Intense few-cycle hard-UV-pulse-induced internal conversion processes

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The internal conversion coefficient for bound-free electron transition of originally energetically forbidden internal conversion processes induced by intense, few-cycle UV laser pulse of Gaussian shape in the case of isomers $^{107}Ag^m$ (K shell, $E3$, 25.47 keV), $^{90}Nb^m$ (L_2 shell, $M2+E3$, 2.3 keV), $^{183}W^{m1}$(M_5 shell, $E2$, 1.79 keV), $^{183}W^{m2}$ (N_1 shell, $E1$, 548 eV), and $^{188}Re^m$ (M_2 shell, $M3+E4$, 2.63 keV), and $^{235}U^m$ (O_4 and O_5 shells, $E3$, 73.5 eV) is determined numerically. Experimental conditions and possibilities of the laser-induced internal conversion process of $^{183}W^{m2}$ from the N_1 shell are discussed in more detail.

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In the last two and a half decades the electron-nucleus-laser combined processes, such as the laser-beam-modified internal conversion (IC) [1] and electronic-bridge processes [2], were systematically investigated [3] and their description could be traced back to laser-field-induced modification of the electromagnetic interaction between the electron and the nucleons [4,5]. The recently created intense pulses of length of a few cycles and of photon energy in the hard-UV and soft-x-ray range [6,7] are expected to measurably modify the electron-nucleus combined processes. In a recent article [8] the superintense, few-cycle x-ray pulse-induced IC process was discussed in more detail and as a numerical example the superintense, few-cycle, soft-x-ray laser-induced IC process in the case of ^{99m}Tc was investigated. The few-cycle x-ray pulse-induced IC coefficient (ICC) of the energetically forbidden IC process that starts from the $2p_{3/2}$ electron shell was given in the case of a pulse of Gaussian shape. In this brief report we discuss similar processes of the isomers $^{107}Ag^m$ (K shell, $E3$, 25.47 keV), $^{90}Nb^m$ (L_2 shell, $M2+E3$, 2.3 keV), $^{183}W^{m1}$ (M_5 shell, $E2$, 1.79 keV), $^{183}W^{m2}$ (N_1 shell, $E1$, 548 eV), $^{188}Re^m$ (M_2 shell, $M3+E4$, 2.63 keV), and $^{235}U^m$ (O_4 and O_5 shells, $E3$, 73.5 eV).

It was shown in [8] that the effect of a few-cycle pulse of peak magnitude E_0 of the electric field strength on electron-nucleus interaction can be equivalently replaced by transforming the electron space coordinates $\vec{x}$ to $\vec{x}_B$ as

$$\vec{x}_B = \vec{x} + \xi\,\vec{\varepsilon}, \tag{1}$$

(Henneberger transformation [9,10]), where $\vec{\varepsilon}$ is the unit vector of polarization of the laser field,

$$\xi(k_0x_0,\phi) = \xi_0 f(k_0x_0,T)\cos(k_0x_0+\phi), \tag{2}$$

with

$$\xi_0 = \frac{-e}{\kappa}Z_0. \tag{3}$$

The notation is the following: $k_0=\omega_0/c$, ω_0 is the carrier angular frequency; c is the velocity of light; $x_0=ct$; $T=\omega_0\tau$, where τ is the pulse length; ϕ is the carrier-envelope phase; $Z_0=E_0/\omega_0^2$, $f(k_0x_0,T)$ describes the pulse form; κ is the rest mass of the electron; and e is the elementary charge. The quantity Z_0 is the peak magnitude of

$$\vec{Z}(t) = -\frac{1}{c}\int \vec{A}_{\rm cl}(t)\,dt, \tag{4}$$

where $\vec{A}_{\rm cl}(t)$ is the vector potential that describes the classical radiation (laser) field. Similarly to (1), as it was done in [8], any function $F(\vec{x})$, such as the interaction potentials, must be transformed as $F(\vec{x}+\xi\,\vec{\varepsilon})$.

The Henneberger transformation (1) is a transformation to an oscillating coordinate system, where generally a useful simplification of the laser-assisted problems occurs only if the energy of a photon of the applied field is substantially greater than the transition energy of the bound system. However, this commonly known restriction is the consequence of the application of the stationary Floquet approach only [11] that has not been applied in our calculation [4,8]. (The condition of applicability of the stationary Floquet approach in a nonstationary problem is that the pulse rise time is not exceedingly short; that is, the pulse rise time should be long in comparison to the light period [11]. Therefore the stationary Floquet approach and its consequences cannot be applied in the case of few-cycle pulses.) The Henneberger transformation is based on the dipole approximation [see (4)]. Recent investigation of the tunneling limit of strong field ionization [12] raises the question of the applicability of the dipole approximation in tunneling theories and at extremely large intensities in general. This is a serious question that imposes intensity limits on the validity and applicability of this work. The intensity limit, however, is higher than the intensity limit originating from experimental limitations. Namely, in experiments aimed at investigating laser-ignited IC processes, radioactive sample material is used and consequently it is desirable to apply peak intensities below the threshold intensity of damage, and up to these peak intensities the applicability of dipole approximation cannot be questioned.

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TABLE I. Computed values of $\Sigma_l \alpha^{\rm pulse}_{l,L,0}$ (in cm^2/W units). In the table the transition energies ($\hbar\omega_{\alpha\beta}$), the multipolarity of the nuclear electric transition with the symbols of the electronic shells that take part in the IC process, the binding energies (E_B) of the electron on the shell in question [14], the energy defect of the shell ($\Delta E = \hbar\omega_{\alpha\beta} - E_B$), and the total laser-free IC coefficients ($\alpha_{\rm tot}$) [see Refs. [15] (a) and [16] (b)] of the transitions investigated are listed.

Isomer	$\hbar\omega_{\alpha\beta}$ (keV)	EL/shell	E_B (keV)	$-\Delta E$ (eV)	$\Sigma_l \alpha^{\rm pulse}_{l,L,0}$(cm^2/W)	$\alpha_{\rm tot}$
^{105}Ag^m	25.47	E3/K	25.514	44	2.6×10^{-16}	3.6×10^4(a)
^{90}Nb^m	2.3	$E3/L_2$	2.368	68	1.2×10^{-13}	1.1×10^{10}(b)
$^{183}W^{m1}$	1.79	$E2/M_5$	1.807	17	3.9×10^{-13}	6.5×10^7(b)
$^{183}W^{m2}$	0.548	$E1/N_1$	0.595	47	6.9×10^{-18}	2240(b)
^{188}Re^m	2.63	$E4/M_2$	2.682	52	3.4×10^{-16}	1.1×10^{15}(b)
^{235}U^m	0.0768	$E3/O_5$	0.096	20	0.038	3.7×10^{20}(b)
^{235}U^m	0.0768	$E3/O_4$	0.103	26	0.02	3.7×10^{20}(b)

It is an important conclusion of our two-decade-long investigation of laser-assisted IC processes that measurable effect can be expected only in the case of originally energetically forbidden IC processes where the laser helps to fulfill energy conservation law by injecting the energy needed to start the process [1,4]. Our former laser-ignited IC coefficient calculation [4] carried out in the plane-wave limit indicates that from the point of view of laser radiation this laser-assisted process is rather similar to a multiphoton ionization process where the binding energy has to be replaced by the energy defect of the originally energetically forbidden IC process. It was found [13] that only the one laser-photon-ignited process is accountable. This recognition was confirmed by our next laser-ignited IC coefficient calculation [8], where the formerly applied method was extended to be valid in the case of extremely short pulses, too. Although the isomers considered here have energy of the isomeric transition ranging from 25.5 keV to 73.5 eV, the really essential parameter, that is, the carrier angular frequency of the applied laser, is determined by the energy defect of the process, and in the case of the preceding isomers it lies in the UV range from 17 to 68 eV.

In the following, we investigate the UV-pulse-ignited IC process of such metastable state of the nucleus that decays mainly by an electric multipole decay mode of order L (denoted as EL) and for which the current-current interaction between the nucleus and the electron can be neglected. The angular momentum change in the electronic transition is l. We take a Gauss function for the envelope function, that is,

$$\xi(k_0 x_0, \phi) = \xi_0 e^{-(\frac{k_0 x_0}{T})^2} \cos(k_0 x_0 + \phi), \tag{5}$$

where ξ_0 is connected with the peak intensity I of the intense UV pulse by the formula $\xi_0 = [4\pi\alpha_f \hbar/(\kappa^2 c^4 k_0^4)]^{1/2} I^{1/2}$ where α_f is the fine structure constant.

Now we briefly summarize our former results [8] that can be applied in the case of UV-pulse-induced IC processes which are originally energetically forbidden. The UV-pulse-induced ICC of a bound-free electronic transition,

$$\alpha_{l,L} = \alpha^{\rm pulse}_{l,L,0} \delta^4 \frac{\tau}{\tau_{ir}} \psi(\phi, T, \delta) I, \tag{6}$$

valid in the case of a Gaussian laser pulse, where $\delta = |\Delta|/k_0$, $\Delta = \Delta E/(\hbar c)$. ΔE is the energy defect of the system, that is, $\Delta E = E_\alpha - E_\beta + E_1$, with E_α and E_β standing for the energy eigenvalues of the initial and final nuclear states, and E_1 denoting the energy eigenvalue of the initial electron state. The energetically forbidden IC processes are investigated near the threshold, that is, the carrier angular frequency $\omega_0 \sim |\Delta E|/\hbar$. For the detailed formula of $\alpha^{\rm pulse}_{l,L,0}$, see Eq. (42) of [8],

$$\psi(\phi, T, \delta) = T \int_0^\infty |G(\beta, T, \phi, \delta)|^2 d\beta, \tag{7}$$

with

$$G(\beta, T, \phi, \delta) = \frac{\pi^{1/2}}{2} \left[e^{-i\phi} e^{-(\frac{(\beta+\delta-1)T}{2})^2} + e^{i\phi} e^{-(\frac{(\beta+\delta+1)T}{2})^2} \right], \tag{8}$$

where $\beta = E_2/(k_0 \hbar c)$ and E_2 denotes the energy eigenvalue of the final electron state. The plane-wave limit determines $\tau_{ir} = 0.627\tau$ (see Eq. (46) of [8]). The ϕ and T dependence of $\alpha_{l,L}$ is illustrated by the plot of $\psi(\phi, T, \delta = 1)$ ($k_0 = |\Delta|$) in Fig. 1 of [8]. The δ ($k_0 = |\Delta|/\delta$) and the T dependence of $\delta^4 \psi$, which is the same as the δ and the T dependence of $\alpha_{l,L}$, is given at $\phi = n\pi$ in Fig. 2 of [8].

The results of our numerical calculation are collected in Table I. The atomic and nuclear data are taken from [14], and the total laser-free ICCs can be found in [15] and [16]. The table contains the sum of the accountable leading terms that contribute to $\Sigma_l \alpha^{\rm pulse}_{l,L,0}$. If we take into account the method of preparation of the desired number nuclei of isomeric state in the sample, then the most promising one is the laser-induced IC process of $^{183}W^{m2}(E1, 548\ {\rm eV})$ from the N_1 shell ($|\Delta E| = 47$ eV).

The $^{183}W^{m2}(11/2^+)$ isomer has half-life $\tau_\alpha = 5.2$ s and its energy is $E_\alpha = 309.493$ keV. It decays with an $E1$ transition to the $9/2^-$ state of energy $E_\beta = 308.9455$ keV; thus the energy of the transition is $\hbar\omega_{\alpha\beta} = 548$ eV. The isomeric state is generated in a β^- decay of the ground state of ^{183}Ta of half-life $\tau_{\rm Ta,183} = 5.1$ d. The ^{183}Ta can be generated in a two-step neutron capture process from ^{181}Ta (99.988%). The thermal neutron cross sections of the ^{181}Ta$(n,\gamma)^{182}$Ta and ^{182}Ta$(n,\gamma)^{183}$Ta processes are $\sigma_{12} = 21\ b$ and $\sigma_{23} = 8200\ b$, respectively. Since $\sigma \ll \sigma_{23}$, one can use the secular balance condition

$$\sigma_{23} N_{\rm Ta,182}(t) = \sigma_{12} N_{\rm Ta,181}(t), \tag{9}$$

solving the coupled rate equations, where $N_{\rm Ta,181}(t)$ and $N_{\rm Ta,182}(t)$ are the instantaneous numbers of ^{181}Ta and ^{182}Ta

nuclei during the neutron irradiation process. The instantaneous number of ^{183}Ta nuclei,

$$N_{\mathrm{Ta},183}(t) = N_0 \frac{\sigma_{12}\Phi(1 - e^{-(\sigma_{12}\Phi - \lambda_{\mathrm{Ta},183})t})}{\sigma_{12}\Phi - \lambda_{\mathrm{Ta},183}} e^{-\lambda_{\mathrm{Ta},183}t}, \qquad (10)$$

during the neutron irradiation process, where N_0 is the initial number of the ^{181}Ta nuclei, Φ is the neutron flux, t is the time of irradiation, and $\lambda_{\mathrm{Ta},183} = \ln 2/\tau_{\mathrm{Ta},183}$. In obtaining (10), the decay of ^{182}Ta was neglected since its lifetime (114 d) is long enough to do this. If $\sigma_{12}\Phi \gg \lambda_{\mathrm{Ta},183}$, then

$$N_{\mathrm{Ta},183}(t) = N_0(1 - e^{-\sigma_{12}\Phi t})e^{-\lambda_{\mathrm{Ta},183}t}. \qquad (11)$$

With the aid of (11), one can obtain the initial number $N_{\mathrm{Ta},183}(0)$ of ^{183}Ta nuclei in the laser-induced experiment, for example, with $\Phi = 10^{18}$ s^{-1} cm^{-2} and $t = 4.8 \times 10^4$ s, one gets $N_{\mathrm{Ta},183}(0) = 0.59 \times N_0$.

Using again the secular balance condition since $\lambda_{\mathrm{Ta},183} \ll \lambda_\alpha = \ln 2/\tau_\alpha$ the instantaneous number of the isomeric state $^{183}W^{m2}(11/2^+)$,

$$N^m_{W,183}(t) = 0.05 \frac{\tau_\alpha}{\tau_{\mathrm{Ta},183}} N_{\mathrm{Ta},183}(0) e^{-\lambda_{\mathrm{Ta},183}t}, \qquad (12)$$

in the laser-irradiated sample. The factor 0.05 in (12) is present since the 5% of the β decay of ^{183}Ta results $^{183}W^{m2}(11/2^+)$. In the case calculated above $N^m_{W,183}(t) = 3.5 \times 10^{-7} N_0 e^{-\lambda_{\mathrm{Ta},183}t}$.

The laser-induced number of vacancies N_v in the N_1 shell, which will be considered as signal of the measurement, can be written as

$$N_v = \sum_{k=1}^{N_p} N^m_{W,183}(t_k) \frac{\Sigma_l \alpha_{l,L}\tau_{ir}}{\alpha_{\mathrm{tot}}\tau_\alpha}. \qquad (13)$$

Here N_p is the number of the pulses. If we take a laser of repetition rate $r = 10$ s^{-1} and the total time of the measurement $T_{\mathrm{tot}} = 86\,400$ s, the carrier angular frequency at the optimum value of $\delta = 2.5$ ($\omega_0 = c|\Delta|/\delta$, $\nu_0 = \omega_0/(2\pi) = 4.5 \times 10^{15}$ s^{-1}, $\hbar\omega_0 = 18.8$ eV, $\tau = 0.22$ fs) and in the case of a one-cycle pulse of $\phi = n\pi$ one gets $N_v = 2.16 \times 10^{-37} N_0 I$, with I measured in W/cm^2 units. It is a technical challenge to reach and maintain the desired number of ^{183}Ta isotopes in the laser beam of necessary intensity. It may be advantageous to increase the length of the sample without focusing the beam since $N_0 I$ is proportional to the length of the sample irradiated. Taking, for example, Ta_2O_5 as sample material, which is optically transmitting, the intensity may be lowered below the threshold of damage that makes it possible to hold the sample material in the beam during the total time of the measurement (e.g., taking $N_0 = 6 \times 10^{23}$, we get $N_v = 1.30 \times 10^{-13} I$).

The number of vacancies N_{back} in the N_1 shell created by other IC decay processes in the sample during the measurement in one pulse of time τ_m, which will be considered as background of the measurement, can be estimated as

$$N_{\mathrm{back}} = \lambda_{\mathrm{Ta},183} \sum_{k=1}^{N_p} N_{\mathrm{Ta},183}(t_k) W_{\mathrm{av}} \tau_m^2, \qquad (14)$$

where

$$W_{\mathrm{av}} = \sum_j \alpha_{\mathrm{IC},j}(N_1) W_{\gamma,j}. \qquad (15)$$

Here $\alpha_{\mathrm{IC},j}(N_1)$ and $W_{\gamma,j}$ are the ICCs and γ rates of the other transitions of the decay of an excited state of ^{183}W that lead to an N_1 vacant state. W_{av} is hard to determine but it may be estimated from the half-lives of the possible populated levels and it is reasonable to expect that $W_{\mathrm{av}} \lesssim 2 \times 10^{10}$ s^{-1}. If the total time of the experiment is shorter than $\tau_{\mathrm{Ta},183}$ then the signal to background ratio

$$\eta = \frac{0.05}{\ln 2} \frac{\Sigma_l \alpha^{\mathrm{pulse}}_{l,L,0} \delta^4 \psi \tau I}{\alpha_{\mathrm{tot}} W_{\mathrm{av}} \tau_m^2}. \qquad (16)$$

In the situation discussed earlier, $\eta \gtrsim 1.5 \times 10^{-17} I \tau_m^{-2}$, where I and τ_m have to be substituted in W/cm^2 and fs units, respectively. If vacancies of the N_1 shell created by one pulse are traced by the method of light-controlled secondary electron emission spectroscopy [17] and/or ion-charge-state chronoscopy [18] of Auger decay, then τ_m may have fs order of magnitude. Consequently, good signal-to-background ratio may be expected. It is hoped that with the aid of recently available sub-fs, one-cycle pulses in the UV range [7] successful experiments may be achieved.

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