Nuclear excitation by laser-assisted electronic transitions

S. Typel and C. Leclercq-Willain
Physique Nucléaire Théorique et Physique Mathématique, Université Libre de Bruxelles, Case Postale 229, Boulevard du Triomphe, B-1050 Bruxelles, Belgium
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The excitation of nuclear states by electric and magnetic multipole transitions of bound electrons in the presence of a strong laser field is studied. The excitation probability is calculated for general time-dependent electronic states in first-order perturbation theory. An adiabatic description of the dressed electron states in the laser field allows an easy calculation of the excitation function. Ionization effects caused by the laser are considered in a simple manner. The probability for the excitation of the nuclei $^{161}_{66}$Dy, $^{189}_{76}$Os, $^{193}_{77}$Ir, $^{197}_{79}$Au, $^{235}_{92}$U, and $^{237}_{93}$Np is investigated as a function of the laser intensity and the photon energy. The increase of the excitation probability is limited by the ionization of the atom and the lifetime of the states. The de-excitation of the $\frac{3}{2}^+$ (3.5 ± 1.0 eV) state in $^{229}_{90}$Th is an example of the laser-assisted discrete internal conversion.

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I. INTRODUCTION

The excitation of atomic nuclei by electronic transitions has repeatedly been discussed in the literature [1–4] and several experiments have been performed [5–11]. In this process an electron in an outer shell makes a transition to a state of lower energy transferring the difference of the binding energies and angular momentum to the nucleus which is excited from the ground state or a long-lived isomeric state to another state. This mechanism corresponds to the inverse internal conversion (IIC) and is a further kind of electronic deexcitation in addition to radiative transitions and the emission of Auger electrons. The electronic transition requires the existence of a vacancy in the electronic cloud. This hole can be produced, e.g., by means of x-ray absorption by an electron or inelastic scattering of electrons with sufficient energy. Some heavy nuclei possess low-lying states with excitation energies close to the energy differences of the electronic orbits. These nuclear states can be reached by IIC in transitions with the according electromagnetic multipolarities. The possibility of influencing electron states by now available powerful lasers has renewed the interest in this excitation mechanism because the application of a laser field has a strong influence on the usual internal conversion [12–17]. There is the hope that an accurate adjustment of the electromagnetic field of the laser can considerably enhance the transition probabilities [18–20].

The theoretical description of the excitation process needs the knowledge of the electromagnetic interaction between the nucleus and the electron cloud and the action of the laser field on the electron states. These problems were already tackled in different approximations. In Ref. [18] the electron states are described in the potential of a harmonic oscillator whose parameters were individually adjusted to give the correct binding energies and orbital radii. In this approximation the time dependent evolution of the states in the laser field can be given analytically. The nuclear excitation is caused by the Coulomb field of the oscillating charge distribution of the electron cloud. This semiclassical approximation (cf. the Coulomb excitation of nuclei [21,22]) considers no actual transitions between initial and final electronic states with definite quantum numbers. Therefore it gives no explicit selection rules in the electronic part as in a real quantum-mechanical calculation. In Ref. [18] the possible escape of the electrons at large laser intensities is discussed but not explicitly included in the calculation. Only simple estimates for the ionization limit are given. More realistic wave functions of the electrons in the Coulomb potential of the nucleus and a real quantum-mechanical treatment of the excitation (considering only the Coulomb interaction of the charge densities) are used in Ref. [20]. The action of the laser field is included by the use of field dressed states [23,24]. It is assumed that electron states with the same principal quantum number are energetically degenerated. Neglecting the coupling between states of different principal quantum number, the time evolution of the electron states can again be given analytically using parabolic coordinates. But these field dressed states can only be used if the interaction of the electrons with the laser field is stronger than the spin-orbit splitting of the electron states [25]. In multi-electron systems this condition is obviously not met. In Ref. [19] the excitation process with high frequency laser fields was studied using a classical description for the motion of the electrons.

In view of the above approximations, we will present a more realistic description of the excitation process. It includes the electric and magnetic interaction between the electrons and the nucleus and permits the use of more realistic electronic wave functions. The initial creation of a hole state in the electron cloud [20] will not be taken into consideration. This process can be assumed to be independent from the excitation in first approximation. The action of the laser field on the electron states is considered in an adiabatic treatment of the time evolution and the ionization effects are considered in a simple way. The dependence of the excitation probability on the laser intensity and photon energy is studied. The paper is organized as follows. In Sec. II we calculate in general the probability of the nuclear excitation during an electronic transition without a detailed specification of the time-dependent electron states. The action of the laser field on the electron states is described in Sec. III and in this adiabatic approximation the nuclear excitation functions are calculated. The finite lifetimes of the states including the

ionization of the atom in the laser field is considered in Sec. IV. In Sec. V we apply our model to the excitation of states with small excitation energy in the nuclei $^{235}_{92}$U, $^{197}_{79}$Au, $^{193}_{77}$Ir, $^{161}_{66}$Dy, $^{189}_{76}$Os, and $^{237}_{93}$Np. A special case is the $^{229}_{90}$Th nucleus where the deexcitation of a nuclear state is studied. Finally, in Sec. VI, we close with a summary and our conclusions.

II. NUCLEAR EXCITATION INDUCED BY ELECTRONIC TRANSITIONS

The probability for the excitation of a nuclear state $|\psi_f^N\rangle$ with energy E_f^N from a ground state $|\psi_i^N\rangle$ with energy $E_i^N<E_f^N$ during the transition of an electron from the state $|\psi_i^e\rangle$ with energy E_i^e to a stronger bound state $|\psi_f^e\rangle$ with energy $E_f^e<E_i^e$ will in general be much smaller than one. Therefore we can calculate the excitation probability in first order perturbation theory proceeding along the lines of the Coulomb excitation theory [21,22]. The excitation amplitude is given by

$$b_{fi}=\frac{1}{i\hbar}\int dt e^{i\omega_N t}\int\int d^3r d^3r'\left[\rho^N(\boldsymbol{r}')\rho^e(\boldsymbol{r},t)-\frac{1}{c^2}\boldsymbol{J}^N(\boldsymbol{r}')\cdot\boldsymbol{J}^e(\boldsymbol{r},t)\right]\frac{\exp(ik_N|\boldsymbol{r}-\boldsymbol{r}'|)}{|\boldsymbol{r}-\boldsymbol{r}'|} \tag{2.1}$$

with the nuclear excitation energy

$$\hbar\omega_N=E_f^N-E_i^N=\hbar ck_N \tag{2.2}$$

and the nuclear and electronic transition matrix elements of the charge density

$$\rho^N(\boldsymbol{r}')=\langle\psi_f^N|\hat{\rho}(\boldsymbol{r}')|\psi_i^N\rangle,\quad \rho^e(\boldsymbol{r},t)=\langle\psi_f^e(t)|\hat{\rho}(\boldsymbol{r})|\psi_i^e(t)\rangle, \tag{2.3}$$

and the current density

$$\boldsymbol{J}^N(\boldsymbol{r}')=\langle\psi_f^N|\hat{\boldsymbol{J}}(\boldsymbol{r}')|\psi_i^N\rangle,\quad \boldsymbol{J}^e(\boldsymbol{r},t)=\langle\psi_f^e(t)|\hat{\boldsymbol{J}}(\boldsymbol{r})|\psi_i^e(t)\rangle. \tag{2.4}$$

We have assumed that the total initial (final) wave function is a simple product state of the initial (final) nuclear and electronic wave functions. We keep the time dependence of the electronic states in this notation as it is not specified as yet.

The excitation amplitude can be expanded into a sum of contributions of different multipolarity by using the expansion of the Green's function. Assuming that the electrons do not penetrate into the nucleus, i.e., $|\boldsymbol{r}'|<|\boldsymbol{r}|$, the integrand factorizes into a nuclear and an electronic part. Using the continuity equation and integrating by parts in both the nuclear and the electronic part, we remain with

$$b_{fi}=-\frac{4\pi k_N}{\hbar c^2}\sum_{LM}\left[\int d^3r'\rho^N(\boldsymbol{r}')j_L(k_Nr')Y_{LM}(\hat{\boldsymbol{r}}')\left(1+\frac{k_e}{k_N}\right)\int dt e^{i\omega_N t}\int d^3r\rho^e(\boldsymbol{r},t)h_l^{(1)}(k_Nr)Y_{LM}^*(\hat{\boldsymbol{r}})+\sum_{a=e,m}\int d^3r'\boldsymbol{J}^N(\boldsymbol{r}')\cdot\boldsymbol{A}_{LM}(\boldsymbol{r}',a)\int dt e^{i\omega_N t}\int d^3r\boldsymbol{J}^e(\boldsymbol{r},t)\cdot\boldsymbol{B}_{LM}^*(\boldsymbol{r},a)\right] \tag{2.5}$$

with the electric ($a=e$) and magnetic ($a=m$) vector fields $\boldsymbol{A}_{LM}(\boldsymbol{r}',a)$ and $\boldsymbol{B}_{LM}(\boldsymbol{r},a)$ defined in Ref. [22]. Usually the energy difference $E_f^e-E_i^e=\hbar ck_e$ between the final and initial electronic states is the negative of the nuclear energy difference so that the scalar contribution from the charge density cancels out the longitudinal contribution from the current density and only the electric and magnetic contributions remain in (2.5). But for the nuclear excitation due to a laser-assisted electronic transition, this is not necessarily the case; the nuclear and electronic energy differences do not compensate exactly.

We introduce the usual electric ($a=E$) and magnetic ($a=M$) multipole operators $\mathscr{M}(a,LM)$ and additionally the "scalar" multipole operator

$$\mathscr{M}(S,LM)=\frac{(2L+1)!!}{k_N^L}\int d^3r\hat{\rho}(\boldsymbol{r})j_L(k_Nr)Y_{LM}(\hat{\boldsymbol{r}}), \tag{2.6}$$

where j_L is the regular spherical Bessel function. For the excitation amplitude we get

$$b_{fi}=\frac{4\pi}{i\hbar(2L+1)}\sum_{\substack{LM\\a=e,m,s}}\langle\psi_f^N|\mathscr{M}(a,LM)|\psi_i^N\rangle F_{LM}^a \tag{2.7}$$

with the electronic time integrals

$$F_{LM}^a=\int dt e^{i\omega_N t}\langle\psi_f^e(t)|\mathscr{N}(a,LM)|\psi_i^e(t)\rangle. \tag{2.8}$$

Here we have introduced the electric, the magnetic, and the scalar multipole operators defined by

$$\mathscr{N}(E,LM)=\frac{ik_N^L}{c(2L-1)!!L}\int d^3r\hat{\boldsymbol{J}}(\boldsymbol{r})\times[\nabla\times\hat{\boldsymbol{L}}h_L^{(2)}(k_Nr)Y_{LM}(\hat{\boldsymbol{r}})]^*, \tag{2.9}$$

$$\mathscr{N}(M,LM)=\frac{k_N^{L+1}}{c(2L-1)!!L}\int d^3r\hat{\boldsymbol{J}}(\boldsymbol{r})\times[\hat{\boldsymbol{L}}h_L^{(2)}(k_Nr)Y_{LM}(\hat{\boldsymbol{r}})]^*, \tag{2.10}$$

and

$$\mathcal{N}(S,LM)=\left(1+\frac{k_e}{k_N}\right)\frac{ik_N^{L+1}}{(2L-1)!!}\int d^3r\hat{\rho}(\mathbf{r}) \times h_L^{(1)}(k_Nr)Y_{LM}^*(\hat{\mathbf{r}}) \tag{2.11}$$

with the spherical Hankel functions of first and second kind $h_L^{(2)}=h_L^{(1)*}=(j_L+in_L)^*$. The electric multipole operators can be put into a more convenient form by applying the formula

$$\nabla\times\hat{\boldsymbol{L}}[f_L(kr)Y_{LM}(\hat{\mathbf{r}})]=i\nabla\left\{\frac{\partial}{\partial r}[rf_L(kr)]Y_{LM}(\hat{\mathbf{r}})\right\} +ik^2\mathbf{r}f_L(kr)Y_{LM}(\hat{\mathbf{r}}) \tag{2.12}$$

for a spherical cylinder function f_L, integrating by parts and using the continuity equation. For the nuclear part the electric multipole operator reads as

$$\mathcal{M}(E,LM)=\frac{(2L+1)!!}{k_N^L(L+1)}\int d^3r\hat{\rho}(\mathbf{r})\frac{\partial}{\partial r}[rj_L(k_Nr)]Y_{LM}(\hat{\mathbf{r}}) -i\frac{(2L+1)!!}{ck_N^{L-1}(L+1)}\int d^3r\hat{\mathbf{J}}\cdot\mathbf{r}j_L(k_Nr)Y_{LM}(\hat{\mathbf{r}}) \tag{2.13}$$

and reduces in the long-wavelength limit ($k_N\to 0$) to

$$\mathcal{M}(E',LM)=\int d^3rr^L\hat{\rho}(\mathbf{r})Y_{LM}(\hat{\mathbf{r}}), \tag{2.14}$$

whereas the magnetic multipole operator assumes the form

$$\mathcal{M}(M,LM)=\frac{-i}{c(L+1)}\int d^3rr^L\hat{\mathbf{J}}\cdot\hat{\boldsymbol{L}}Y_{LM}(\hat{\mathbf{r}}). \tag{2.15}$$

This approximation is well justified by the small dimensions of the nucleus as compared to the length $1/k_N$. The long-wavelength limit of the nuclear scalar operator (2.6) leads to the same result as the electric operator so that we can combine both contributions to the excitation amplitude by introducing the operator

$$\mathcal{N}(E',LM)=\mathcal{N}(E,LM)+\mathcal{N}(S,LM) \tag{2.16}$$

in the electronic part. The application of relation (2.12) leads to

$$\mathcal{N}(E',LM)=i\frac{k_N^L}{(2L-1)!!}\int d^3r\hat{\rho}(\mathbf{r})\left\{\frac{k_e}{L}\frac{\partial}{\partial r}[rh_L^{(1)}(k_Nr)]+(k_N+k_e)h_L^{(1)}(k_Nr)\right\}Y_{LM}^*(\hat{\mathbf{r}}) +\frac{k_N^{L+2}}{c(2L-1)!!L}\int d^3r\hat{\mathbf{J}}\cdot\mathbf{r}h_L^{(1)}(k_Nr)Y_{LM}^*(\hat{\mathbf{r}}) \tag{2.17}$$

where the factor k_e appears as before from the application of the continuity equation to the electron current. For $k_N\to 0$ we get the long-wavelength limit

$$\mathcal{N}(E,LM)=\int d^3r\frac{1}{r^{L+1}}\hat{\rho}(\mathbf{r})Y_{LM}^*(\hat{\mathbf{r}}). \tag{2.18}$$

The magnetic operator reduces to

$$\mathcal{N}(M,LM)=\frac{i}{cL}\int d^3r\frac{1}{r^{L+1}}\hat{\mathbf{J}}\cdot\hat{\boldsymbol{L}}Y_{LM}^*(\hat{\mathbf{r}}) \tag{2.19}$$

in this limit. In the electronic part we can use these approximations only for very small nuclear transition energies, the electronic wave functions being much more extended than the nuclear wave functions.

The excitation probability for the transition between nuclear states with total angular momentum J_i^N and J_f^N during the electronic transition from state J_i^e to state J_f^e is now obtained as

$$P_{fi}=\frac{1}{(2J_i^N+1)(2J_i^e+1)}\sum_{\substack{M_i^NM_f^N\\M_i^eM_f^e}}|b_{fi}|^2. \tag{2.20}$$

With the reduced transition probability for the nuclear excitation ($a=E,M$)

$$B(aL,J_i^N\to J_f^N)=\frac{1}{(2J_i^N+1)}\sum_{M_i^NM_f^NM}|\langle J_f^NM_f^N|\mathcal{M}(a,LM)|J_i^NM_i^N\rangle|^2, \tag{2.21}$$

we finally get for the excitation probability

$$P_{fi}=\sum_{\substack{L\\a=e,m}}B(aL,J_i^N\to J_f^N)f(aL,J_i^e\to J_f^e) \tag{2.22}$$

with the electronic excitation function

$$f(aL,J_i^e\to J_f^e)=\frac{1}{(2L+1)^3}\left(\frac{4\pi}{\hbar}\right)^2\frac{1}{(2J_i^e+1)}\sum_{M_i^eM_f^eM}|F_{LM}^a|^2. \tag{2.23}$$

This function depends on the initial and final wave functions of the electronic states which are affected by the action of the laser field. Multiplying the excitation probability P_{fi} with the

cross section σ for the production of the hole state we will finally get the cross section for the excitation process assuming two independent processes. Because the cross section σ depends on the chosen mechanism for the hole creation we will not investigate this in the following. The full information of the nuclear excitation during the electronic transition is already contained in the probability P_{fi}.

III. DYNAMIC OF ELECTRON STATES IN A LASER FIELD

The description of bound electron states in the time-dependent laser field is, in general, very complicated since one has to solve a time-dependent many-body problem. For simplicity we describe the electrons as independently moving in a Coulomb field with a nuclear charge number which is individually adjusted to yield the correct binding energies. The effect of the perturbing electromagnetic field is calculated for the one-electron states separately. The laser field of frequency ω_L and amplitude $\mathscr{E}_0$ is treated classically. The field can be considered homogeneous as its wavelength is much larger than the atomic dimensions. Assuming a polarization in the z direction, the time-dependent perturbation of the electron states is given by

$$V(\boldsymbol{r},t)=e\mathscr{E}(t)z \quad \text{with} \quad \mathscr{E}(t)=\mathscr{E}_0\sin(\omega_L t). \tag{3.1}$$

The unperturbed electron eigenfunctions $|n\rangle$ are solutions of the unperturbed Schrödinger equation

$$H_0|n\rangle=E_n^0|n\rangle \tag{3.2}$$

with the energy $E_n^0=\hbar\omega_n^0$. The wave function under the action of the perturbation can in general be expanded as

$$|\psi(t)\rangle=\sum_n a_n(t)|n\rangle\exp(-i\omega_n^0 t) \tag{3.3}$$

with time-dependent amplitudes $a_n(t)$. They obey the set of differential equations

$$\frac{d}{dt}a_m(t)=\frac{1}{i\hbar}\sum_n a_n(t)V_{mn}\sin(\omega_L t)\exp[-i(\omega_n^0-\omega_m^0)t] \tag{3.4}$$

with

$$V_{mn}=\langle m|e\mathscr{E}_0 z|n\rangle \tag{3.5}$$

and the initial condition $a_m(0)=\delta_{mi}$ when the electron was in the state $|i\rangle$ before the laser was switched on at time $t=0$. The set of equations (3.4) can be solved, in principle, numerically. Depending on the number of considered states and the used step size of the integration the calculation will be very extensive. Carrying out this procedure we notice however that the time dependence of the wave function shows a very simple behavior. The electronic states follow adiabatically the slowly changing perturbation field because the electronic transition energies $E_m^0-E_n^0$ are much larger than the laser energy $\hbar\omega_L$. If the adiabaticity criterion

$$\left|\frac{\omega_n^0-\omega_m^0}{\omega_L}\right|\gg 1 \tag{3.6}$$

is valid the following simple adiabatic approximation can be used. The time-dependent wave function of an initial state $|l\rangle$ for $t>0$ can be written as

$$|\psi_l(t)\rangle=|\phi_l[\mathscr{E}(t)]\rangle\exp\left[-\frac{i}{\hbar}\int_0^t E_l(t')dt'\right] \tag{3.7}$$

where $|\phi_l\rangle$ is the solution of the time-independent Schrödinger equation

$$[H_0+V(t)]|\phi_l[\mathscr{E}(t)]\rangle=E_l(t)|\phi_l[\mathscr{E}(t)]\rangle \tag{3.8}$$

with a constant perturbation depending on the electric-field strength $\mathscr{E}(t)$ at each time t. The adiabatic energy is given in second-order time-independent perturbation theory by

$$E_l(t)=E_l^0[1+C_l\sin^2(\omega_L t)] \tag{3.9}$$

with

$$C_l=\frac{1}{E_l^0}\sum_{n\neq l}\frac{V_{ln}V_{nl}}{E_l^0-E_n^0}. \tag{3.10}$$

For the spatial part of the wave function we get in this approximation

$$|\phi_l\rangle=\sum_k\ [\delta_{lk}+B_{lk}\sin(\omega_L t)+A_{lk}\sin^2(\omega_L t)]|k\rangle \tag{3.11}$$

with dimensionless coefficients

$$B_{lk}=\begin{cases}0, & k=l\\ \dfrac{V_{kl}}{E_l^0-E_k^0}, & k\neq l\end{cases}$$

$$A_{lk}=\begin{cases}-\dfrac{1}{2}\displaystyle\sum_{m\neq l}\left|\frac{V_{ml}}{E_l^0-E_m^0}\right|^2, & k=l\\ \dfrac{1}{E_l^0-E_k^0}\displaystyle\sum_{m\neq l}\frac{V_{km}V_{ml}}{E_l^0-E_m^0}, & k\neq l.\end{cases} \tag{3.12}$$

All the numbers A_{lk}, B_{lk}, and C_l depend on the amplitude $\mathscr{E}_0$ of the laser field. In the calculation we have made use of the fact that the perturbation does not couple states of equal quantum numbers due to the negative parity of the coupling potential V. The coefficients B_{lk} and A_{lk} determine the coupling of the unperturbed state $|l\rangle$ to states $|k\rangle$ which have the opposite and the same parity as the state $|l\rangle$, respectively. The quantity $C_l E_l^0$ is simply the Stark shift of the level $|l\rangle$ in a constant electric field of amplitude $\mathscr{E}_0$. The time dependence for the exponential factor in (3.7) has the form

$$\exp\left[-\frac{i}{\hbar}\int_0^t E_l(t')dt'\right]$$

$$=\exp(-i\omega_l t)\exp\left[i\frac{C_l}{4}\frac{\omega_l^0}{\omega_L}\sin(2\omega_L t)\right] \tag{3.13}$$

with the shifted frequencies

$$\omega_l = \omega_l^0 \left(1 + \frac{C_l}{2} \right). \tag{3.14}$$

The action of the laser field introduces a time dependence with the double laser frequency because the Stark effect gives a change in the energy only in the contribution of second order. Using the generating function of Bessel functions

$$\exp[i\lambda \sin(\omega t)] = \sum_{N=-\infty}^{\infty} J_N(\lambda) \exp(iN\omega t) \tag{3.15}$$

we obtain for the total wave function

$$|\psi_l(t)\rangle = \sum_{Nrn} J_N\left(\frac{C_l \omega_l^0}{4\omega_L} \right) D_{ln}^r |n\rangle \exp(-i\omega_l t) \times \exp[i(2N+r)\omega_L t] \tag{3.16}$$

with the coefficients

$$D_{ln}^0 = \delta_{ln} + \frac{A_{ln}}{2}, \quad D_{ln}^1 = \frac{B_{ln}}{2i} = -D_{ln}^{-1}, \quad D_{ln}^2 = -\frac{A_{ln}}{4} = D_{ln}^{-2}. \tag{3.17}$$

The summation over r covers the range from -2 to 2 because we have restricted our approximation to second-order perturbation theory. Simultaneously with the initial state $|l\rangle$, each state $|n\rangle$ which couples to $|l\rangle$ gets a ladder of Floquet states ($N=-\infty,\ldots,\infty$) with an energy separation of $2\hbar\omega_L$ and an amplitude determined by the Bessel function J_N. The argument of the Bessel function is the ratio between the half Stark shift of the initial level and the double laser frequency. This factor rises linearly with the laser intensity. The use of the second-order perturbation theory for $|\phi_l\rangle$ limits the application of our approximation to not too large laser intensities. But as the intensity rises the electrons (beginning with the outer shells) will be removed from the atom in a ionization process before this will be a problem.

Comparing our result for the field-dressed electron states with the description [20] we note a structural similarity but characteristic differences. Instead of the first-order perturbation in the linear Stark effect, our solution is determined by the quadratic Stark effect. The energies of the states are not separated by the single but by the double photon energy. We are not limited to the coupling of states with equal principal quantum number and equal energy in our description.

The time integral for the duration T of the laser pulse in the electronic excitation function (2.8) can now be done analytically

$$F_{LM}^a = \int_0^T dt e^{i\omega_N t - \gamma t} \langle \psi_f^e(t) | \mathcal{N}(a,LM) | \psi_i^e(t) \rangle = \sum_{\substack{Nrn \\ Msm}} J_N\left(\frac{C_i \omega_i^0}{4\omega_L} \right) J_M\left(\frac{C_f \omega_f^0}{4\omega_L} \right) D_{in}^r D_{fm}^{s*} \langle m | \mathcal{N}(a,LM) | n \rangle \frac{[\exp(i\Omega T - \gamma T) - 1]}{i\Omega - \gamma} \tag{3.18}$$

with

$$\Omega = \omega_N - \omega_i + \omega_f + [2(N-M) + r - s]\omega_L. \tag{3.19}$$

We have introduced an additional factor $\exp(-\gamma t)$ in the integral to account for the finite lifetime of the electronic and nuclear states. The number of participating photons from the laser field is given by

$$n_\gamma = 2(|N| + |M|) + |r| + |s|. \tag{3.20}$$

For a certain transition multipolarity the contributions to the electronic excitation function with either even or odd n_γ do not vanish. Through the application of the laser, transitions which are not possible in the unperturbed case due to selection rules may get a finite probability.

To remove the dependence of the excitation probability on the time T, we assume that the duration of the laser pulse is much longer than the lifetime of the hole states. The widths of electron hole states in heavy atoms are usually in the range of some eV [28] where 1 eV corresponds to a half-life of about 0.5 fs. We can carry out the limit $T \to \infty$ and get

$$F_{LM}^a = \sum_{\substack{Nrn \\ Msm}} J_N\left(\frac{C_i \omega_i^0}{4\omega_L} \right) J_M\left(\frac{C_f \omega_f^0}{4\omega_L} \right) D_{in}^r D_{fm}^{s*} \frac{\langle m | \mathcal{N}(a,LM) | n \rangle}{\gamma - i\Omega}. \tag{3.21}$$

Choosing a certain laser frequency the possible enhancement of the excitation probability due to the application of the laser is determined by the value of the Bessel functions, the D coefficients, the energy $\hbar\Omega$, and the width $\hbar\gamma$ which all depend on the intensity of the laser. Without the laser field the electronic time integral reduces simply to

$$F_{LM}^a = \frac{\langle f | \mathcal{N}(a,LM) | i \rangle}{\gamma - i(\omega_N - \omega_i^0 + \omega_f^0)}. \tag{3.22}$$

The excitation probability may then be expressed by

$$P_{fi} = \sum_{\substack{L \\ a=e,m}} \frac{\omega_{\rm int}^2(aL)}{(\omega_N - \omega_i^0 + \omega_f^0)^2 + \gamma^2} \tag{3.23}$$

with the squared interaction energies

$$E^2_{\rm int}(aL)=[\hbar\omega_{\rm int}(aL)]^2 = \frac{(4\pi)^2}{(2L+1)^3}B(aL,J^N_i\to J^N_f) \times \sum_{M^e_i M^e_f M}\frac{|\langle f|\mathcal{M}(a,LM)|i\rangle|^2}{(2J^e_i+1)}. \quad (3.24)$$

These quantities serve as measure for the strength of the electron-nucleus interaction in the laser-free case.

If the frequency Ω in Eq. (3.21) goes to zero we get into resonance with the nuclear excitation. This can be achieved in principle by adjusting a multiple of the laser frequency ω_L to the difference between the nuclear excitation frequency ω_N and the difference $\omega_i-\omega_f$ of the Stark-shifted electron frequencies. The amplification factor $\mathcal{A}$ for the probability by the resonant laser interaction can be calculated easily in a rough approximation when the resonant contribution dominates in Eq. (3.21). We compare the expressions for the squared modulus of the excitation function F^a_{LM} in the resonant $2N$-photon excitation and in the process without the laser. Because the outermost electrons, i.e., the initial electron states, have a much larger Stark shift than the more tightly bound electrons we get the expression

$$\mathcal{A}_{2N}\approx\left[J_N\left(\frac{C_i\omega_i^0}{4\omega_L}\right)\frac{1}{\gamma}\right]^2 \Big/ \frac{1}{(2N\omega_L)^2+\gamma^2} \quad (3.25)$$

which can be simplified assuming that the width $\hbar\gamma$ and the Stark shift $C_i\hbar\omega_i^0$ of the initial electron state are much smaller than the photon energy $\hbar\omega_L$. Using the approximation of the Bessel function for small arguments, the amplification factor becomes

$$\mathcal{A}_2\approx\left(\frac{C_i\omega_i^0}{4\gamma}\right)^2 \quad (3.26)$$

for the two-photon resonance, independent of the photon energy. Therefore a large amplification can be obtained if the total width of the states is much smaller than the Stark shift of the initial electron state.

IV. FINITE LIFETIME OF STATES AND EFFECTS OF IONIZATION

There are different contributions to the width $\hbar\gamma$ in Eq. (3.21)

$$\gamma=\frac{1}{2\hbar}[\Gamma^N(i)+\Gamma^N(f)+\Gamma^e(i)+\Gamma^e(f)] \quad (4.1)$$

due to the finite lifetime of the initial and final nuclear and electronic states. The width $\Gamma^N(i)$ of the initial nuclear state is usually zero or very small determined by, e.g., the β-decay rate of the ground state. The width $\Gamma^N(i)$ of the final nuclear state will mainly depend on the probability for the transition into lower nuclear states either by electromagnetic or nuclear decay. The electronic contributions $\Gamma^e(l)$ of the initial and final states ($l=i,f$) to the width $\hbar\gamma$ contain spontaneous processes and laser-induced transitions.

The lifetime of the initial state depends on the probability for removing the electron from this state. This can be caused either by the action of the laser field, the radiative transition of the electron to a state with lower energy which is not occupied, or an Auger process. The width of the initial electron state will increase with growing laser intensity on account of ionization; comparatively the spontaneous processes become less important.

We estimate the ionization widths for the electron states in a simple approximation to take into account the action of the laser field. As the electron states adjust immediately to the change of the potential by the electric field of the laser we can use a simple barrier penetration model to calculate in a rough approximation the ionization width $\Gamma^e_L(l)$ due to the laser field for a state with wave function $|\psi_l(t)\rangle$. This procedure is consistent with the adiabatic description of the time development of the electron states. During one laser period the electron has a time-dependent probability to be found in a unperturbed electronic state $|k\rangle$ [cf. Eq. (3.11)]. Using the mean probabilities we can calculate the width

$$\Gamma^e_L(l)=(1+A_{ll}+\tfrac{3}{8}A^2_{ll})\Gamma_l+\sum_{k\neq l}(\tfrac{1}{2}B^2_{lk}+\tfrac{3}{8}A^2_{lk})\Gamma_k \quad (4.2)$$

from the widths $\Gamma_k=\Gamma(|nJMl\rangle)$ of the usual one-electron wave functions $|k\rangle=|nJMl\rangle$ with principal quantum number n, total angular momentum J, magnetic quantum number M, and orbital angular momentum l. These states in the spherical basis are linear combinations of states in the parabolic basis with the same principal quantum number n, the parabolic quantum number q, and the magnetic quantum number m. Accordingly, we calculate the width as a sum

$$\Gamma(|nJMl\rangle)=\sum_{q=0}^{n-|m|-1}\sum_{m=-l}^{l}C^{nJl}_{qMm}\Gamma(|nqm\rangle), \quad (4.3)$$

where

$$C^{nJl}_{qMm}=(2J+1)(2l+1) \times\begin{pmatrix} l & \frac{1}{2} & J \\ m & M-m & -M\end{pmatrix}^2\begin{pmatrix} \frac{n-1}{2} & \frac{n-1}{2} & l \\ \frac{m+k}{2} & \frac{m-k}{2} & -m\end{pmatrix}^2 \quad (4.4)$$

with

$$k=2q-n+|m|+1 \quad (4.5)$$

the probability of finding the electron in the respective state. A more rigorous treatment would directly use the wave functions in the representation in parabolic coordinates for the electronic states from the beginning. The widths $\Gamma(|n,q,m\rangle)$ of the parabolic wave functions are finally calculated from the barrier penetration probability in the Wentzel-Kramers-Brillouin (WKB) approximation [26]. When the laser amplitude $\mathcal{E}_0$ rises and the barrier vanishes there will be no bound electron state any more so that the

TABLE I. Characteristics of the nuclear and electronic transitions for different atoms.

	$^{161}_{66}$Dy	$^{189}_{76}$Os	$^{193}_{77}$Ir	$^{197}_{79}$Au	$^{229}_{90}$Th	$^{235}_{92}$U	$^{237}_{93}$Np
Multipolarity aL	$M1$	$M1$	$M1$	$M1$	$M1$	$E3$	$E1$
Nuclear transition	$\frac{5}{2}^+ \rightarrow \frac{7}{2}^+$	$\frac{3}{2}^- \rightarrow \frac{5}{2}^-$	$\frac{3}{2}^+ \rightarrow \frac{1}{2}^+$	$\frac{3}{2}^+ \rightarrow \frac{1}{2}^+$	$\frac{3}{2}^+ \rightarrow \frac{5}{2}^+$	$\frac{7}{2}^- \rightarrow \frac{1}{2}^+$	$\frac{3}{2}^+ \rightarrow \frac{1}{2}^+$
Excitation energy (keV)	43.8211 [a]	69.537 [b]	73.041 [c]	77.351 [d]	−0.0035 [e]	0.0768 [f]	102.96 [g]
Reduced transition probability $B(aL,\downarrow)$ (W.u.)	0.026[a]	0.00252 [b]	0.00098 [c]	0.00409 [d]	0.0198 [h]	1.0	0.0198 [g]
Electronic transition	$2s\frac{1}{2} \rightarrow 1s\frac{1}{2}$	$3s\frac{1}{2} \rightarrow 1s\frac{1}{2}$	$3s\frac{1}{2} \rightarrow 1s\frac{1}{2}$	$3s\frac{1}{2} \rightarrow 1s\frac{1}{2}$	$7s\frac{1}{2} \rightarrow 8s\frac{1}{2}$	$6p\frac{3}{2} \rightarrow 5d\frac{5}{2}$	$2p\frac{3}{2} \rightarrow 1s\frac{1}{2}$
This work	3.1×10^{-4}	1.1×10^{-5}	1.6×10^{-6}	8.2×10^{-6}	2.1×10^{-14}	2.4×10^{-23}	1.7×10^{-6}
$E^2_{\rm int}$ $(eV)^2$ i	4.62×10^{-5}	3.8×10^{-4}	4.97×10^{-5}	3.07×10^{-4}		1.9×10^{-17} [j]	9.6×10^{-6}
Two-photon resonance energy $\hbar\omega_L$ (eV)	455.13	632.245	61.13	37.59	0.1065	0.575	954.46

[a]Reference [30].
[b]Reference [31].
[c]Reference [32].
[d]Reference [33].
[e]Reference [35].
[f]Reference [29].
[g]Reference [34].
[h]Reference [36].
[i]Reference [4].
[j]Value converted to $B(E3)=1.0$ W.u.

electron will be immediately removed from the atom. The time constant of this process will be determined by the time dependence of the laser field. As a simple approximation we assume the value $4\hbar\omega_L$ (the field reaches its first maximum after one quarter of the laser period) for the width at the ionization threshold. For larger amplitudes of the electric field, the ionization threshold will be reached before the field attains its maximum strength. For simplicity, we assume a linear scaling of the width with the field strength. This method should give a reasonable approximation to the ionization widths.

We also have to consider that the interaction with the laser field leads not only to larger widths of the initial electron states but also to a decrease of the widths of the final states. The more electrons are removed from the outer shells of the atom in the ionization, the smaller is the width of the final electron state, a hole state in an inner shell. Finally it gets an infinite lifetime when the intensity reaches its ionization threshold. We can take this effect into account in a very simple method for a rough estimation. The width of a final state is assumed to be reduced from its value in the laser-free case proportionally to the number of ionized electron states above it.

V. APPLICATION TO INDIVIDUAL NUCLEI

We will now examine the excitation of some heavy nuclei employing the theory developed before. This will give a feeling about the importance of the various effects in the laser-assisted excitation process. The characteristics of the nuclear and electronic transitions for the examined atoms are given in Table I.

The matrix elements for the coupling of the electron states by the perturbation of the laser field $\langle m|e\mathscr{E}_0 z|n\rangle$ and of the transition operators $\langle m|\mathscr{N}(a\lambda\mu)|n\rangle$ are calculated with nonrelativistic one-electron wave functions in a Coulomb potential where the nuclear charge has been adjusted to give the correct binding energies of the states. This should be a reasonable approximation for a first estimation of the excitation probability. The binding energies were taken from Ref. [27]. The screening of the nuclear charge by the more strongly bound electrons leads to quite small effective charge numbers for the Coulomb potential of the outer electrons. A calculation with relativistic wave functions of the electrons is possible but would increase the numerical expenses and is beyond the aim of the present work. The long-wavelength limit of the electronic excitation operators was only used for the cases of $^{235}_{92}$U and $^{229}_{90}$Th, in the other cases the full expressions were used.

Of course, we cannot expect to get really quantitative results. However, the essential dependences will become clear and may indicate the direction for future investigations. The absolute values of the excitation probability are rather uncertain and may change considerably in more exact calculations of the electronic transition matrix elements. This is clearly seen in Table I where the squared interaction energies [Eq. (3.24)] in our calculation are compared with the results of Ref. [4] where a relativistic variant of the Hartree-Fock-Slater method was used for the generation of the electronic

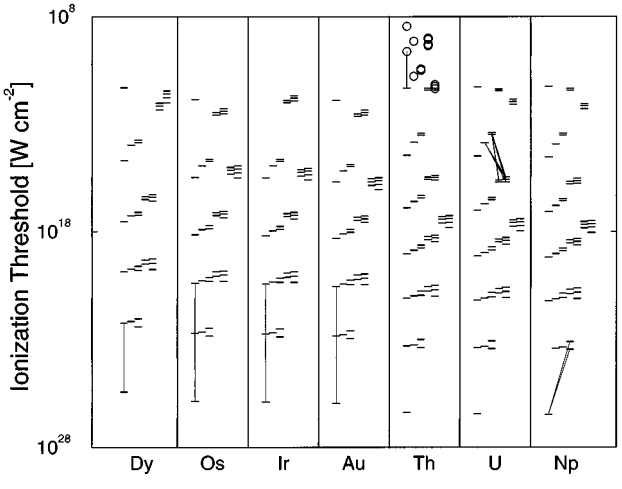

FIG. 1. Ionization thresholds of the electron states in the investigated atoms. The states are arranged for each element from left to right in the order $s\frac{1}{2}$, $p\frac{1}{2}$, $p\frac{3}{2}$, $d\frac{3}{2}$, $d\frac{5}{2}$, $f\frac{5}{2}$, $f\frac{7}{2}$, and from bottom to top with increasing principal quantum number. States not occupied by electrons are given by circles in the case of Th. The considered electronic transitions are marked with thin lines. Corresponding to the Stark shift, the sublevels for total angular momentum J are splitted where the ionization threshold increases with increasing $|M|$.

wave functions. In general, the electronic-nuclear interaction becomes larger for lower multipolarities and transitions between inner electron shells. The influence of the shape of the electronic wave function on the value of the matrix element is easily understandable from the radial dependence of the transition operators. For small radii we have a r^{-L-1} dependence from the Hankel functions enhancing the contributions of the inner part of the wave function where relativistic and many-body effects like the antisymmetrization are very important. This is most noticeable in the $E3$ transition of $^{235}_{92}$U. On the other hand, the calculated values of the Stark shift should not be so sensitive to the used wave functions because the contributions in the inner region of the radial integral are suppressed.

The assumption of independent one-electron states will also limit the predictive power of our calculations. Perturbations in the other electronic states through the interaction of the laser field will have an influence on the binding energy and wave function of each electron because of many body effects. The changes in the energies may be larger than the widths and Stark shifts of the initial states.

The contributions of the spontaneous electronic transitions to the widths of the initial electron states are neglected as they are small compared to the width of the final electronic states (taken from [28] except for $^{235}_{92}$U and $^{229}_{90}$Th). The ionization widths of the initial electronic states are taken into account as described in Sec. IV. In Fig. 1 we give the calculated ionization thresholds for the electron states of the considered atoms. The threshold for the ionization of the atom for the different electron states span a wide range of laser intensities. The outermost electrons will escape the atom at laser intensities below 10^{12} W cm^{-2}, whereas the 1s electrons are affected only for much larger intensities of about 10^{26} W cm^{-2}. The widths of the nuclear states can be neglected due to their long lifetimes as compared to the lifetimes of the electronic states.

A. $^{235}_{92}$U

The long-lived ($T\frac{1}{2}\approx 25$ min [29]) isomeric $\frac{1}{2}^{+}$ state can be reached in an $E3$ transition from the $\frac{7}{2}^{-}$ ground state. The nuclear energy difference corresponds almost exactly to the difference in the binding energies of the $6p\frac{3}{2}$ and $5d\frac{5}{2}$ electrons. By carefully adjusting the laser frequency a resonant transition can be achieved.

The contributions of spontaneous transitions to the widths of the final electronic states are estimated from the radiative widths of these states. A calculation with the one-electron wave functions gives approximately 1 meV for the O_1, 0.5 meV for the $O_{2,3}$, 0.1 meV for the $O_{4,5}$, 0.02 meV for the P_1, and 0.01 meV for the $P_{2,3}$ shells. The higher shells are assumed to have zero widths. These numbers should be taken as lower limits as compared to the actual widths of these states since further decay processes than radiative transitions contribute.

To begin with, we examine the dependence of the excitation probability on the laser intensity for the $6p\frac{3}{2} \rightarrow 5d\frac{5}{2}$ electron transition (Fig. 2). Without the perturbation by the laser, we calculate a probability of only 0.185×10^{-22}. Without the long-wavelength approximation this value would change by about 0.5×10^{-2}% because of the small 76.8-eV excitation energy. The photon energy of the laser field is chosen as $\hbar\omega_L = 0.575$ eV, half the difference between the nuclear and electronic energy differences, to get a resonant excitation in case of a two-photon absorption from the laser. With increasing laser intensity the excitation probability first rises to a maximum at approximately 3×10^{12} W cm^{-2} and finally drops far below the unperturbed value. This behavior can be explained by investigating the electronic excitation function [Eq. (3.21)]. For small laser intensities contributions with $n_\gamma \neq 0$ are insignificant and ionization effects can be neglected. The weak dependence of the probability on the laser intensity is only determined by the magnitude of the stark shift of the electronic energies and the coupling of initial and final states to other electronic states. The relative shift of the energies amounts to only 0.4×10^{-4} for the initial and 0.2×10^{-5} for the final state at 10^{13} W cm^{-2}, corresponding to 4 and 0.06 meV, respectively. With increasing laser intensity the two-photon exchange contribution becomes important as the comparison with the excitation probability in the case of a static electric field of equal strength shows. The argument of the Bessel function J_1 in Eq. (3.21) rises. The energy denominator, limited from below by the width of the electronic state, is very small because of the tuning of the photon energy $\hbar\omega_L$.

The enhancement of the probability through the absorption of photons from the laser field is not very large. It reaches a factor of about 2 at approximately 3×10^{12} W cm^{-2}. Not much above 3×10^{13} W cm^{-2} ionization effects become strong. The probability drops steeply as the intensity exceeds the ionization threshold of the initial state. The steps in the probability curve are related to the crossing of ionization thresholds of states which couple to the initial state by the laser perturbation. The effect of ionization is very pronounced in looking at the static electric field. In this

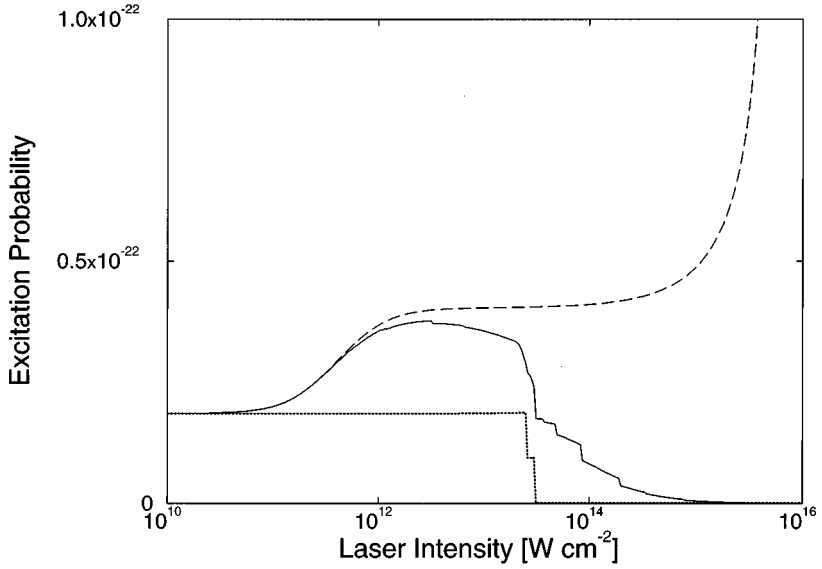

FIG. 2. Excitation probability of the $\frac{1}{2}^+$ state in the $^{235}_{92}$U nucleus for the $6p\frac{3}{2} \rightarrow 5d\frac{5}{2}$ electronic $E3$ transition as a function of the laser intensity with a photon energy of $\hbar\omega_L = 0.575$ eV. Solid line: full calculation; long-dashed line: without ionization; short-dotted line: excitation probability in a constant electric field with corresponding field strength.

calculation the ionization widths have been taken as the energies of the states giving a rough approximation of the typical time constant of the process. Without the escape of the electrons a rather large excitation probability could be reached with the help of the laser. At intensities much above 10^{15} W cm^{-2} processes with more than two photons from the laser field become significant. The Stark-shift of the electronic energies increases rapidly and the theoretical description using only second-order perturbation theory for the electron-laser coupling is no longer valid.

In addition to the $6p\frac{3}{2} \rightarrow 5d\frac{5}{2}$ transition investigated before, the energy differences of the $6p\frac{1}{2} \rightarrow 5d\frac{5}{2}$ and $6p\frac{3}{2} \rightarrow 5d\frac{3}{2}$ transitions are also similar to the nuclear energy difference. To get into resonance, photon energies of 4.695 and 6.235 eV, respectively, are needed. For these energies the adiabaticity condition Eq. (3.6) is not met and the theoretical description of the time dependence of the electron states fails. Because of these larger energies the probability for the nuclear excitation through these two electronic transitions is smaller than in the case of the $6p\frac{3}{2} \rightarrow 5d\frac{5}{2}$ transition. We get the values 0.115×10^{-23} and 0.308×10^{-24}, respectively, in the laser-free case. With a photon energy of 0.575 eV no enhancement can be reached by the application of the laser.

An electronic transition $6p\frac{1}{2} \rightarrow 5d\frac{3}{2}$ with an $E3$ multipolarity is not possible without the laser interaction due to the selection rules. The coupling of initial and final states to other electronic states with increasing laser intensity allows this transition but it is too weak to give a sufficiently large contribution to the excitation probability. We get a value of 0.11×10^{-30} for this transition at an intensity of 10^{13} W cm^{-2} and a photon energy of 0.575 eV. This is eight orders of magnitude smaller than the $6p\frac{3}{2} \rightarrow 5d\frac{5}{2}$ transition.

The dependence of the excitation probability on the photon energy is studied in Fig. 3 for the $6p\frac{3}{2} \rightarrow 5d\frac{5}{2}$ electronic transition. At an intensity of about 3×10^{12} W cm^{-2} we get the maximum enhancement of the probability in the resonance case (cf. Fig. 2) because ionization effects are still small. The sharp resonance from the two-photon absorption is well seen at an energy around 0.575 eV. At an intensity of 3×10^{13} W cm^{-2}, close to the ionization thresholds for the

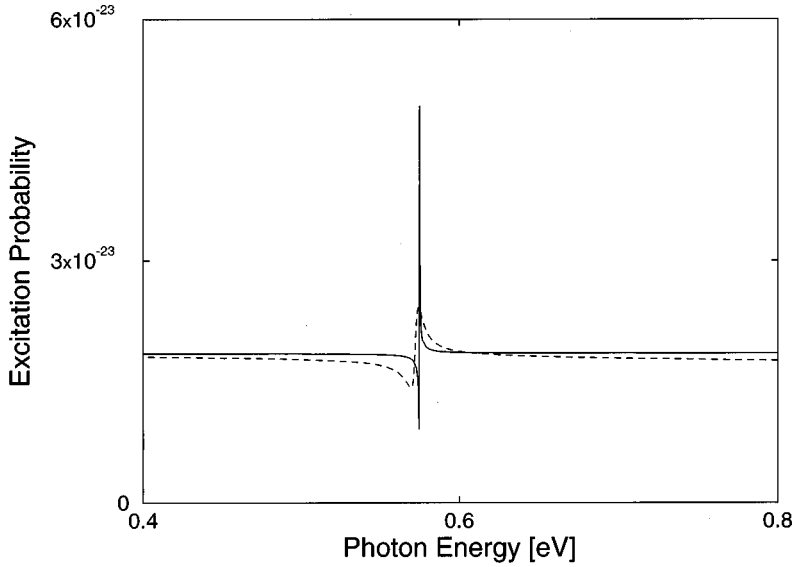

FIG. 3. Excitation probability of the $\frac{1}{2}^+$ state in the $^{235}_{92}$U nucleus for the $6p\frac{3}{2} \rightarrow 5d\frac{5}{2}$ electronic $E3$ transition as a function of the photon energy with a laser intensity of 3×10^{12} W cm^{-2} (solid line) and 3×10^{13} W cm^{-2} (dashed line).

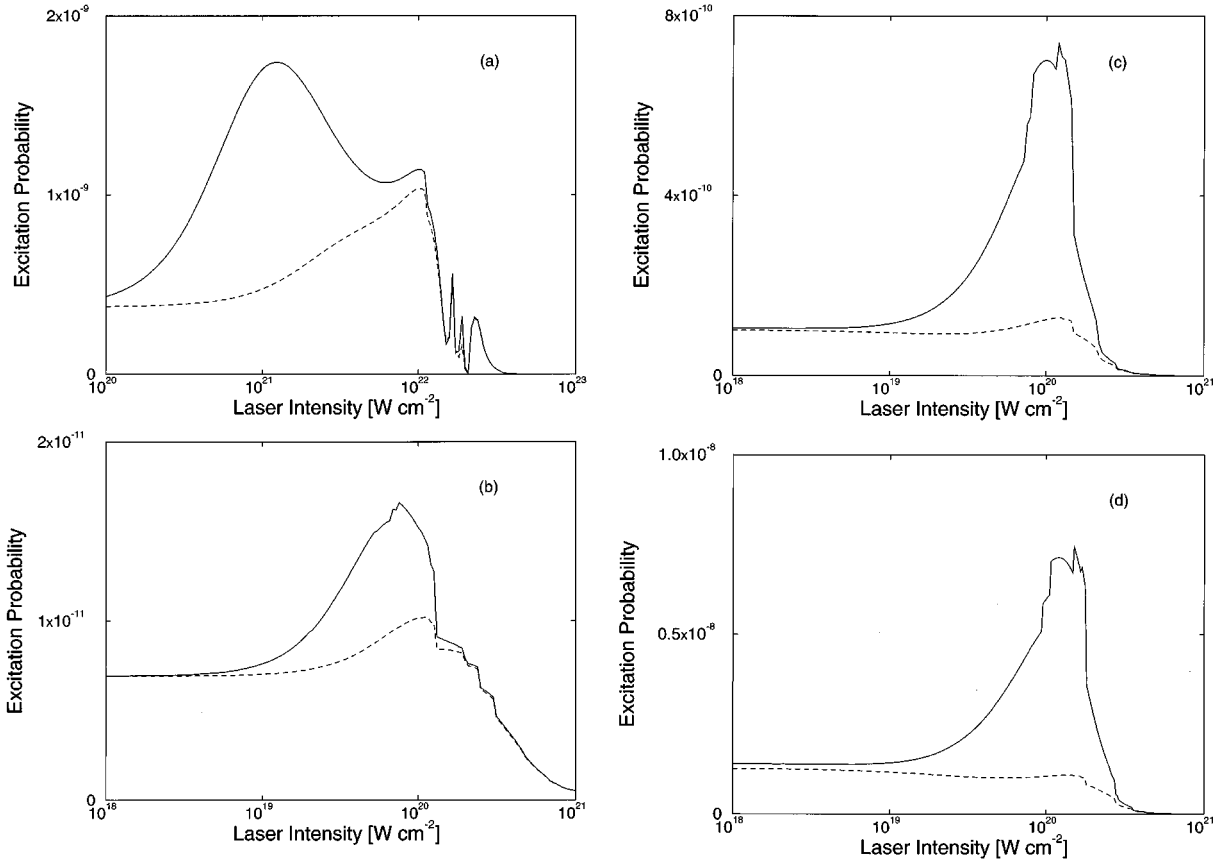

FIG. 4. Excitation probability as a function of the laser intensity. The photon energy is fixed to the energy of a resonant two-photon process. Calculation with (solid line) and without (dashed line) reduction of the final state width by the laser induced ionization. Excitation of the (a) $\frac{7}{2}^+$ state in $^{161}_{66}$Dy with a $2s\frac{1}{2} \rightarrow 1s\frac{1}{2}$ electronic $M1$ transition, (b) $\frac{5}{2}^-$ state in $^{189}_{76}$Os with a $3s\frac{1}{2} \rightarrow 1s\frac{1}{2}$ electronic $M1$ transition, (c) $\frac{1}{2}^+$ state in $^{193}_{77}$Ir with a $3s\frac{1}{2} \rightarrow 1s\frac{1}{2}$ electronic $M1$ transition, (d) $\frac{1}{2}^+$ state in $^{197}_{79}$Au with a $3s\frac{1}{2} \rightarrow 1s\frac{1}{2}$ electronic $M1$ transition.

initial states, the resonance is less pronounced due to the larger widths and a little shifted to smaller energies because of the Stark shift. The probability also becomes smaller with increasing intensity. The resonant contributions from the four-photon, six-photon, or higher photon processes at the corresponding smaller resonance energies are insignificant to the total excitation probability due to the small ratio of the Stark shift to the laser frequency in the argument of the Bessel functions.

We can hardly compare our results for the increase of the excitation probability by the application of the laser with earlier calculations. The useful laser intensities are completely limited by the ionization of the atom, at least six orders of magnitude smaller than the values used in Ref. [18]. These authors give only some rough approximation for the limit in intensity for the laser-induced electron escape from s states. Accordingly their calculated enhancement of the excitation probabilities is many orders of magnitude larger than our results. They did not explicitly study the dependence on the photon frequency too. In Ref. [20] no explicit probabilities for the nuclear excitation are given since the more complicated process with the initial hole creation and only relative yields as compared to nuclear γ-ray absorption and Coulomb excitation are considered.

B. $^{161}_{66}$Dy

The second excited state of $^{161}_{66}$Dy with a half-life of $T_{\frac{1}{2}} = 0.83$ ns [30] can be reached from the ground state during an electronic $M1$ transition from the $2s\frac{1}{2}$ to the $1s\frac{1}{2}$ state (Table I). In Fig. 4(a) the dependence of the excitation probability on the laser intensity is examined. The photon energy is adjusted to a resonant two-photon process to get the maximum enhancement. Much higher laser intensities can be used as compared to the uranium case because the participating electrons are much more tightly bound (cf. Fig 1). The lower multipolarity and the electronic transition in the inner shells lead to a much stronger interaction between the electrons and the nucleus and with that to a higher excitation probability. The larger energy difference between the nuclear and the electronic energy differences as compared to the uranium case, however, leads not to a corresponding increase of the excitation probability as expected from the squared interaction energy (Table I). Taking into account the reduction of the widths of the final electron states with increasing laser intensity in the procedure of Sec. IV we notice a distinct enhancement of the excitation probability. We get an amplification of about 4 at an energy already one order below the ionization threshold of the $2s\frac{1}{2}$ state beyond which the probability decreases quickly. The amplification is limited by the large width of the final electron state.

The two-photon resonance energy depends on the intensity through the Stark shift of the electronic levels and decreases from 455.13 eV at no laser application to 425.42 eV at a laser intensity of 1.25×10^{21} W cm^{-2} [Fig. 5(a)]. Additionally to the distinctly visible two-photon resonance the

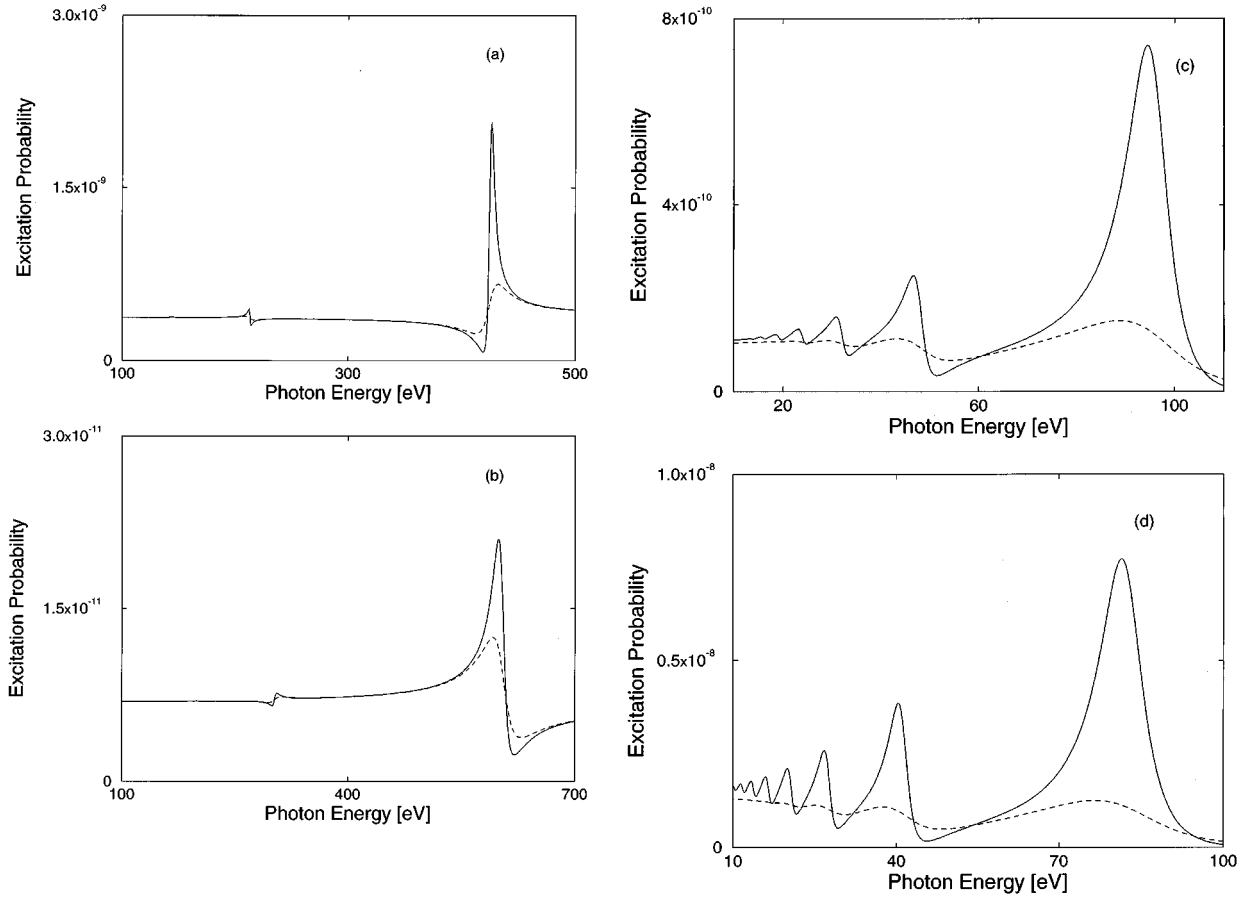

FIG. 5. Excitation probability as a function of the photon energy. Calculation with (solid line) and without (dashed line) reduction of the final state width by the laser induced ionization. Excitation of the (a) $\frac{7}{2}^+$ state in $^{161}_{66}$Dy with a $2s\frac{1}{2} \rightarrow 1s\frac{1}{2}$ electronic $M1$ transition at a laser intensity of 1.25×10^{21} W cm^{-2}, (b) $\frac{5}{2}^-$ state in $^{189}_{76}$Os with a $3s\frac{1}{2} \rightarrow 1s\frac{1}{2}$ electronic $M1$ transition at a laser intensity of 0.75×10^{20} W cm^{-2}, (c) $\frac{1}{2}^+$ state in $^{193}_{77}$Ir with a $3s\frac{1}{2} \rightarrow 1s\frac{1}{2}$ electronic $M1$ transition at a laser intensity of 1.5×10^{20} W cm^{-2}, (d) $\frac{1}{2}^+$ state in $^{197}_{79}$Au with a $3s\frac{1}{2} \rightarrow 1s\frac{1}{2}$ electronic $M1$ transition at a laser intensity of 1.5×10^{20} W cm^{-2}.

four- and six-photon resonances can be seen at a half and a third of this energy with much smaller amplitudes mainly determined by the value of the Bessel functions J_2 and J_3.

C. $^{189}_{76}$Os

The excitation of the $\frac{5}{2}^-$ state ($T\frac{1}{2}=1.62$ ns [31]) from the ground state occurs also during a magnetic dipole transition (Table I). Instead of an initial $2s\frac{1}{2}$ as in the dysprosium case we have a $3s\frac{1}{2}$ state and only smaller laser intensities can be used before ionization effects become noticeable. The dependence of the excitation probability on the laser intensity in Fig. 4(b) shows that the laser-induced reduction of the final-state width is important to achieve an increase of the excitation probability. In Fig. 5(b) we look at the dependence of the excitation probability on the photon energy for a laser intensity of 0.75×10^{20} W cm^{-2}. The behavior is similar to the $^{161}_{66}$Dy case. The resonance energy of 604.36 eV at this laser intensity is even higher than in the case of dysprosium. The four-photon resonance can be detected at half of this energy.

D. $^{193}_{77}$Ir

The excitation of $\frac{1}{2}^+$ state ($T\frac{1}{2}=6.09$ ns [32]) in $^{193}_{77}$Ir also proceeds during an electronic $3s\frac{1}{2} \rightarrow 1s\frac{1}{2}$ transition (Table I). The main differences to the case of osmium are the smaller reduced transition probability for the nuclear excitation and the smaller photon energy neccessary for a two-photon resonance. In Fig. 4(c) we look again at the dependence of the excitation probability on the laser intensity. The reduction of the width of the final electronic state is again the important ingredient to obtain an increase of the probability slightly below the ionization threshold of the initial electron state. The dependence of the probability on the photon energy at a laser intensity of 10^{20} W cm^{-2} [Fig. 5(c)] shows clearly the two-photon resonance. It is shifted by the Stark effect from 61.5 eV in the laser-free case to the higher 95.7 eV at this high intensity. The four-photon, six-photon, and higher photon resonances can also be seen. They are very prominent due to the small values in the argument of the Bessel functions.

E. $^{197}_{79}$Au

The excitation of the $\frac{1}{2}^+$ state ($T\frac{1}{2}=1.91$ ns [33]) resembles very much in its characteristics the excitation of iridium (Table I). The reduced transition probability is larger and the two-photon resonance energy is smaller so that the overall excitation probability is about a magnitude larger. We calculate a probability of 0.129×10^{-8} for the excitation of

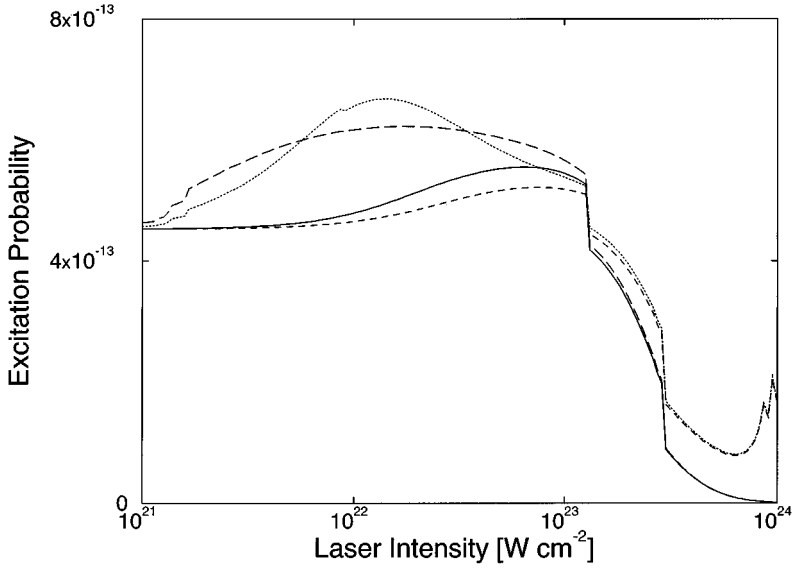

FIG. 6. Excitation probability of the $\frac{7}{2}^-$ state in the $^{237}_{93}$Np nucleus for the $2p\frac{3}{2} \rightarrow 1s\frac{1}{2}$ electronic $E1$ transition as a function of the laser intensity. The photon energy is fixed to the energy of a resonant two-photon process in the $M = \pm\frac{3}{2} \rightarrow \pm\frac{1}{2}$ (solid and long-dashed lines) and $M = \pm\frac{1}{2} \rightarrow \pm\frac{1}{2}$ (short-dashed and dotted lines) transition. Calculation with (long-dashed and dotted lines) and without (solid and short-dashed lines) reduction of the final state width by the laser induced ionization.

the $\frac{1}{2}^+$ state without the application of the laser. The use of the long-wavelength limit in the calculation of the electronic matrix elements would lead to a probability of 0.124×10^{-8}. This is a change of only 4%, not very significant at the current level of theoretical precision. In Fig. 4(d) the dependence of the excitation probability on the laser intensity is studied. As expected, the shape of the excitation function is very similar to the case of $^{193}_{77}$Ir. When the ionization thresholds for the electrons in the $n=3$ shell are reached the probability drops off quickly. The dependence of the excitation probability on the photon energy for a laser intensity of 1.5×10^{20} W cm^{-2} shows again clearly the multiphoton resonances when the reduction of the widths of the final states is taken into account [Fig. 5(d)]. The highest maximum is reached at an energy of 82.4 eV for the two-photon process Stark shifted from 37.59 eV at vanishing laser intensity.

The other maxima can be found at $\frac{1}{2}$, $\frac{1}{3}$, $\frac{1}{4}$, ..., of this energy. The many photon resonances can only be seen so clearly because the Stark shift of the initial state and the photon frequency are of the same order of magnitude: the argument of the Bessel function takes on the large value 0.544 for the two-photon resonance energy. Unfortunately, the photon energies, where we get the strong variation in the excitation probability, seem to be still too high to be reached in an experiment with current available lasers.

F. $^{237}_{93}$Np

The excitation of the $\frac{7}{2}^-$ state in $^{237}_{93}$Np with a half-life of $T_{\frac{1}{2}}=80$ ps [34] from the ground state is made possible by an $E1$ transition (Table I). The initial electron level is now a $2p\frac{3}{2}$ state. The electronic transition matrix elements have to be calculated without using the long-wavelength approximation because of the high excitation energy. We calculate a probability of 0.45×10^{-12} in the case of no laser perturbation. The use of the long-wavelength approximation would reduce this value by 26.6%, a rather large amount. The excitation probability does not increase much with increasing laser intensity (Fig. 6). When the ionization threshold for the initial electronic states is reached we again observe the decrease. The effect of the width reduction by the ionization can be clearly seen but it is not very pronounced. Because the electric field of the laser acts differently on the $2p\frac{3}{2}$ substates the photon energy can be adjusted to a resonant two-photon process either for the $|M|=\frac{3}{2}$ or $\frac{1}{2}$ levels of the initial electron states.

This can also be seen in Fig. 7 where the dependence of the excitation probability on the photon energy is shown for a laser intensity of 7.5×10^{22} W cm^{-2}. The contributions with different $|M|$ in the initial electron state to the total probability show a clearly different behavior. The two-photon resonances are shifted by the Stark effect to different energies and have a different energy dependence. The states with $M=\pm\frac{1}{2}$ are much stronger affected by the Stark effect than the $\pm\frac{3}{2}$ states so that the four-photon resonance in their contribution is also visible. At a laser intensity of 7.5×10^{22} W cm^{-2} the resonances are already quite broad and the ionization induced reduction of the electronic widths has only a small effect. The photon energies for a resonant excitation are considerably larger than the energies that can be reached with current lasers.

G. $^{229}_{90}$Th

Contrary to the examples before we will now study the deexcitation of a nucleus by laser-assisted internal conversion with a bound final electron state. The nucleus $^{229}_{90}$Th has a first excited $\frac{3}{2}^+$ state at an uniquely small energy of (3.5 ± 1.0) eV [35]. The half-life of this state which is populated in the α decay of $^{233}_{92}$U has been estimated to be in the order of 45 h [35] for a M1 radiative transition to the $\frac{5}{2}^+$ ground state ($T_{\frac{1}{2}}=7340$ yr) [36]. A decay of the $\frac{3}{2}^+$ state through internal conversion with an unbound electron in the final state is not possible because the excitation energy is smaller than the binding energy of the most loosely bound electrons in $^{229}_{90}$Th. A resonant or discrete internal conversion with a bound electron in the final state can only be achieved by applying a laser field with the appropriate frequency. This process would lead to a drastic acceleration of the nuclear decay [17].

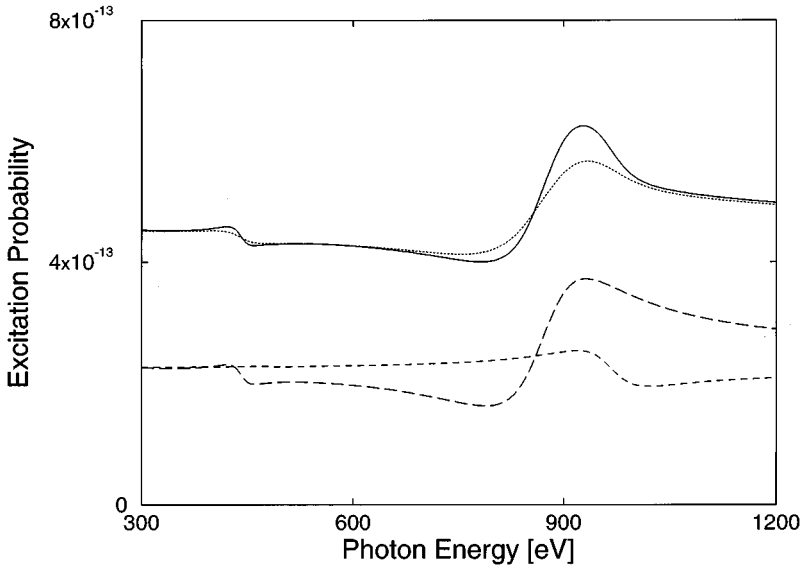

FIG. 7. Excitation probability of the $\frac{7}{2}^-$ state in the $^{237}_{93}$Np nucleus for the $2p\frac{3}{2} \rightarrow 1s\frac{1}{2}$ electronic $E1$ transition as a function of the photon energy with a laser intensity of 7.5×10^{22} W cm^{-2}. Total probability with (solid line) and without (dotted line) reduction of the final state width by the laser induced ionization; contribution of the $M=\pm\frac{1}{2}\rightarrow\pm\frac{1}{2}$ (long-dotted line) and $M=\pm\frac{3}{2}\rightarrow\pm\frac{1}{2}$ (short-dashed line) transition to the probability (calculated with the width reduction).

The electronic binding energies for the electronic states in $^{229}_{90}$Th are taken from Ref. [27] for the $7s\frac{1}{2}$ and lower states, and from Ref. [17] for the upper levels. The energy difference of 3.713 eV between the $7s\frac{1}{2}$ and $8s\frac{1}{2}$ states is very close to the 3.5-eV nuclear decay energy. This electronic transition in a M1 excitation needs only a photon energy of 0.1065 eV in a resonant two-photon process. It is the most favorite candidate for the laser-assisted internal conversion. The experimental signal for the excitation could be the photon emission during the decay of the excited $8s\frac{1}{2}$ state via the $7p\frac{1}{2}$ and $7p\frac{3}{2}$ states.

In Ref. [17] the $8p\frac{1}{2}$ state is chosen as the final electronic state with the excitation to the $8s\frac{1}{2}$ state as intermediate state and the absorption of a single laser photon in the second step. This corresponds to a first-order process in the laser interaction whereas we consider a second-order process. Taking the $8p\frac{1}{2}$ states as final states there is no $M1$ transition possible without the laser field; only the one-photon coupling from the laser with the $8s\frac{1}{2}$ states gives a finite transition probability. The necessary photon energy of 0.712 eV is much higher than in the second-order process. An $8p\frac{1}{2}$ electron will escape from the atom at smaller laser intensities than an $8s\frac{1}{2}$ electron limiting the useful intensities. The probability increases in the first-order process only linearly with the intensity whereas in the second-order process we can expect to observe a quadratic increase.

For the calculation of the $M1$ transition probability we can adopt the theoretical description used before with slight modifications. The width of the initial and final electron states depends on the laser induced ionization. There is no contribution to the width of the initial state from spontaneous transitions because all electron levels with lower energy are occupied. The radiative width of the $8s\frac{1}{2}$ state is calculated from the transitions to the $7p\frac{1}{2}$ and $7p\frac{3}{2}$ levels to be 37.1 neV. Without the application of the laser we get a very small probability of 0.47×10^{-12} for the excitation of the $8s\frac{1}{2}$ electron state during the decay of the $\frac{3}{2}^+$ state.

In Fig. 8 the strong dependence of the excitation probability on the laser intensity is shown where the photon energy is adjusted to the two-photon and four-photon resonances, respectively. We obtain a strong increase with the intensity until the ionization thresholds of the electron states are reached and the probability decreases rapidly. The observed enhancement is enabled by the small width of the final electron state. With a Stark shift of 0.5 meV at a laser intensity of 10^8 W cm^{-2} and the small width of the final electron state we calculate in the approximation of Eq. (3.26) an amplification factor of $\mathscr{A}_2=4.5\times10^7$ independent of the photon energy. This compares well with the value of 2.5×10^7 in the full calculation at a nuclear excitation energy of 3.5 eV. The absolute value of the deexcitation probability depends, of course, on the photon energy. This can be seen in Fig. 8 by comparing the results for different nuclear excitation energies. For large photon energies the theoretical description of the process is no longer valid as the adiabaticity criterion is not fulfilled. The amplification factor explicitly shows the increase of the excitation probability with the square of the laser intensity through the dependence on the Stark shift in the case of the two-photon resonance. In the four-photon process we get an increase with the fourth power of the intensity but at comparatively smaller excitation probabilities.

VI. SUMMARY AND CONCLUSIONS

We studied the excitation of nuclear states by transitions of bound electrons in heavy atoms in the presence of a strong laser field. First-order time-dependent perturbation theory was used for the calculation of the excitation probability including general electric and magnetic multipolarities and considering relativistic retardation effects in the interaction. The time-dependent field dressed electron states were described in an adiabatic approximation which is valid under the condition of a small laser frequency as compared to the electronic transition energies and not too large laser intensities. This method improves the description for many-electron atoms, where the strength of the electron-laser coupling is much smaller than the splitting of states with equal principal quantum number as compared to hydrogenlike atoms. The ionization of the atom for increasing laser intensities was considered in a rough approximation. The limitation of the

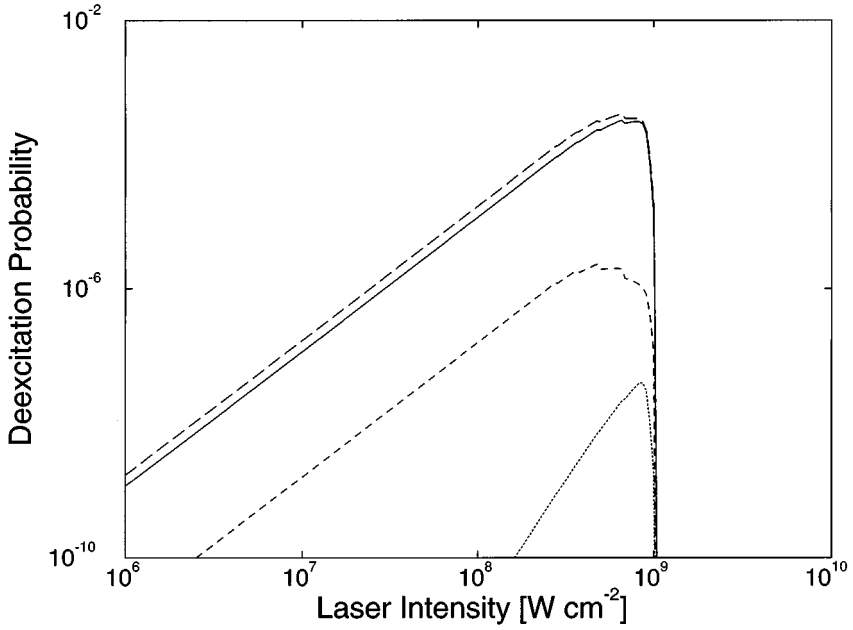

FIG. 8. Deexcitation probability of the $\frac{3}{2}^+$ state in the $^{229}_{90}$Th nucleus for the $7s\frac{1}{2} \rightarrow 8s\frac{1}{2}$ electronic $M1$ transition as a function of the laser intensity. The photon energy is fixed to the energy of a resonant two-photon process for a nuclear excitation energy of 3.5 eV (solid line), 4.0 eV (long-dashed line), and 3.0 eV (short-dashed line) or the resonant four-photon process for an energy of 3.5 eV (dotted line).

laser intensity in the theoretical description is not a problem due to the set in of ionization. The adiabaticity condition is often well met for the envisaged excitation processes.

The laser field gives rise to a characteristic time dependence and coupling of the electronic states determined by the second order Stark effect. This is different from the hydrogenlike atoms where the first order Stark effect dominates. The theoretical description allows an easy application of selection rules for simultaneous electronic and nuclear transitions. Even if the nuclear transition multipolarity does not match the possible multipolarity of the unperturbed electronic transition, there may be a finite excitation probability from the coupling to other electronic states. A resonant increase of the excitation probability can be achieved by adjusting a multiple of the photon energy to the difference of the electronic and nuclear energy differences. These energy differences should match as close as possible to get small laser frequencies and a strong enhancement. The largest increase is expected for large laser intensities sufficiently below the ionization threshold of the participating electron states. The ionization of the atom limits the attainable enhancement of the process by restricting the useful laser intensities. The widths of the states have to be very small. For this, the laser-induced ionization of the atom can contribute by reducing the widths of the final electronic states. The exact values of the ionization thresholds of the electron states calculated in a nonrelativistic approximation can change appreciably in a more detailed calculation. The actual ionization threshold may be larger than the value suggested by the simple barrier penetration model. In Ref. [37] it was demonstrated in a nonrelativistic calculation for the hydrogen atom that a laser field can keep the electron in the atom at very high intensities. The question of the ionization rates in an intense laser field needs certainly more work to be fully understood. The electronic transition matrix elements are very sensitive to the inner structure of the wave functions. A really quantitative calculation will have to use a relativistic description of the electron states. An improvement of the theory is of course possible. More complicated electronic wave functions can be used taking into account relativistic and many-body effects. The adiabatic approximation for the time-dependence of the electron states could be replaced by a full Floquet calculation [38] reducing the limitations in laser frequency and intensity. The ionization process and its effects on the electronic states and their lifetimes should be treated in more detail.

The excitation of the low-lying states in the investigated nuclei exhibits some typical features of the laser-assisted excitation process. The calculated probabilities are very small in the case of the $\frac{1}{2}^+$ isomeric state of $^{235}_{92}$U even in the resonant excitation due to the weak electric octupole coupling of the outer electrons with the nucleus. The excitation with the help of electronic transition with multipolarity $L=1$ in the inner shells of, e.g., $^{161}_{66}$Dy, $^{189}_{76}$Os, and $^{197}_{79}$Au are much more probable. An actual experimental observation of the nuclear excitation by laser-assisted electronic transitions seems to be very difficult in the $^{235}_{92}$U case. The needed photon energies for a resonant excitation of the nuclei $^{197}_{79}$Au, $^{193}_{77}$Ir, and especially $^{161}_{66}$Dy, $^{189}_{76}$Os, and $^{237}_{93}$Np seem to be too large for current lasers. The deexcitation of the $\frac{3}{2}^+$ state in $^{229}_{90}$Th in a resonant internal conversion is really worth an experimental study. In comparison with experiments we have however to bear in mind the idealized description of a single atom. In real situations especially the outer electron states will depend on the atomic environment. This is clearly seen from the influence of the chemical composition on the lifetime of the isomeric state in $^{235}_{92}$U which decays by internal conversion [39,40]. The ionization of the atom in the strong laser field will also have an effect on the binding energies of the remaining electrons. In favorable cases, this can lead to a shift of the resonance energies to experimental accessible regions.

An investigation of other nuclei is worthwhile. Low multipolarities and transitions between inner electron states would be advantageous to get a strong nuclear-electron cou-

pling. The necessary photon energy to achieve resonance has to be within reach experimentally. The chance of finding out favorable experimental conditions depends mainly on the precise knowledge of the nuclear and electronic energies. The actual number of experimentally accessible cases will be quite small.

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