Internal-conversion process in superintense ultrashort x-ray pulses

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The electron-nucleus interaction in a super-intense few-cycle x-ray pulse is investigated. The super-intense few-cycle x-ray pulse-induced internal conversion (IC) process is discussed in detail. The x-ray laser-pulse induced IC coefficient is calculated, and in particular, it is derived in the case of a pulse of Gaussian shape and for a bound-free electron transition. The IC coefficient of the IC process induced by a super-intense few-cycle soft-x-ray laser pulse in the case of the ^{99m}Tc isomer is determined numerically. The results obtained for the IC coefficient show significant carrier angular frequency, carrier-envelope phase, and pulse-length dependencies. The infinite pulse-length limit and experimental aspects are also discussed.

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I. INTRODUCTION

It was long ago recognized [1] that the direct interaction between a nucleus and a laser beam was strongly hindered by the shielding effect of the electron cloud. It was also shown in the case of low-energy nuclear transitions [2] that the rate of an internal conversion process was lowered if an outer electron was removed with the aid of laser radiation. Later it was concluded [3] that the direct laser-nucleus interaction cannot cause measurable change in γ-decay rates. These results focused the interest on the laser beam modified internal conversion (IC) [4] and electronic-bridge processes [5]. Afterward, in a number of works it was stated that laser fields of high enough intensity may cause significant modification in the rate of electron-nucleus-laser combined processes [6]. Finally it was found that all these processes can be traced back to laser-field induced modification of the electromagnetic interaction between the electron and nucleons [7].

The progress made in laser technology aimed at reaching extremely high laser intensities has led to pulses of length of few cycles and of photon energy in the hard UV and soft-x-ray range [8,9] that seem to be appropriate to measurably modify electron-nucleus combined processes. The subfemtosecond x-ray pulses are currently produced by high-harmonic generation combined with spectral filtering and phase matching and their peak intensity is about two to three orders of magnitude higher than that of synchrotron sources, but their duration is six to eight orders of magnitude shorter and thus their use does not seem to increase the feasibility of experimental verification of the laser-induced IC coefficient. However, recently about a four orders of magnitude increase with a decreasing pulse length of the two photonic ionization process was stated [10] in the x-ray regime that indicates a similar phenomenon in the case of laser-assisted electron-nucleus combined processes. The calculations in Ref. [7] were carried out in the plane-wave (infinitely long pulse) limit for the transition probability per unit time of laser-induced IC processes and for IC coefficients of energetically forbidden IC processes. Therefore, we reformulate the problem discussed in Ref. [7] to be valid in the few-cycle case as well.

One aim of this article is to give the transition probability of electron-nucleus-laser combined processes valid also in the case of super-intense few-cycle x-ray pulses. The super-intense few-cycle x-ray pulse-induced IC process is discussed in more detail. Numerically the super-intense few-cycle soft-x-ray laser-induced IC process in the case of ^{99m}Tc is investigated and the few-cycle x-ray pulse-induced IC coefficient of the energetically forbidden IC process that starts from the $2p_{3/2}$ electron shell is given. The infinitely long pulse limit is determined for comparison. Some experimental aspects are also discussed.

II. ELECTRON-NUCLEUS INTERACTION IN A FEW-CYCLE LASER FIELD

It was shown in Ref. [7] that the effect of the laser radiation on electron-nucleus interaction can be equivalently replaced by transforming the electron space coordinates $\vec{x}$ to $\vec{x}_B$ as

$$\vec{x}_B = \vec{x} + \frac{e_e}{\kappa}\vec{Z}, \tag{1}$$

(Henneberger transformation [11,12]) where κ is the rest mass of the electron, $e_e = -e$ is the electronic charge (e is the elementary charge),

$$\vec{Z}(t) = -\frac{1}{c}\int \vec{A}_{\rm cl}(t)dt, \tag{2}$$

where c is the velocity of light, and $\vec{A}_{\rm cl}(t)$ is the vector potential that describes the classical radiation (laser) field. Similarly to Eq. (1), any function $F(\vec{x})$ must be transformed as $F(\vec{x} + \frac{e_e}{\kappa}\vec{Z})$.

In Ref. [7] $\vec{A}_{\rm cl}(t) = \vec{\varepsilon} A_0 \cos(\omega_0 t)$ was used to correspond to an infinitely long pulse of carrier angular frequency ω_0. In the case of a pulse of length τ, the electric field strength of a few-cycle pulse can be well described [13] with

$$\vec{E}(k_0x_0) = E_0\vec{\varepsilon}\,h(k_0x_0/T)\cos(\alpha), \tag{3}$$

where $\vec{\varepsilon}$ is the unit vector of polarization, $h(k_0x_0/T)$ stands for the pulse envelope function, $T = \omega_0\tau$ is the parameter describing the pulse length, and $\alpha(\phi) = k_0x_0 + \phi$ with ϕ as the carrier-envelope phase. (We introduce new variables $k_0 = \omega_0/c$, $x_0 = ct$.) We found that pulses of the form similar to Eq. (3) can be obtained by deriving the vector potential $\vec{A}_{\rm cl}(x_0/c)$ and the electric field strength $\vec{E}(k_0x_0)$ from

$$\vec{Z}(k_0x_0) = Z_0\vec{\varepsilon}\,f(k_0x_0,T)\cos(k_0x_0+\phi), \tag{4}$$

with $Z_0 = E_0/\omega_0^2$ and $f(k_0x_0,T)$ describing the pulse form. As the radii of both the atomic and nuclear states are much smaller than the carrier wavelength of the radiation, the dipole approximation is justified. As a consequence of Eq. (1) and what was said earlier the calculation in Ref. [7] must be repeated with the substitution

$$\vec{x}_B = \vec{x} + \xi\vec{\varepsilon}, \tag{5}$$

where

$$\xi(k_0x_0,\phi) = \xi_0 f(k_0x_0,T)\cos(k_0x_0+\phi), \tag{6}$$

with

$$\xi_0 = \frac{e_e}{\kappa}Z_0. \tag{7}$$

As a result, in D_F, that is the causal (Feynman) Green-function [14] used in the electromagnetic four-vector potential [7], an $\exp[i\xi(k_0x_0,\phi)\vec{\varepsilon}\times\vec{q}\,]$ multiplier appears

$$D_F(x'-y_N) = \frac{-1}{4\pi^3}\int e^{i[\xi(k_0x_0,\phi)]\,\vec{\varepsilon}\cdot\vec{q}}\,\frac{e^{-iq(x-y_N)}}{q^2+i\varepsilon}d^4q. \tag{8}$$

Here the four-vector $q^\mu = \{q_0,\vec{q}\}$, $x^{\mu\prime}$ represents the Henneberger-transformed electron four-coordinate, $(x^{\mu\prime} = \{x_0,\vec{x}_B\})$, and $y_N^\mu = \{y_0,\vec{y}_N\}$ as the four-coordinate of one of the nucleons. Let $J_\mu^{21}(x)$ denote the four-transition current density of the electron in the transition $1\to 2$ and $j_{\beta\alpha}^\mu(y_N)$ the four-transition current density of a nucleon in the $\alpha\to\beta$ transition that are

$$J_\mu^{21}(x) = J_\mu^{21}(\vec{x})e^{ik_{21}x_0}, \tag{9}$$

with $J_\mu^{21}(\vec{x}) = e_e\bar{\psi}_2(\vec{x})\gamma_\mu\psi_1(\vec{x})$ and

$$j_{\beta\alpha}^\mu(y_N) = e^{ik_{\beta\alpha}y_0}j_{\beta\alpha}^\mu(\vec{y}_N). \tag{10}$$

Here γ^μ denotes the γ matrices, ψ_2 and ψ_1 stand for the electron bispinors of energy eigenvalues E_2 and E_1 corresponding to the final and initial electron states of the process, respectively, $k_{21} = (E_2-E_1)/(\hbar c)$, and $k_{\beta\alpha} = (E_\beta - E_\alpha)/(\hbar c)$, where E_β and E_α stand for the energy eigenvalues of the final and initial nuclear states, respectively. The transition amplitude of the process [14] is

$$c_{fi}(x_{02},x_{01}) = -\frac{i}{\hbar c}\sum_N\int_{x_{01}}^{x_{02}}dx_0\int d^3x\int d^4y_N \times J_\mu^{21}(x)D_F(x'-y_N)j_{\beta\alpha}^\mu(y_N). \tag{11}$$

Integrating over y_0 and q_0 and using the expansion of $\exp(iq\xi\cos\vartheta_q) = \sum_{n=0}^\infty i^n(2n+1)P_n(\cos\vartheta_q)j_n(q\xi)$, we obtain

$$\int dq_0\int dy_0 e^{ik_{\beta\alpha}y_0}D_F(x'-y_N) = \sum_n e^{ik_{\beta\alpha}x_0}V_n(\vec{x},\vec{y}_N;\xi(x_0)), \tag{12}$$

where

$$V_n(\vec{x},\vec{y}_N;\xi(x_0)) = \frac{-i^n(2n+1)}{2\pi^2}\int d^3q\,P_n(\cos\vartheta) \times\frac{j_n(\xi q)e^{i\vec{q}\cdot(\vec{x}-\vec{y}_N)}}{k_{\beta\alpha}^2-\vec{q}^{\,2}+i\varepsilon}, \tag{13}$$

and j_n stands for the spherical Bessel function. Carrying out the integration over x_0

$$c_{fi}(x_{02},x_{01}) = \int_{x_{01}}^{x_{02}}K_{fi}(\xi(x_0))e^{i(k_{\beta\alpha}+k_{21})x_0}dx_0, \tag{14}$$

with

$$K_{fi}(\xi(x_0)) = \sum_n K_{fi}^{(n)}(\xi(x_0)), \tag{15}$$

and

$$K_{fi}^{(n)}(\xi(x_0)) = -\frac{i}{\hbar c}\sum_N\int d^3x d^3y_N J_\mu^{21}(\vec{x})j_{\beta\alpha}^\mu(\vec{y}_N) \times V_n(\vec{x},\vec{y}_N;\xi(x_0)). \tag{16}$$

The transition probability

$$P_{\alpha\beta,12} = \sum_f\langle|c_{fi}(-\infty,\infty)|^2\rangle_i, \tag{17}$$

where $\sum_f$ denotes the sum over all the possible final states and $\langle\rangle_i$ denotes the average over all the possible initial states. As a consequence of the large bandwidth of the short pulse the terms that correspond to n-photonic processes cannot be traced out with the aid of an energy-Dirac-delta and so they are mixed in Eq. (15) and consequently in Eq. (14).

III. X-RAY PULSE-INDUCED IC PROCESS

We deal with energetically forbidden IC processes ($|\beta\rangle\neq|\alpha\rangle$, $k_{\beta\alpha}<0$) that are induced by the coherent, super-intense UV soft or hard x-ray radiation. It was found [7] that it can switch on the IC processes more effectively if its carrier angular frequency $\omega_0\sim|\Delta E|/\hbar$, where $\Delta E = E_{\alpha\beta}-|E_1|$ is the energy defect of the system with $E_{\alpha\beta} = E_\alpha - E_\beta$. Therefore in the following, the energetically forbidden IC process is investigated near the threshold.

Expanding the exponent in Eq. (13) and following the train of thought of Ref. [7] we obtain

$$V_n = \sum_{l,L,M}(-8)i^l(-i)^L Y_{lM}(\hat{x})Y_{LM}^*(\hat{y}_N) \times\int_{-1}^1\varphi_{l,L,M}^n(u)du\int_0^\infty I_n(q)dq, \tag{18}$$

where Y_{lM} and Y_{LM} denote the spherical harmonics, $\hat{x}$ and $\hat{y}_N$ represent the unit vectors pointing in the directions $\vec{x}$ and $\vec{y}_N$, respectively, and we introduce

$$\varphi^n_{l,L,M}(u) = N i^n (2n+1) P_n(u) P_l^M(u) P_L^M(u), \tag{19}$$

with

$$N = \sqrt{\frac{(2l+1)(l-M)!}{2(l+M)!}}\sqrt{\frac{(2L+1)(L-M)!}{2(L+M)!}}. \tag{20}$$

P_n is the Legendre polinomial and P_l^M and P_L^M are the associated Legendre-polinomials of the first kind

$$I_n(q) = \frac{j_n(\xi q) j_l(qx) j_L(q y_N) q^2}{k^2_{\alpha\beta} - \vec{q}^{\,2} + i\varepsilon}, \tag{21}$$

where j_n, j_l, and j_L are spherical Bessel functions of order n, l, and L, respectively. The frame of reference was chosen as $\vec{q_z} \parallel \vec{\varepsilon}$ (the laser is polarized in the z direction).

Expanding the expression $P_n(u)P_l^M(u)P_L^M(u)$ into the power series of u [15], it can be shown that it is an even function of u if $n+l+L$ is even. In this case the integration over u can be written as $\int_{-1}^1 du = 2\int_0^1 du$. On the other hand, if $n+l+L$ is odd the integration results in $\int_{-1}^1 P_n P_l^M P_L^M du = 0$, which gives $P_{\alpha\beta,12} = 0$. Therefore we have to evaluate the transition probability of the process for $l+L+n =$ even cases only. However, for $l+L+n =$ even, one can change the integration as $\int_0^\infty dq = 1/2\int_{-\infty}^\infty dq$. Supposing that $\xi + y_N \ll x$, one can carry out the integration over q with the aid of contour integration techniques [7]. It results

$$\int_0^\infty I_n(q)dq = \frac{\pi}{2} i k_{\alpha\beta} j_n(-\xi k_{\alpha\beta}) j_L(k_{\alpha\beta} y_N) h_l^{(1)}(k_{\alpha\beta}x), \tag{22}$$

where $h_l^{(1)}(k_{\alpha\beta}x)$ is the spherical Hankel function of the first kind of order l and $k_{\alpha\beta} = -k_{\beta\alpha}$. Collecting everything we obtain

$$V_n = \frac{\pi}{2} i k_{\alpha\beta} \sum_{l,L,M} (-8) i^l (-i)^L Y_{lM}(\hat{x}) Y^*_{LM}(\hat{y}_N) \times j_L(k_{\alpha\beta} y_N) h_l^{(1)}(k_{\alpha\beta}x) Q^n_{l,L,M}\, j_n(-\xi k_{\alpha\beta}), \tag{23}$$

where

$$Q^n_{l,L,M} = \int_{-1}^1 \varphi^n_{l,L,M}(u)du. \tag{24}$$

In the following we investigate the x-ray pulse-ignited IC process of a certain metastable state of the nucleus that decays mainly by an electric multipole decay mode of order L (denoted as EL) and for this reason the current-current interaction between the nucleus and the electron can be neglected. We approximate $j_L(k_{\alpha\beta}y_N) \simeq (k_{\alpha\beta}y_N)^L/(2L+1)!!$ [16] in Eq. (23), which makes it possible to use the multipole transition operator of the nucleus that is defined as $\mathcal{M}(EL,M) = \sum_N e_N y_N^L Y_{LM}(\hat{y}_N)$, where e_N is the charge of the Nth nucleon. Its transition matrix element can be written as

$$\langle I_i, M_i|\mathcal{M}(EL,M)|I_f, M_f\rangle = (-1)^{I_i-M_i}\langle I_i\|\mathcal{M}(EL)\|I_f\rangle \times \begin{pmatrix} I_i & L & I_f \\ -M_i & M & M_f \end{pmatrix}, \tag{25}$$

where $\langle I_i\|\mathcal{M}(EL)\|I_f\rangle$ is the reduced matrix element of $\mathcal{M}(EL,M)$, I_i and I_f are the angular momentum quantum numbers, M_i and M_f denote the magnetic quantum numbers of the initial and final nuclear states, respectively, and the usual notation of $3j$ symbols is applied.

After summing up all the magnetic quantum numbers of the final states (m_f, M_f), averaging all the initial ones (m_i, M_i)—where m_i and m_f are the magnetic quantum numbers of the initial and final electronic states, respectively,—and introducing the reduced transition probability $B(EL, I_i \to I_f) = |\langle I_i\|\mathcal{M}(EL)\|I_f\rangle|^2/(2I_i+1)$ [17], we obtain for the probability $[P_{\alpha\beta,12}]_{l,L}$ of the transition in the lth partial wave and of multipolarity L

$$[P_{\alpha\beta,12}]_{l,L} = \frac{4\pi\alpha_f}{\hbar c} B(EL, I_i\to I_f) k_{\alpha\beta}^{2L+2} D_{l,L} \times \int \sum_M |G_{l,L,M}(\varepsilon_2)|^2 \left|R_l^{fi}\right|^2 \frac{K_2^2 dK_2}{(2\pi)^3}. \tag{26}$$

Here K_2 is the magnitude of $\vec{K_2}$ (i.e., the wave vector of the outgoing electron) and we define the quantities

$$D_{l,L} = \frac{(2l_f+1)}{(2L+1)[(2L+1)!!]^2}\begin{pmatrix} l_i & l & l_f \\ 0 & 0 & 0\end{pmatrix}^2, \tag{27}$$

$$G_{l,L,M}(\varepsilon_2) = \sum_n Q^n_{l,L,M}\int_{-\infty}^\infty j_n(k_{\alpha\beta}\xi(x_0)) e^{i(\varepsilon_2-\Delta)x_0} dx_0, \tag{28}$$

where $\Delta = \Delta E/(\hbar c)$ with $\Delta E = E_\alpha - E_\beta + E_1$, $\varepsilon_2 = E_2/(\hbar c)$, and

$$R_l^{fi} = \int_0^\infty R_f(r) h_l^{(1)}(k_{\alpha\beta}r) R_i(r) r^2 dr. \tag{29}$$

Here R_f and R_i denote the radial parts of the nonrelativistic wave functions in the final and initial electronic states, respectively, and l_i and l_f are the angular momentum quantum numbers of the electron in the initial and final states, respectively.

We introduce the averaged IC coefficient (ICC) $\alpha_{l,L}$ induced by an ultrashort, super-intense x-ray pulse

$$\alpha_{l,L} = \frac{(P_{\alpha\beta,12})_{l,L}}{\tau_{\text{ir}} W_\gamma}, \tag{30}$$

where τ_{ir} is the irradiation time and

$$W_\gamma = \frac{8\pi(L+1)k_{\alpha\beta}^{2L+1}}{L[(2L+1)!!]^2\hbar} B(EL, I_i\to I_f), \tag{31}$$

is the rate of direct γ decay [17]. (We may take $\tau_{\text{ir}} = \tau$ as a first estimation.) Thus in the case of bound-free electronic transitions

$$\alpha_{l,L} = \frac{\alpha_f k_{\alpha\beta}}{2c\tau_{\text{ir}}} C_{l,L} \times \int \sum_M |G_{l,L,M}(\varepsilon_2)|^2 \left|R_l^{fi}\right|^2 \frac{K_2^2 dK_2}{(2\pi)^3}, \tag{32}$$

with α_f as the fine structure constant and

$$C_{l,L} = \frac{(2l_f+1)L}{(2L+1)(L+1)}\begin{pmatrix} l_i & l & l_f \\ 0 & 0 & 0\end{pmatrix}^2, \quad (33)$$

and $k_{\alpha\beta} = (E_\alpha - E_\beta)/\hbar c$.

IV. GAUSSIAN X-RAY PULSE AND FREE FINAL ELECTRONIC STATE

The transition probability contains the $G_{l,L,M}(\varepsilon_2)$ function that depends on the parameters of the external x-ray field. Using the condition $\xi k_{\alpha\beta} \ll 1$, which is easy to meet, we approximate the spherical Bessel-function in the integral (28) with its low argument approximation $j_n(k_{\alpha\beta}\xi) \approx (k_{\alpha\beta})^n \xi^n/(2n+1)!!$ [16]. (If the pulse peak intensity $I \ll 10^{22}$ W/cm^2 then $\xi k_{\alpha\beta} \ll 1$ is fulfilled in the case of the ^{99m}Tc isomer to be investigated later numerically.)

In the function ξ [defined by Eq. (6)] we take a Gauss function for the envelope function

$$\xi(k_0x_0,\phi) = \xi_0 e^{-(\frac{k_0x_0}{T})^2}\cos(k_0x_0+\phi). \quad (34)$$

Now we can calculate the $G_{l,L,M}(\varepsilon_2)$ functions. If $n=0$, then in $G_{l,L,M}(\varepsilon_2)$ the $Q^0_{l,L,M}\int e^{i(\varepsilon_2-\Delta)x_0}dx_0 = Q^0_{l,L,M}2\pi\delta(\varepsilon_2 - \Delta) = 0$. This is the x-ray pulse-free case and it expresses that the process is energetically forbidden if the laser is not on. Because of the $\xi k_{\alpha\beta} \ll 1$ condition, the contribution of the terms $n \geqslant 2$ in the sum over n in Eq. (28) is negligible. The only term that yields a contribution corresponds to $n=1$. Thus

$$G_{l,L,M} = \frac{1}{3}k_{\alpha\beta}\xi_0 Q^1_{l,L,M}\frac{T}{k_0}G(\varepsilon_2,T,\phi,\Delta,k_0), \quad (35)$$

with

$$G(\varepsilon_2,T,\phi,\Delta,k_0) = \frac{k_0}{T}\int_{-\infty}^{+\infty} e^{-(\frac{k_0x_0}{T})^2}\cos(k_0x_0+\phi) \times e^{i(\varepsilon_2-\Delta)x_0}dx_0. \quad (36)$$

Carrying out the integration, it reads

$$G(\beta,T,\phi,\delta) = \frac{\pi^{1/2}}{2}\left[e^{-i\phi}e^{-(\frac{(\beta+\delta-1)T}{2})^2} + e^{i\phi}e^{-(\frac{(\beta+\delta+1)T}{2})^2}\right], \quad (37)$$

where $\beta = \varepsilon_2/k_0$ and $\delta = |\Delta|/k_0$. As a result of the factorized form of $G_{l,L,M}$ [see Eq. (35)] the angular momentum conservation in $\alpha_{l,L}$ [see Eq. (32)] is partly expressed through the quantity

$$S^1_{l,L} = \sum_M |Q^1_{l,L,M}|^2. \quad (38)$$

The only nonvanishing $S^1_{l,L}$ values are $S^1_{2,3}=9$ and $S^1_{4,3}=12$. (The $n+l+L=$ even condition results the $l+L=$ odd restriction for $n=1$.)

In Eq. (32) the quantity R^{fi}_l [defined by Eq. (29)] contains radial parts of the initial (bound) and final (free) electronic states that are represented by radial parts of both hydrogen-like (R_i) and free Coulomb [$R_{K_2l_f}(r)$] wave functions, respectively [18]. If the kinetic energy approaches zero ($E_2 \to 0$) (i.e., near the threshold) we may use the low argument approximation of the radial part of the Coulomb function as $R_{K_2l_f}(r) = \sqrt{\frac{4\pi K_2}{r}}J_{2l_f+1}(\sqrt{\frac{8Zr}{a_0}})$, where a_0 is the Bohr radius, $J_{2l_f+1}(\sqrt{\frac{8Zr}{a_0}})$ is the Bessel function of the first kind, and Z is the charge number. To separate the K_2 dependence in Eq. (29) we use the identity

$$|R^{fi}_l(K_2)|^2 = \frac{32\pi^4 a_0^2}{K_2}|\widetilde{R}^{fi}_l|^2, \quad (39)$$

where

$$\widetilde{R}^{fi}_l = \int \widetilde{R}_{n_il_i}(x)J_{2l_f+1}(\sqrt{8x})e^{-\frac{x}{n_i}}x^{3/2}h_l\left(\frac{k_{\alpha\beta}a_0x}{Z}\right)dx, \quad (40)$$

is a K_2 independent quantity. Here $\widetilde{R}_{n_il_i}(x) = a_0^{3/2}R_i(x)$ with $x = Zr/a_0$ and $h_l(k_{\alpha\beta}a_0x/Z)$ denotes the spherical Hankel function.

Using the replacement $\xi_0 = [4\pi\alpha_f\hbar/(\kappa^2c^4k_0^4)]^{1/2}I^{1/2}$ in Eq. (35), where I denotes the peak intensity of the super-intense x-ray pulse, we obtain the x-ray pulse-induced ICC of a bound-free electronic transition

$$\alpha_{l,L} = \alpha^{\text{pulse}}_{l,L,0}\delta^4\frac{\tau}{\tau_{\text{ir}}}\psi(\phi,T,\delta)I, \quad (41)$$

valid in the case of a Gaussian laser pulse, where

$$\alpha^{\text{pulse}}_{l,L,0} = AC_{l,L}S^1_{l,L}|\widetilde{R}^{fi}_l|^2, \quad (42)$$

with

$$A = \frac{2\pi\alpha_f^2a_0^2k^3_{\alpha\beta}}{9\kappa c^3|\Delta|^4}, \quad (43)$$

and

$$\psi(\phi,T,\delta) = T\int_0^\infty |G(\beta,T,\phi,\delta)|^2 d\beta. \quad (44)$$

V. SUPER-INTENSE SUBFEMTOSECOND SOFT-X-RAY LASER PULSE-INDUCED IC PROCESS OF ^{99m}TC ISOMER

The x-ray pulse-induced IC decay of ^{99m}Tc [7,19] is numerically investigated. The ^{99m}Tc isomeric state decays by an $E3$ transition of transition energy $E_{\alpha\beta} = 2.1726$ keV. The half-life of the isomeric state is $\tau_\alpha = 6.01$ h and the total laser-free IC coefficient is $\alpha_{\text{tot}} \simeq 1.6\times 10^7$ [20]. $E_{\alpha\beta}$ is lower than the binding energy of the K and L shell electrons, therefore the IC channel for these shells is energetically forbidden. However, if the x-ray pulse is switched on then the IC process from these shells may start.

It was found in Ref. [7] that the laser-induced IC process from the $2p_{3/2}$ shell is the most probable, therefore this case is investigated further. The binding energy $E_1 = -2676.9$ eV of the $2p_{3/2}$ shell, therefore the energy that is required to ignite the IC process is $|\Delta E| = 504.3$ eV. For the angular frequency of the carrier wave $\sim|\Delta E|/\hbar$ is chosen. The shielding effect of the other electrons is taken into account by introducing the effective charge number, thus $Z_{\text{eff}} = n\sqrt{|E_1|/\mathcal{R}y} = 28.054$ is substituted for Z in Eq. (40), where $\mathcal{R}y$ is the Rydberg energy and the principal quantum number of the shell is $n=2$.

The results of the $\alpha_{l,L}$ calculation show that $\alpha_{4,3}$ is dominant: $\alpha^{\text{pulse}}_{4,3,0} = 1.21\times 10^{-11}$ W^{-1} cm^2 and $\alpha^{\text{pulse}}_{2,3,0}$ is more than

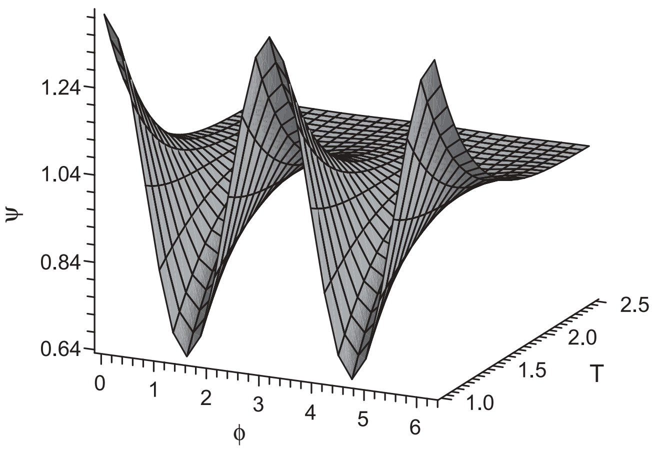

FIG. 1. The carrier-envelope phase (ϕ) ($-2\pi \leqslant \phi \leqslant 0$) and pulse length ($T$) dependence of $\psi(\phi, T)$ in the $n = 1$ process at $\delta = 1$. $T = \omega_0\tau$, $\omega_0 = ck_0$ is the carrier angular frequency and τ is the pulse length, $\delta = |\Delta|/k_0$. $\hbar c|\Delta|$ is the magnitude of the energy defect.

seven orders of magnitude smaller. The ϕ and T dependence of $\alpha_{4,3}$ is illustrated by the plot of $\psi(\phi, T, \delta = 1)$ ($k_0 = |\Delta|$) in Fig. 1. It can be seen that $\alpha_{4,3}$ has significant ϕ dependence at $T = 1$, but the ϕ dependence practically disappears at $T \simeq 2$. However, the drastic increase of $\alpha_{4,3}$ with decreasing pulse length obtained in the case of multiphoton ionization of phase-controlled ultrashort x-ray pulses [10] does not appear.

The plane-wave laser-induced IC coefficient has a threshold property, that is, in the weak field limit the laser-induced IC process may start with the absorption of one photon of $k_0 \geqslant |\Delta|$, but for photons of $k_0 < |\Delta|$ the plane-wave induced IC process is forbidden. However, in the case of few-cycle x-ray pulses the bandwidth of the pulse is comparable with the carrier angular frequency; thus even for $k_0 < |\Delta|$ the energetically forbidden IC process may be started. This situation is shown in Fig. 2 where the $\delta(k_0 = |\Delta|/\delta)$ and the T dependence of $\delta^4\psi$ is plotted at $\phi = n\pi$.

Figure 3 shows the $|G(\beta, \phi)|^2$ function that gives the $\varepsilon_2 = \beta k_0$ and the ϕ dependence of the (differential) IC coefficient at $T = 1$, (a) at $\delta = 0.8$, and (b) at $\delta = 2.4$. It can be seen that $|G(\beta, \phi)|^2$ has maxima at $\beta = 0$ and with $\phi = n\pi$ ($n = 0, 1, 2, \ldots,$).

The $\alpha_{4,3}$ in the plane-wave limit with the $k_0 \geqslant |\Delta|$ condition

$$\alpha_{4,3}^{\text{pw}} = \frac{\pi}{2}\alpha_{l,L,0}^{\text{pulse}}\delta^4 I. \qquad (45)$$

At the threshold ($k_0 = |\Delta|$) it gives $\alpha_{4,3}^{\text{pw}} = 1.91 \times 10^{-11} I$ [21]. Comparing the $\lim_{T\to\infty} \alpha_{4,3}$ (41) and the $\alpha_{4,3}^{\text{pw}}$ (45) expressions yields

$$\frac{\tau}{\tau_{\text{ir}}} \lim_{T\to\infty} \psi(\phi, T, \delta = 1) = \frac{\pi}{2}, \qquad (46)$$

that gives $\tau_{\text{ir}} = 0.627\tau$ in our case.

From the experimental point of view, the number N_v of super-intense subfemtosecond soft-x-ray laser pulse-induced events (the number of x-ray pulse-induced $2p_{3/2}$ vacancies) may be interesting. Their observation may be effectively done with the aid of light-controlled secondary electron emission spectroscopy [22] and ion-charge-state chronoscopy [23] of Auger decay. For a sample containing $N_\alpha(t)$ number of ^{99m}Tc isomer nuclei, N_v may be calculated as

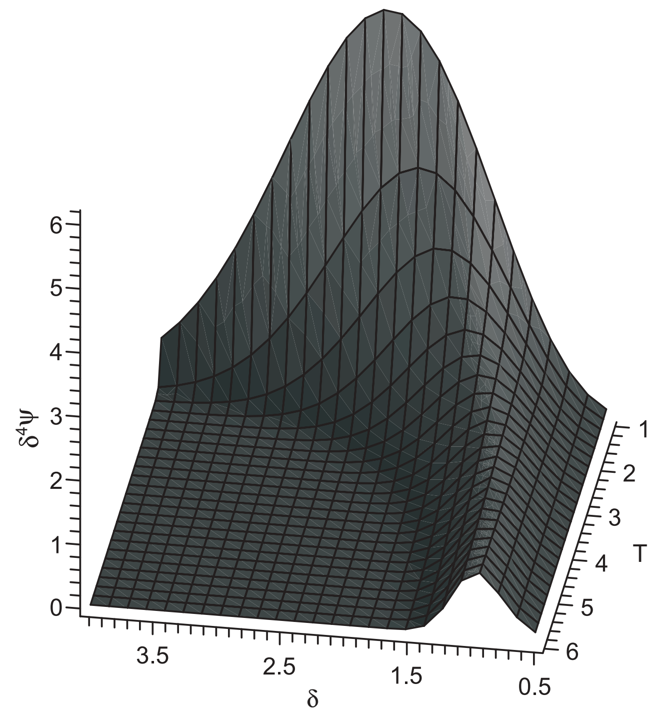

FIG. 2. The ($\delta = |\Delta|/k_0$) and T dependence of $\delta^4\psi$ at $\phi = n\pi$. $T = \omega_0\tau$, $\omega_0 = ck_0$ is the carrier angular frequency and τ is the pulse length, $\hbar c|\Delta|$ is the magnitude of the energy defect.

$$N_v \simeq \sum_{k=1}^{N_p} N_\alpha(t_k)\frac{\alpha_{4,3}}{\alpha_{\text{tot}}}\frac{\tau_{\text{ir}}}{\tau_\alpha}, \qquad (47)$$

where N_p is the number of pulses in the experiment. The decay of the ^{99m}Tc isomer is followed by an $M1 + E2\gamma$ transition of energy 140 keV, of half-life $\tau_\beta = 0.19$ ns, and of mixing ratio $+0.134$ [19]. The number N_{back} of those $2p_{3/2}$ vacancies that are created in the 140 keV transition can mainly be considered as background. For $\delta > 1$ the laser-induced bound-free transition from the $2p_{3/2}$ shell needs more than six photons; consequently the creation of a vacancy in that manner is negligible. Using the secular balance condition

$$N_{\text{back}} = \sum_{k=1}^{N_p} N_\alpha(t_k)\alpha_{140}\frac{\tau_m}{\tau_\alpha}, \qquad (48)$$

where τ_m is the time of measurement of determining the existence of a vacancy. The estimated internal conversion coefficient α_{140} from the $2p_{3/2}$ (L_2) shell of the 140 keV transition is less than 10^{-2} ($\alpha_{140} \lesssim 10^{-2}$). It is reasonable to suppose that $\tau_m \ll \tau_\beta$ and the time between ultrashort x-ray pulses is $t_p \gg \tau_\beta$. Thus the estimated signal-to-background ratio ($\eta = N_v/N_{\text{back}}$) can be written as

$$\eta = \frac{\alpha_{4,3}\tau_{\text{ir}}}{\alpha_{tot}\alpha_{140}\tau_m}. \qquad (49)$$

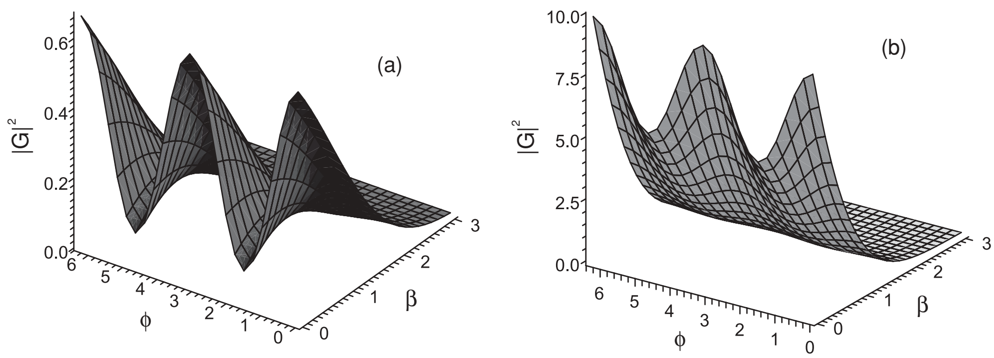

FIG. 3. The $|G(\beta,\phi)|^2$ function, the $\beta = \varepsilon_2/k_0$ and the ϕ dependence of the (differential) IC coefficient at $T = 1$. $\varepsilon_2 = E_2/\hbar c$ where E_2 is the energy of the outgoing electron. $T = \omega_0\tau$, $\omega_0 = ck_0$ is the carrier angular frequency and τ is the pulse length $\delta = |\Delta|/k_0$. $\hbar c|\Delta|$ is the magnitude of the energy defect. (a) $\delta = 0.8$ and (b) $\delta = 2.4$.

Substituting the expression (41) of $\alpha_{4,3}$ and the numerical values of $\alpha^{\text{pulse}}_{l,L,0}$, α_{tot}, and α_{140} we have

$$\eta = \eta_0 \tau I \delta^4 \psi(\phi, T, \delta)/\tau_m, \tag{50}$$

with $\eta_0 = \alpha^{\text{pulse}}_{4,3,0}/(\alpha_{\text{tot}}\alpha_{140}) \gtrsim 7.6 \times 10^{17}\ \text{W}^{-1}\ \text{cm}^2$. From an experimental point of view, the $\rho = N_v/\sqrt{N_{\text{back}}}$ ratio is also informative, where $\sqrt{N_{\text{back}}} = \sigma_{\text{back}}$ stands for the standard deviation of the background. Considering that τ_a is long enough to approximate $\sum_{k=1}^{N_p} N_\alpha(t_k) = A_{q\alpha}\tau_\alpha N_p$, where $A_{q\alpha}$ is the initial activity of the sample and if the x-ray pulse has repetition rate r and the total time of the measurement is T_m then $N_p = rT_m$ and

$$\rho = \sqrt{\frac{A_{q\alpha} r T_m}{\alpha_{140}\tau_m}}\,\frac{\alpha_{4,3}\tau_{\text{ir}}}{\alpha_{\text{tot}}}. \tag{51}$$

Furthermore, if we measure $A_{q\alpha}$ in $\text{mCi} = 3.7 \times 10^7\ \text{s}^{-1}$ units and take $T_m = \mu\tau_a$ ($\tau_\alpha = 6.01$ h), then $\rho = \rho_0\delta^4\psi\tau I\sqrt{A_{q\alpha}r\mu/\tau_m}$ with $\rho_0 = 6.7 \times 10^{-12}\ \text{W}^{-1}\ \text{cm}^2\ \text{s}^{1/2}$. Considering the rapid progress in the field of the generation of few-cycle soft-x-ray pulses and in subfemtosecond metrology [9] it is expected that the required $\rho > 3$ may be achieved in the future.

Finally, for the sake of illustration the $\delta(k_0 = |\Delta|/\delta)$ dependence of N, the number of $2p_{3/2}$ vacancies created by a hypothetical short x-ray laser pulse of laser peak intensity $I = 10^{21}\ \text{W cm}^{-2}$, repetition rate $10\ \text{s}^{-1}$, and $T = 1$, is shown in Fig. 4. The total time of measurement $T_m = 21,600$ s (6 h) and the activity of the sample is 100 Ci. An advantage of the application of an extra short pulse can be seen from Fig. 4. The curve has a maximum at $\delta_m = 2.5$, consequently it is advantageous if the carrier angular frequency $\omega_0 \simeq |\Delta E|/(\delta_m\hbar)$, that is, 0.4 times smaller than the one we need in the plane-wave limit (e.g., in the case of syncrotron radiation). On the other hand, at this angular frequency the efficiency of soft-x-ray pulse generation is about 20 times higher [9,24] than in the case of $\omega_0 \simeq |\Delta E|/\hbar$.

VI. SUMMARY

The results of intense laser-field modified electron-nucleus interaction obtained in the plane-wave case [7] are adapted for the case of a few-cycle x-ray laser field. The few-cycle x-ray pulse-induced IC process is investigated in more detail and the x-ray pulse-induced IC coefficient is deduced. Specifically, the IC coefficient induced by a Gaussian x-ray pulse is derived for bound free electron transitions. The x-ray pulse-induced IC process from the $2p_{3/2}$ shell of the ^{99m}Tc isomer that is energetically forbidden in the laser-free case is investigated numerically. The process has a moderate x-ray pulse length dependence in our case, compared to the one obtained in the case of multiphoton ionization by phase-controlled ultrashort x-ray pulses [10], and it shows significant carrier angular frequency and carrier-envelope phase dependence near the $\tau = \omega_0^{-1}$ pulse length case. The result of the obtained infinite pulse length limit agrees with the plane-wave results. The super-intense peak intensity required to reach the experimentally observable laser-induced $\alpha_{4,3}$ value is hoped to be available in the future [9] and it is also expected that the existence of a Tc ion having a vacant $2p_{3/2}$ electron state may be traced by the method of light-controlled secondary electron emission spectroscopy [22] and ion-charge-state chronoscopy [23] of Auger decay.

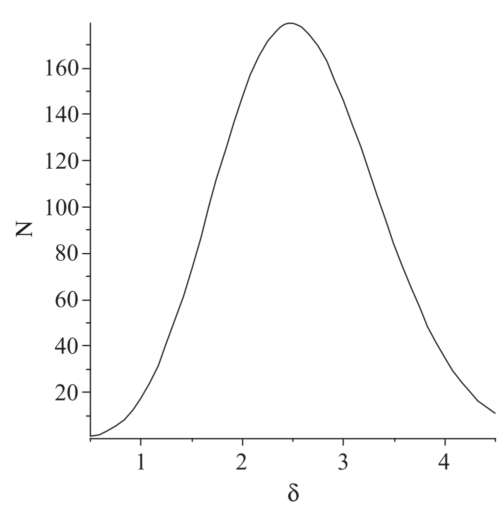

FIG. 4. The δ ($\delta = |\Delta|/k_0$) dependence of N, the number of $2p_{3/2}$ vacancies created by a hypothetic short x-ray laser pulse, with $T = 1$ and $T_m = 21,600$ s (6 h). The activity of the sample is 100 Ci, the repetition rate is $10\ \text{s}^{-1}$, and the laser peak intensity $I = 10^{21}\ \text{W cm}^{-2}$.

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