $\Delta\omega_L$ are the Doppler width and laser linewidth, respectively. Here, both these widths are assumed to be larger than the radiative (natural) width of the excited ionic state. This assumption might not always be true for the two-photon absorption of high-Z ions. For such ions, one should use the generalization of Eq. (17) as given, for example, in Ref. [26].

III. RESULTS AND DISCUSSIONS

A. Two-photon absorption of hydrogenlike ions

The expressions for the second-order amplitudes (8) and (9) and transition rates (14)–(16), derived in the previous section, can be utilized for any shell structure of the atoms and ions, if the corresponding (reduced) elements S^{J_ν} are properly generated. Within the relativistic framework, however, the evaluation of the two-photon matrix elements is generally not a simple task since it requires a summation over the complete Dirac spectrum [cf. Eq. (6)]. A number of methods has been developed in the past to perform such an intermediate-state summation consistently. For the hydrogenlike ions, the matrix elements S^{J_ν} can be calculated very accurately by making use of either the relativistic Coulomb-Green's function approach [27–29] or the various finite-basis-set methods [30–35]. In the present paper, we followed the second line and employed the finite-basis solutions constructed from a B-spline set. Based on this approach, the second-order matrix elements and, in turn, the parameter α_0 were evaluated for the two-photon excitation of hydrogenlike systems for a single linearly polarized laser. For this particular case, the results are obtained immediately from Eqs. (8)–(10) by choosing $\omega_1 = \omega_2 = (E_f - E_i)/2$, $\hat{k}_1 = \hat{k}_2$, and $\chi_1 = \chi_2$.

TABLE I. The parameter (14) for the two-photon absorption of neutral hydrogen. Predictions have been obtained for two gauges for the coupling of the radiation field within the electric dipole ($2E1$) approximation as well as by taking the higher (nondipole) terms into account. Our fully relativistic predictions are compared with the results of the nonrelativistic dipole calculations by Tung *et al.* [38]. Powers of 10 are given in brackets.

		α_0 (cm^2/s)/(W/cm^2)	
Transition	Multipolarity	Coulomb	Babushkin
$1s_{1/2} \rightarrow 2s_{1/2}$	$2E1$	2.747 952[−17]	2.747 952[−17]
			2.75[−17][a]
	Total	2.747 935[−17]	2.747 935[−17]
$1s_{1/2} \rightarrow 3s_{1/2}$	$2E1$	2.419 411[−18]	2.419 411[−18]
			2.42[−18][a]
	Total	2.419 404[−18]	2.419 404[−18]
$1s_{1/2} \rightarrow 4s_{1/2}$	$2E1$	6.687 125[−19]	6.687 125[−19]
			6.69[−19][a]
	Total	6.687 113[−19]	6.687 113[−19]
$2s_{1/2} \rightarrow 3s_{1/2}$	$2E1$	5.709 419[−15]	5.709 419[−15]
			5.72[−17][a]
	Total	5.709 405[−15]	5.709 405[−15]
$2s_{1/2} \rightarrow 4s_{1/2}$	$2E1$	3.079 139[−17]	3.079 139[−17]
			3.52[−17][a]
	Total	3.079 124[−17]	3.079 124[−17]

[a]Tung *et al.* [38].

Tables I, II, and III display parameter α_0 for various $ns \rightarrow n's$ transitions in neutral hydrogen as well as hydrogenlike xenon Xe^{53+} and uranium U^{91+} ions. The calculations have been performed in Coulomb (velocity) as well as Babushkin (length) gauges for the coupling of the radiation field. Although an agreement between these two gauges does not prove the correctness of the predicted data *directly*, it is usually used as an indicator for the accuracy of the atomic-structure calculations [36,37]. Moreover, predictions of the exact relativistic theory that account for all allowed multipole components (p_1L_1, p_2L_2) in the amplitude (9) are compared with those from the electric dipole approximation, i.e., if the multipole summation is restricted to the single term with $L_1 = L_2 = 1$ and $p_1 = p_2 = 1$. As expected, both approaches yield almost identical results for neutral hydrogen, for which the relativistic and nondipole effects are known to be negligible. For $Z = 1$, moreover, our calculations reproduce the nonrelativistic dipole calculations by Tung *et al.* [38] as well as by Gontier and Trahin [39] very well, and they also show good agreement with the experimental finding for the $1s \rightarrow 2s$ effective excitation cross section σ from Ref. [40]. In that paper, a value of $\sigma^{\rm exp} = 3.3 \pm 0.8 \times 10^{-28}$ cm^4/W was determined, which can be compared directly to our prediction of $\sigma^{\rm th} = 5.2 \pm 1.3 \times 10^{-28}$ cm^4/W and where the latter value has been obtained from Eq. (15) for the two-photon statistical factor $G^{(2)} = 2$ and the line-shape function $g(\omega) = 9.6 \pm 2.5 \times 10^{12}$ s (as recommended in Ref. [40]). One may note that, despite the numerical accuracy of the α_0 parameters in Table I, the theoretical value $\sigma^{\rm th}(1s \rightarrow 2s)$ has an uncertainty of about 25%. As seen from Eq. (17), for the line-shape function, this uncertainty accounts for the incomplete knowledge about the properties of the laser beam as well as of the ensemble of target

TABLE II. The same as Table I but for the two-photon excitation of hydrogenlike xenon Xe^{53+}.

		α_0 (cm^2/s)/(W/cm^2)	
Transition	Multipolarity	Coulomb	Babushkin
$1s_{1/2} \rightarrow 2s_{1/2}$	$2E1$	8.495 884[−28]	8.495 884[−28]
	Total	8.338 306[−28]	8.338 306[−28]
$1s_{1/2} \rightarrow 3s_{1/2}$	$2E1$	6.421 630[−29]	6.421 630[−29]
	Total	6.369 145[−29]	6.369 145[−29]
$1s_{1/2} \rightarrow 4s_{1/2}$	$2E1$	1.650 110[−29]	1.650 117[−29]
	Total	1.643 400[−29]	1.643 405[−29]
$2s_{1/2} \rightarrow 3s_{1/2}$	$2E1$	1.311 563[−25]	1.311 563[−25]
	Total	1.299 959[−25]	1.299 959[−25]
$2s_{1/2} \rightarrow 4s_{1/2}$	$2E1$	1.471 467[−28]	1.471 465[−28]
	Total	1.529 490[−28]	1.529 488[−28]

TABLE III. The same as Table I but for the two-photon excitation of hydrogenlike uranium U^{91+}.

Transition	Multipolarity	α_0 (cm^2/s)/(W/cm^2)	
		Coulomb	Babushkin
$1s_{1/2} \to 2s_{1/2}$	$2E1$	1.849 824[−29]	1.849 824[−29]
	Total	1.743 653[−29]	1.743 653[−29]
$1s_{1/2} \to 3s_{1/2}$	$2E1$	9.063 289[−31]	9.063 420[−31]
	Total	8.937 087[−31]	8.937 237[−31]
$1s_{1/2} \to 4s_{1/2}$	$2E1$	1.856 756[−31]	1.856 662[−31]
	Total	1.868 169[−31]	1.867 428[−31]
$2s_{1/2} \to 3s_{1/2}$	$2E1$	5.398 854[−28]	5.398 855[−28]
	Total	5.053 379[−28]	5.053 380[−28]
$2s_{1/2} \to 4s_{1/2}$	$2E1$	2.422 525[−29]	2.422 506[−29]
	Total	2.306 343[−29]	2.306 325[−29]

atoms in a *particular* experiment. For example, in the paper reported by Bickel and McRae in Ref. [40], a 243-nm XeCl dye laser was used for which the linewidth $\Delta\omega_L = 18.8$ GHz was known only within about 20% uncertainty. An almost equally large uncertainty was found for the Doppler width $\Delta\omega_D$ and was attributed to the wide velocity spread of the (target) hydrogen atoms, which were produced by the laser ablation of the zirconium alloys. Any improvement in the accuracy of the two-photon absorption studies, therefore, will require a better control of the temporal and spatial laser-beam characteristics and of the target properties. This can be achieved, for example, by using cold-atomic targets together with the single-frequency high-power lasers (see, e.g., Ref. [41]) or narrow cw single-mode lasers as well as by performing Doppler-free spectroscopy [2,42–46].

As seen from Tables II and III, we observe a significant reduction in parameter α_0 as the nuclear charge Z increases. This Z behavior can easily be understood—at least, within the framework of the nonrelativistic dipole approximation—if we consider the individual trends of the matrix elements and energy denominator in expression (14): While the two-photon matrix element (8) is weakly dependent on Z [11,12,47], the transition energy $2\omega = E_f - E_i$ follows a Z^2 law. That yields a simple estimate $\alpha_0 \sim Z^{-6}$ which, however, takes neither the relativistic contraction of the wave functions toward the nucleus nor the nondipole effects in the electron-photon interaction into account. The relativistic contraction is known to further reduce the two-photon cross sections by a factor of about 2 for high-Z ions (see, for example, Refs. [11,48]). In contrast, the influence of the higher-multipole contributions is not so significant. As seen from Tables II and III, for example, the probability of the $1s \to 2s$ two-photon absorption is decreased only by about 2% for Xe^{53+} and by 6% for U^{91+} if, apart from the leading $2E1$ channel, the nondipole terms are taken into account. This reduction, which was also predicted for the two-photon *decay* of the $2s$ metastable state [27,49], arises mainly from the interference between the $2E1$, $2M1$, and $2E2$ multipole components as can be deduced from Eq. (14),

$$\alpha_0(1s \to ns) \simeq \frac{8\pi^5}{\alpha^2\omega^3} |\mathcal{S}_{E1}|^2 \left(1 + 2\frac{\mathcal{S}_{M1}}{\mathcal{S}_{E1}} + 2\frac{\mathcal{S}_{E2}}{\mathcal{S}_{E1}} + \cdots\right), \tag{18}$$

and where we have assumed again that both incident photons have the same polarization and wave vectors. Moreover, for the sake of brevity, here, we introduced the notation $\mathcal{S}_{Lp} = S^{J'_\nu}_{Lp,\,Lp}(\omega) - S^{J'_\nu+1/2}_{Lp,\,Lp}(\omega)$ where $J'_\nu = 1/2$ for $2E1$ and $2M1$ transitions and $J'_\nu = 3/2$ in the $2E2$ case. As seen from Eq. (18), no photon pair with different multiplicities (for example, $E1$-$M2$) contribute to the $ns \to ns'$ (two-photon) absorption process. Also, this immediately follows from the properties of the angular polarization tensor (7) which—in the absorption of photons from a single polarized laser beam—does not allow any photon pair with multipole combination $L_1 \neq L_2$.

Up to the present, we have discussed the $ns \to ns'$ two-photon excitations of hydrogenlike ions. Besides this case, which has been reasonably established in the literature, we also consider the $ns \to n'p$ induced transitions. Although these transitions are rather weak, they can provide a very promising playground for studying PV phenomena in simple atomic systems. For the two-photon excitation to the p states, moreover, one expects that nondipole terms in the electron-photon interaction play a more significant role than for the $ns \to ns'$ excitations. This is due to the fact that s and p states can be coupled by two $E1$ photons only owing to the tiny PV term and, hence, the $ns \to n'p$ transitions proceed mainly via the leading $E1$-$M1$–$M1$-$E1$ and $E1$-$E2$–$E2$-$E1$ channels. In order to investigate the role of these (and higher-order) multipole terms, in Table IV, we present the α_0 parameter for the excitation of neutral hydrogen as well as hydrogenlike xenon and uranium ions by a single linearly polarized laser (i.e., $\hat{\boldsymbol{k}}_1 = \hat{\boldsymbol{k}}_2$, $\boldsymbol{\epsilon}_1 = \boldsymbol{\epsilon}_2$). The angular polarization tensor T_{00} identically vanishes for such a beam according to Eq. (13), and the properties of the $ns \to n'p_{1/2}$ two-photon transitions are defined by just the tensor component T_{10}. As we see later in Sec. III B, this is not the case for the $S_0 \to P_0$ transitions in helium- and berylliumlike ions for which rank k cannot be greater than zero [cf. Eq. (9)] and, hence, $\alpha_0(\hat{\boldsymbol{k}}_1 = \hat{\boldsymbol{k}}_2, \boldsymbol{\epsilon}_1 = \boldsymbol{\epsilon}_2) = 0$.

Calculations for the two-photon excitation to the $np_{1/2}$ levels have been performed within both, the rigorous relativistic theory (9), where the summation runs over all allowed multipole combinations, and the $E1$-$M1 + E1$-$E2$ approximation, which only includes the leading channels.

TABLE IV. The parameter (14) for the $ns \to n'p$ two-photon transitions in neutral hydrogen as well as hydrogenlike xenon and uranium ions. Predictions have been obtained within the leading $E1$-$M1 + E1$-$E2$ approximation as well as by taking the higher multipole terms using the Babushkin gauge into account. Powers of 10 are given in brackets.

		α_0 (cm^2/s)/(W/cm^2)		
Transition	Multipolarity	$Z = 1$	$Z = 54$	$Z = 92$
$1s_{1/2} \to 2p_{1/2}$	$E1$-$M1$–$E1$-$E2$	1.277 404[−22]	1.028 584[−29]	4.753 682[−31]
	Total	1.277 407[−22]	1.035 005[−29]	4.857 008[−31]
$1s_{1/2} \to 3p_{1/2}$	$E1$-$M1$–$E1$-$E2$	1.406 394[−24]	5.340 185[−32]	5.354 621[−35]
	Total	1.406 396[−24]	5.341 826[−32]	7.417 931[−35]
$1s_{1/2} \to 4p_{1/2}$	$E1$-$M1$–$E1$-$E2$	3.289 574[−26]	1.622 427[−34]	1.171 781[−33]
	Total	3.289 564[−26]	1.814 492[−34]	1.271 929[−33]
$2s_{1/2} \to 3p_{1/2}$	$E1$-$M1$–$E1$-$E2$	7.462 732[−21]	6.096 173[−28]	2.620 305[−29]
	Total	7.462 732[−21]	6.092 971[−28]	2.596 358[−29]
$2s_{1/2} \to 4p_{1/2}$	$E1$-$M1$–$E1$-$E2$	1.728 988[−22]	1.424 077[−29]	5.300 456[−31]
	Total	1.728 992[−22]	1.435 945[−29]	5.549 392[−31]

As seen from Table IV, this approximation provides a good estimate of the parameter α_0 along the entire isoelectronic sequence. That is, the discrepancy between the $E1$-$M1 + E1$-$E2$ calculations and rigorous results does not exceed 5%–7% for the heaviest species. The leading role of the $E1$-$M1$ and $E1$-$E2$ excitation channels also allows us to qualitatively understand the Z scaling of the α_0 parameter for the $ns \to n'p$ transitions. The matrix elements for these two channels scale approximately as Z, thus, leading to an $\alpha_0 \sim Z^{-4}$ dependence. In the relativistic case, again, this dependence has to be corrected for the relativistic contraction and higher-term effects.

B. Intrashell transitions in few-electron ions

Apart from analyzing the transitions in hydrogenlike ions, Eqs. (8)–(16) can be employed to investigate the two-photon absorption of many-electron atoms and ions. Similar to before, the reduced matrix elements (6) contain complete information about the electronic structure of the particular system. In contrast to hydrogenlike ions, however, the relativistic second-order calculations of S^{J_ν} are more intricate for few-electron ions since we then have to account for the interelectronic interaction effects. For the radiative transitions between the ground and the excited states of high-Z ions, whose (excitation) energies usually reach thousands of eV, the independent particle model was found suitable to treat these effects and to compute the total and differential rates [50–52]. During the past decades, however, laser-induced $J_i = 0 + 2\gamma \to J_f = 0$ *intrashell* transitions have attracted much attention, both in theory and in experiment. Having transition energies in the optical-to-extreme-UV range, these induced two-photon excitations may provide an alternative and very promising tool for studying many-body and PV phenomena in heavy atomic systems [13–15].

For the theoretical analysis of such two-photon absorption processes, however, a more systematic treatment of the electron-electron interaction and QED effects is needed, which cannot be performed within the independent particle model [53]. In the present paper, for example, we make use of the MCDF approach [36,54] in order to generate the ionic spectrum and to perform the intermediate-state summation in Eq. (6). Since the excitation energies of the intrashell transitions are orders of magnitude smaller than the binding energies of the active electron in the (initial and final) levels, such a summation can be restricted to just a few states that contribute most to the absorption rates. By applying such a direct summation method, we have re-investigated, in particular, the $1s^2 2s^2\,{}^1S_0 \to 1s^2 2s 2p^3\,P_0$ transition in berylliumlike ions. The two-photon absorption rates for these ions, reported by Maul *et al.* [14], have to be questioned since wrong energies were employed at that time in the electron-photon interaction operators $\hat{\mathcal{R}}(\boldsymbol{k},\boldsymbol{\epsilon})$ that enter the amplitudes (6). Instead of $\omega_1 = \omega_2 = (E_f - E_i)/2$, the authors of Ref. [14] have mistakenly used the energies of transitions between the initial and the intermediate $\omega_1 = E_\nu - E_i$ as well as the intermediate and final states $\omega_2 = E_f - E_\nu$. As we see below, this results in a misestimation of the excitation probabilities.

In Table V, we display the α_0 parameter for the leading $E1$-$M1$–$M1$-$E1$ $1s^2 2s^2\,{}^1S_0 \to 1s^2 2s 2p^3\,P_0$ excitation channel in berylliumlike iron Fe^{22+}, barium Ba^{52+}, and uranium U^{88+} ions. Since, according to Eq. (13), such a transition cannot be induced by a single (coherent) beam, calculations have been performed for two counterpropagating photons, having equal energies $\omega = (E_f - E_i)/2$ and orthogonal polarization vectors $(\boldsymbol{\epsilon}_1 \times \boldsymbol{\epsilon}_2) = 1$. For this two-photon configuration, moreover, the competing $E1$-$E2$–$E2$-$E1$ channel is forbidden owing to the selection rules. As before, predictions have been obtained within two (Coulomb and Babushkin) gauges for the coupling of the radiation field. As seen from the table, the agreement between the gauges is worse than what was obtained for the hydrogenlike ions (cf. Tables I, II, and III). Although our calculations show a very rapid convergence of the summation over the $1s^2 2snl_j$ intermediate states in both gauges, the convergence limits differ by about 30% for medium-Z and 15% for high-Z ions. We attribute these differences to the fact that the summation over the intermediate states in Eq. (6) has been restricted to the *bound* spectrum of

TABLE V. The parameter (14) for the $1s^2 2s^2\,{}^1S_0 \to 1s^2 2s2p\,{}^3P_0$ two-photon transition in berylliumlike iron, barium, and uranium ions. Calculations have been performed within the leading $E1$-$M1$–$M1$-$E1$ approximation and by making use of the direct summation approach. Within this approach, the intermediate-state summation in Eq. (6) is restricted to all (allowed) levels $1s\,nl_j\,{}^{2S+1}L_{J=1}$ with $n \leqslant N_{\max}$. Powers of 10 are given in brackets.

		α_0 (cm^2/s)/(W/cm^2)	
Nuclear charge	$N_{\max}$	Coulomb	Babushkin
$Z = 26$	2	1.989[−27]	2.924[−27]
	3	2.113[−27]	2.927[−27]
$Z = 56$	2	1.494[−27]	2.383[−27]
	3	1.638[−27]	2.384[−27]
	4	1.668[−27]	2.384[−27]
$Z = 92$	2	3.832[−28]	5.458[−28]
	3	4.399[−28]	5.462[−28]
	4	4.521[−28]	5.463[−28]

the ion. As known from previous theoretical studies on the two-photon transitions of one-electron systems [34,55,56], the *continuum* part of the spectrum also contributes to the absorption (and emission) rates, although its contribution is usually less important in the length (Babushkin) gauge. Therefore, our discussion below is based on the Babushkin-gauge results whose accuracy we (conservatively) estimate at about ±20%. We note that such an accuracy is definitely sufficient for the analysis of the forthcoming two-photon absorption experiments that will be carried out at the GSI facility in Darmstadt.

Having calculated the α_0 parameters, now, we are ready to estimate the two-photon rates for the induced $1s^2 2s^2\,{}^1S_0 \to 1s^2 2s2p\,{}^3P_0$ transition. As seen from Eqs. (16) and (17), these calculations require some knowledge about the laser parameters. By choosing a spatially and temporally incoherent UV radiation with an energy width of $\Gamma_{\rm laser} = 1$ eV as suggested in Ref. [14], we obtain the reduced rates $W/I^2 = 1.06 \times 10^{-24}$, 3.55×10^{-25}, and 3.46×10^{-26} cm^4/(W^2 s) for iron, barium, and uranium ions, respectively, which are about 1 order of magnitude larger than those predicted by Maul *et al.* As already mentioned above, this is due to the fact that wrong energies were used in Ref. [14] for the evaluation of the second-order transition amplitudes S^{J_ν}. A further enhancement of the (theoretically predicted) rates W/I^2 can be achieved by using the energy widths $\Gamma_{\rm laser}$ in the meV range. This range is easily accessible at present-day optical- and UV-light facilities.

Until now, our calculations were based on the usual assumption that the parity is well defined for each state of an atom or ions. This conservation of parity places certain restrictions on the choice of allowed multipole components $(L_1 p_1, L_2 p_2)$ in the summation in Eq. (9). These restrictions, however, do not apply if the parity conservation is *violated* due to some mechanism as, for instance, mediated by the weak interaction between the nucleons and the electrons of the ion. For berylliumlike ions, such a PV mechanism leads to a mixing between the ${}^1S_0$ and ${}^3P_0$ berylliumlike states and allows the $1s^2 2s^2\,{}^1S_0 \to 1s^2 2s2p\,{}^3P_0$ two-photon transition to proceed via the electric dipole ($2E1$) channel if both photons are absorbed from a single (coherent) laser beam. In order to estimate the rate of such an excitation process, again, we may use Eqs. (6)–(9) and (14)–(16) but may now consider the initial- and final-ionic states,

$$|1s^2 2s^2\,{}^1S_0\rangle_{\rm PV} \approx |1s^2 2s^2\,{}^1S_0\rangle + \eta_{\rm PV}|1s^2 2s2p\,{}^3P_0\rangle,$$

$$|1s^2 2s2p\,{}^3P_0\rangle_{\rm PV} \approx |1s^2 2s2p\,{}^3P_0\rangle + \eta_{\rm PV}|1s^2 2s^2\,{}^1S_0\rangle, \quad (19)$$

where $\eta_{\rm PV}$ is the parity-mixing coefficient. For berylliumlike ions, this coefficient takes the values $\eta_{\rm PV} = -9.929 \times 10^{-12}$, -6.029×10^{-10}, and -1.916×10^{-8} for $Z = 26, 56$, and 92, respectively (cf. Table I in Ref. [14]). By using these values and the energy width of the laser of 1 eV, we obtain the reduced rates for the PV $2E1$ $1s^2 2s^2\,{}^1S_0 \to 1s^2 2s2p\,{}^3P_0$ transition: $W/I^2 = 2.79 \times 10^{-42}$, 3.48×10^{-41}, and 5.21×10^{-40} cm^4/(W^2 s). Apart from the low-Z end of the isoelectronic sequence, these predictions are about ten times smaller than those reported by Maul *et al.* [14] but can be increased further if more realistic laser parameters are applied.

Besides the two-photon excitation from the ground state of the berylliumlike species, the $1s2p\,{}^3P_0 \to 1s2s\,{}^1S_0$-induced transition in heliumlike heavy ions has attracted some recent interest as an alternative for probing atomic PV [13]. Similar as above, this transition should proceed via the PV electric dipole ($2E1$) channel if induced by two equivalent photons. In the past, a more detailed analysis of this excitation process was hampered by a lack of reliable data on the energies of the low-lying ionic levels. Only recently, systematic investigations of the (heliumlike) spectra and transition energies have been carried out within the QED framework [57–60]. Special attention has been paid to the heliumlike uranium ^{238}U, which is considered to be the most suitable candidate for atomic PV studies in the high-Z domain. For this ion, the ${}^3P_0$-${}^1S_0$ energy splitting was found to be as small as $\Delta E \simeq -2.8$ eV, thus, implying a very strong PV mixing of about $\eta_{\rm PV} = 1.7 \times 10^{-6}$. For such a splitting, moreover, the $1s2p\,{}^3P_0 \to 1s2s\,{}^1S_0$ ($2E1$) transition can be induced by an *optical* light. For this transition, if we assume a mixing between the $|1s2s\,{}^1S_0\rangle$ and $|1s2p\,{}^3P_0\rangle$ states, analog to Eq. (19), we find a reduced rate $W/I^2 = 1.26 \times 10^{-33}$ cm^4/(W^2 s), which agrees with the previous prediction $W/I^2 \approx 10^{-33}$ cm^4/(W^2 s) by Schäfer *et al.* [13]. This value is of direct interest, first, for preparing and analyzing heavy-ion PV experiments that hopefully are to be carried out at the GSI storage ring in Darmstadt and the Helmholtz Institute in Jena. In such experiments, (a beam of) singly excited heliumlike (U^{90+} ions, produced by ion-atom collisions [61], will be irradiated with optical light. As mentioned above, a polarized optical laser will *stimulate* the $2E1$ PV transition between the $2\,{}^3P_0$ and $2\,{}^1S_0$ levels and, hence, will change the lifetime of the metastable ions (owing to the fact that lifetimes of the triplet-P and the singlet-S states differ by a factor of about 400). In order to perform such lifetime measurements in the high-Z domain, however, one needs to apply lasers that are intense enough so that the induced $2E1$ transition can compete with the spontaneous decay of metastable ionic states. Because of this requirement and making use of our theoretical (stimulated)

two-photon absorption rate, one can estimate the necessary laser intensity $I \sim 10^{20}$–10^{21} W/cm^2. Today, these intensities are accessible with modern laser sources (see, for example, Ref. [62]). Apart from the determination of the required laser intensity, an accurate theoretical prediction for this rate will also be employed to extract the information on the mixing parameter $\eta_{\rm PV}$ from the measured lifetimes (as a function of the laser intensity). A more detailed proposal and analysis of such a PV experiment with highly charged heliumlike ions will be presented elsewhere [63]; here, let us just mention that the estimated 20% accuracy for parameter α_0 appears to be sufficient for a first generation of such PV experiments owing to the limitations in controlling the properties of the ion beam, the target for the excitation of the ions, and the overlap of the beams with the profile of the (high-intensity) optical laser.

IV. SUMMARY AND OUTLOOK

Second-order perturbation theory, based on Dirac's relativistic equation, has been applied to investigate the excitation of few-electron ions under the simultaneous absorption of two photons. In particular, we have derived the general expressions for the two-photon transition rates, which include a full account of the higher-order multipole effects in the electron-photon interaction. These expressions can be applied to all many-electron ions, independent of their particular shell structure, if they are exposed to two incident light beams with arbitrary propagation directions and polarization properties. In order to illustrate the application of the approach, detailed calculations have been carried out for the $ns \to n's$ and $ns \to n'p$ two-photon-induced transitions in neutral hydrogen as well as hydrogenlike xenon Xe^{53+} and uranium U^{91+} ions. These calculations, which made use of the finite-basis-set approach, demonstrated the importance of the relativistic contraction of the wave functions as well as the nondipole excitation channels for the accurate analysis of the absorption rates.

Apart from the excitation of single-electron systems, we have also revisited the two-photon intrashell transitions in helium- and berylliumlike ions. During the last two decades, these transitions have attracted a good deal of interest since they may provide an alternative and very promising route for studying PVs in medium- and high-Z ions. Reliable theoretical estimates for the (two-photon) absorption rates are required to plan and to prepare future PV experiments. In this contribution, we employed the MCDF approach in order to compute the probabilities for the parity-allowed $E1$-$M1$–$M1$-$E1$ as well as the PV $2E1$ $1s^2 2s^2\,{}^1S_0 \to 1s^2 2s 2p\,{}^3P_0$ transitions in berylliumlike ions and the PV $2E1$ $1s 2p\,{}^3P_0 \to 1s 2s\,{}^1S_0$ transition in heliumlike ions. Results of these calculations are found in good agreement with the nonrelativistic predictions of Schäfer *et al.* [13] for the two-electron species but yield some discrepancies with the previously reported excitation rates for berylliumlike ions. These discrepancies can be assigned to the wrong energies, which were used in Ref. [14] in the evaluation of the second-order transition amplitudes.

Theoretical predictions for the PV two-photon transitions in few-electron ions, reported in our and previous publications, clearly indicate the need for high-intensity lasers in order to make these transitions visible. However, such intense laser fields also may lead to sizable Stark shift and mixing of different ionic levels even in the high-Z regime. An accurate treatment of these effects will be required for a better understanding and analysis of future two-photon absorption experiments and is currently under the way.

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