Laser-Induced Nuclear Excitation

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Abstract—An analysis is presented of the Coulomb excitation of low-lying nuclear levels by the electrons produced by strong-field ionization of atoms. It is shown that the resulting short-lived radioactivity can be as high as on the order of 10^3 Ci for certain isotopes excited by using modern laser systems. Relativistic effects are demonstrated that substantially increase radioactivity as compared to that predicted by nonrelativistic theory results.

1. INTRODUCTION

The moving photoelectron released from an atom by strong-field ionization can be driven back to the ion core by the same laser field and produce a multiply charged ion via inelastic scattering from the core. This multiple ionization mechanism, proposed in [1, 2] and currently known as rescattering, can be tentatively described as a three-step scenario: (1) ionization of a neutral atom by direct interaction between the laser field and a valence electron; (2) acceleration of the outgoing photoelectron by the laser field, followed by its return to the parent ion; (3) impact ionization of the core by inelastic electron scattering from the parent ion.

The second step is modeled as the classical center-of-mass motion of a wave packet representing the electron. In the nonrelativistic limit, the maximum energy of its first recollision with the parent ion is $3.17U_\mathrm{p}$ [2], where

$$U_\mathrm{p} = \frac{(eF)^2}{4m\omega^2}$$

is the ponderomotive potential of the laser field, e is the elementary charge, m is the electron mass, F is the laser electric field amplitude, and ω is frequency. Therefore, rescattering-induced multiple ionization is a threshold process that can compete with direct ionization, which leads to a "knee structure" of double-ionization yield plotted versus laser intensity.

The rescattering model can be applied not only to multiple ionization processes [3], but also to other secondary effects. Examples include recent studies of high-order harmonic generation by infrared laser pulses [4] and above-threshold ionization [5]. The rescattering scenario provides an explanation for the existence of a plateau in the hard-photon and above-threshold electron spectra analyzed in those studies.

Rescattering plays an important role only in a linearly polarized field and is negligible in a circularly polarized field. As the laser intensity increases, so does the Lorentz deflection of the classical electron trajectory by its magnetic component even in the case of linear polarization, which leads to suppression of rescattering. When a Ti:sapphire laser operating at a wavelength of 800 nm is used, rescattering is suppressed at intensities higher than 10^{16} W/cm^2 [6]. However, the Lorentz deflection can be eliminated by using two counterpropagating equal-handed, circularly polarized, mutually coherent laser beams, which makes rescattering possible at any laser intensity [7].

In this paper, we analyze the effect of rescattering on internal degrees of freedom of atomic nuclei. Our analysis is based on the three-step scenario outlined above, but with nuclear Coulomb excitation via photoelectron rescattering as the last step (rather than impact ionization of the ion core). In our previous study [8], this approach was used to calculate nuclear excitation rates for linearly polarized fields with intensities no higher than 10^{17} W/cm^2, when relativistic effects can be ignored. Furthermore, it was shown that radioactivity as high as 10^3 Ci can be obtained for certain isotopes, such as Pu^{239}, by using of a modern laser system. However, being limited to nonrelativistic intensities, our analysis could not be extended to higher nuclear excitation energies. This limitation can be removed by using the experimental configuration proposed in [7]. In the present study, laser-induced excitation rates are found for several isotopes by partly taking into account relativistic effects.

A competing mechanism to that considered here is the nuclear excitation by "foreign" electrons in a high-temperature plasma [9]. However, that heating by this mechanism is feasible only when the laser pulse duration exceeds 1 ps. It plays no significant role when a modern high-power laser is used to generate pulses of duration within a few femtoseconds.

2. GENERAL FORMULAS

Before calculating the yield of ions with nuclei excited by laser-induced recollision, we recall some well-known formulas. The differential cross section for nuclear Coulomb excitation by inelastic electron scattering is expressed as follows [10]:

$$\frac{d\sigma_{Xl}}{d\Omega} = \left(\frac{e}{\hbar c}\right)^2 \frac{4\pi(l+1)}{\lambda[(2l+1)!!]^2}\frac{K^{2l}}{k_i^2}B_{if}(Xl)V(\theta). \quad (1)$$

Here, $\hbar\mathbf{K} = \hbar\mathbf{k}_i - \hbar\mathbf{k}_f$ is the transferred momentum ($\hbar\mathbf{k}_i$ and $\hbar\mathbf{k}_f$ are the initial and final electron momenta, respectively), θ is scattering angle, $d\Omega$ is a solid angle element in the $\mathbf{k}_f$-space,

$$V(\theta) = k_i k_f \frac{(k_i^2 + k_f^2 - \kappa^2)K^2 - 2(\mathbf{k}_i\cdot\mathbf{K})(\mathbf{k}_f\cdot\mathbf{K})}{K^2(K^2-\kappa^2)^2},$$

$\kappa = \Delta E/\hbar c$ (ΔE is nuclear excitation energy), X stands for the type of nuclear transition (E and M for electric and magnetic ones, respectively), and l is the transition multipolarity.

The reduced transition probability $B_{fi}(Xl)$ is related to the γ-decay constant $w(f \longrightarrow i)$ as

$$w(f \longrightarrow i) = \frac{8\pi(l+1)\kappa^{2l+1}}{\hbar l[(2l+1)!!]^2}B_{fi}(Xl),$$
$$B_{if}(Xl) = \frac{2I_f+1}{2I_i+1}B_{fi}(Xl), \quad (2)$$

where I_i and I_f are the initial (ground-state) and final (excited-state) nuclear angular momenta, respectively.

The γ-decay constant $w(f \longrightarrow i)$ is calculated by using the tabular value of the half-life $T_{1/2}$ [11]:

$$w(f \longrightarrow i) = \frac{\ln 2}{T_\gamma}, \quad T_\gamma = T_{1/2}(1+\alpha),$$

where α is the internal conversion coefficient. For mixed $M1 + E2$ transitions, the partial mean lifetimes are expressed in terms of mixing ratio δ as

$$T_\gamma^{M1} = T_\gamma(1+\delta^2), \quad T_\gamma^{E2} = T_\gamma(1+\delta^{-2}).$$

3. CLASSICAL RELATIVISTIC DYNAMICS OF A PHOTOELECTRON

The nuclear excitation rate is evaluated here by taking into account only the first recollision between a relativistic electron and an ion core, which was analyzed in detail in our previous study [12]. This section recapitulates the results reported therein.

The recollision time is calculated by analyzing the classical equations of motion of the wave-packet center, which are valid if the de Broglie wavelength of the photoelectron is much shorter than its displacement. Indeed, the recollision time is $\Delta t \sim \pi/\omega$; the corresponding displacement of a relativistic electron is $l \sim c\Delta t = \pi c/\omega$; and the momentum gained is $p \sim eF\Delta t = \pi eF/\omega$, where F is the laser electric field amplitude. Estimating the photoelectron's de Broglie wavelength as $\lambda = 2\pi\hbar/p \sim 2\hbar\omega/eF$, we have $\lambda/l \sim \hbar\omega^2/ceF$. When the pulse produced by a Ti:sapphire laser tuned to 800 nm ($\omega = 0.056$ au) has an intensity of 3.45×10^{18} W/cm^2 ($F = 10$ au), this ratio is 2×10^{-6}.

The one-dimensional relativistic motion of an electron driven by the electric field $F\sin\omega t$ of a light wave linearly polarized along the z axis is governed by the equations

$$mc\frac{du^0}{ds} = \frac{e}{c}Fu_1\sin\omega t,$$
$$mc\frac{du^1}{ds} = -\frac{e}{c}Fu_0\sin\omega t. \quad (3)$$

Here, $u^0 = u_0$ and $u^1 = -u_1$ are the components of the proper velocity

$$(u^0 \;\; u^1) = \left(1 - \frac{v^2}{c^2}\right)^{-1/2}\left(1 \;\; \frac{v}{c}\right)$$

in the Minkowski space with coordinate time $x^0 = ct$, spatial coordinate $x^1 = z$, and spacetime interval s ($s = ct$ for light waves).

By introducing the proper phase $\zeta = s\omega/c$ related to the proper time $\tau = s/c$, system (3) is simplified to

$$\frac{du^0}{d\zeta} = -fu^1\sin\zeta,$$
$$\frac{du^1}{d\zeta} = -fu^0\sin\zeta, \quad (4)$$

where the dimensionless field strength parameter

$$f = \frac{eF}{mc\omega} \quad (5)$$

determines the relativistic motion of the electron.

The general solution to system (4) is

$$u^0(\zeta) = C_1e^{-f\cos\zeta} + C_2e^{f\cos\zeta},$$
$$u^1(\zeta) = -C_1e^{-f\cos\zeta} + C_2e^{f\cos\zeta}$$

with constants C_1 and C_2 to be determined. The initial conditions to system (4) are set by assuming zero electron velocity at the ionization time t_0 in the laboratory frame: $u^0(\zeta_0) = 1$ and $u^1(\zeta_0) = 0$, where $\zeta_0 = \omega t_0$. The

final expressions for the incoming velocity components of a rescattered electron are

$$u^0(\zeta) = \cosh[f(\cos\zeta - \cos\zeta_0)],$$
$$u^1(\zeta) = \sinh[f(\cos\zeta - \cos\zeta_0)]. \quad (6)$$

The spatial coordinates in the Minkowski space are found by solving the equations

$$\frac{dx^0}{d\zeta} = \frac{c}{\omega}\cosh[f(\cos\zeta - \cos\zeta_0)],$$
$$\frac{dx^1}{d\zeta} = \frac{c}{\omega}\sinh[f(\cos\zeta - \cos\zeta_0)]$$

subject to the initial conditions $x^0(\zeta_0) = ct_0$ and $x^1(\zeta_0) = 0$. As a result, a law of motion $z(t)$ is obtained in parametric form:

$$z = \frac{c}{\omega}\int_{\zeta_0}^{\zeta}\sinh[f(\cos\zeta' - \cos\zeta_0)]d\zeta', \quad (7)$$

$$t = t_0 + \frac{1}{\omega}\int_{\zeta_0}^{\zeta}\cosh[f(\cos\zeta' - \cos\zeta_0)]d\zeta'. \quad (8)$$

The electron's energy and momentum at time t are expressed as

$$E = mc^2\cosh[f(\cos\zeta - \cos\zeta_0)], \quad (9)$$

$$p = mc\sinh[f(\cos\zeta - \cos\zeta_0)]. \quad (10)$$

It is easily checked that (7)–(10) reduce to nonrelativistic formulas when $f \ll 1$. In particular,

$$z(t) = \frac{eF}{m\omega^2}[\sin\omega t - \sin\omega t_0 - \omega(t - t_0)\cos\omega t_0].$$

The proper time $\tau_1 = \tau(t_1)$ corresponding to the first recollision time t_1 is determined by solving the equation $x^1(\zeta_1) = 0$, by virtue of (7) equivalent to

$$\int_{\omega t_0}^{\omega\tau_1}\sinh[f(\cos\zeta' - \cos\zeta_0)]d\zeta' = 0. \quad (11)$$

In the laboratory frame, the time t_1 is calculated by using formula (8) with $\zeta = \zeta_1 = \omega\tau_1$.

In the nonrelativistic limit ($f \ll 1$) Eq. (11) reduces to a well-known equation [3]:

$$\sin\omega t_1 - \sin\omega t_0 = \omega(t_1 - t_0)\cos\omega t_0. \quad (12)$$

As mentioned above, the maximum recollision energy in the nonrelativistic limit is

$$E_{\max}^{(0)} = 0.79mc^2f^2 = 3.17U_p^{(0)}, \quad (13)$$

where

$$U_p^{(0)} = \frac{(eF)^2}{4m\omega^2} \quad (14)$$

is the ponderomotive potential, if the electron is emitted from the atom at the instant corresponding to the field phase

$$\Phi_{0,\max} = \omega t_{0,\max} = 1.884. \quad (15)$$

Hereinafter, subscript (0) denotes nonrelativistic values, as in (13) and (14).

To find relativistic corrections to the nonrelativistic dynamics of a laser-driven electron as described by Eqs. (9)–(11), we expand the hyperbolic functions contained therein in Taylor series in powers of f. Retaining the lowest nonvanishing term in (11), we obtain an equation similar to (12) up to a correction:

$$\sin\zeta - \sin\zeta_0 - (\zeta - \zeta_0)\cos\zeta_0 = f^2A(\zeta, \zeta_0), \quad (16)$$

where

$$A(\zeta, \zeta_0) = \frac{1}{24}(\zeta - \zeta_0)(9\cos\zeta_0 + \cos3\zeta_0)$$
$$-\frac{1}{8}(3 + 2\cos2\zeta_0)\sin\zeta + \frac{1}{8}\cos\zeta_0\sin2\zeta \quad (17)$$
$$-\frac{1}{72}\sin3\zeta + \frac{3}{16}\sin\zeta_0 + \frac{11}{144}\sin3\zeta_0.$$

Analogously, expression (10) for the momentum gained by the electron before the first recollision becomes

$$p_1 = p_1^{(0)} + mcf^3\left[\frac{1}{6}(\cos\zeta_1^{(0)} - \cos\zeta_0)^3 - A(\zeta_1^{(0)}, \zeta_0)\sin\zeta_1^{(0)}\right], \quad (18)$$

where $\zeta_1^{(0)}$ is the root of Eq. (16) with zero right-hand side and $A(\zeta_1^{(0)}, \zeta_0)$ is given by (17). (Relativistic problems are more amenable to analysis in terms of momentum rather than kinetic energy.)

To lowest order in f, the maximum relativistic momentum of the electron before the recollision is expressed as

$$p_{\max} = mcf(1.26 + 0.34f^2), \quad (19)$$

where the first term corresponds to the nonrelativistic limit [12]. For moderately relativistic electrons, approximation (19) is accurate up to a few percent, as illustrated by Fig. 1a.

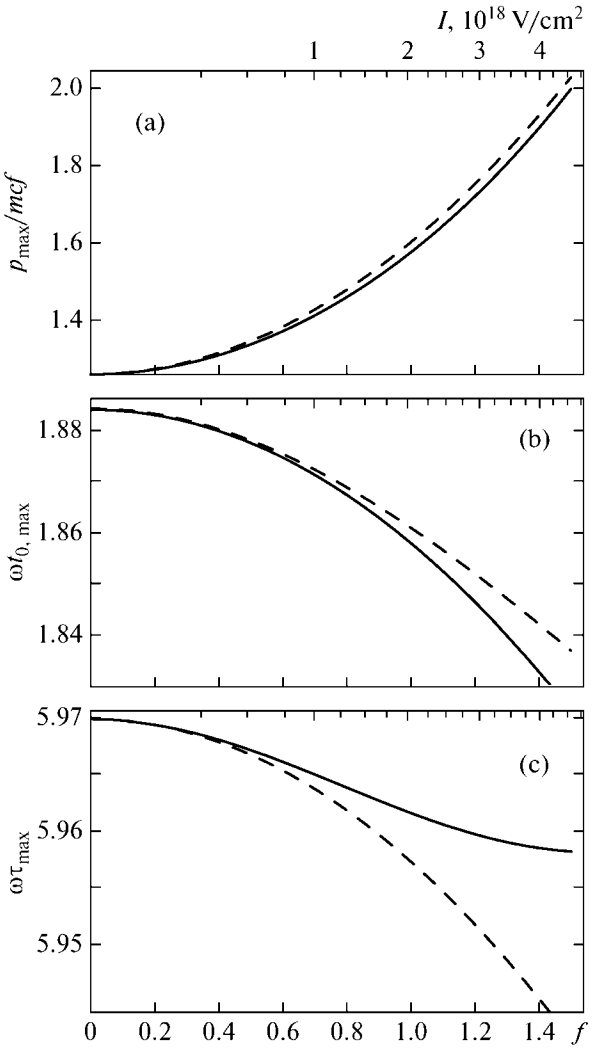

Fig. 1. Curves of p_{max} (a), $\zeta_{0,max}$ (b), and τ_{max} (c) vs. parameter f. Solid curves represent results of exact relativistic calculations; dashed curves are obtained by using approximate formulas (19), (22), and (23).

When the energy gained by the electron is comparable to its rest energy, the expression for ponderomotive potential must take into account the relativistic mass shift [13]:

$$U_p = mc^2\left(\sqrt{1+\frac{2U_p^{(0)}}{mc^2}}-1\right), \qquad (20)$$

where $U_p^{(0)}$ is given by (14).

In the weakly relativistic limit, expression (19) can be used to refine well-known formula (13) for the maximum recollision energy:[1]

$$E_{max} = 3.17U_p\left(1+1.07\frac{U_p}{mc^2}\right). \qquad (21)$$

In the nonrelativistic limit ($U_p \ll mc^2$), expression (21) reduces to (13).

Thus, relativistic effects lead to a steeper increase in recollision energy with f.

The electron emission time $t_{0,max}$ corresponding to the maximum recollision momentum is a much weaker function of f than p_{max}:

$$\zeta_{0,max} = 1.884 - 0.025f^2. \qquad (22)$$

As in (19), the first term in (22) corresponds to the nonrelativistic limit. Figure 1b compares the exact behavior of $\zeta_{0,max}$ with that predicted by (22).

The proper time τ_{max} corresponding to the maximum recollision energy is an even weaker function of f:

$$\omega\tau_{max} = 5.970 - 0.012f^2. \qquad (23)$$

Figure 1c demonstrates that formula (23) has a narrower scope of application than both (19) and (22).

4. NUCLEAR EXCITATION RATE

The nuclear excitation rate is the product of cross section (1) with the incident probability current density j of the rescattered electron:

$$\frac{dP_{if}}{d\Omega} = j\frac{d\sigma}{d\Omega}. \qquad (24)$$

Using the classical model proposed in [3], we calculate j from the lateral spreading of the electron wave packet. For a Gaussian packet, the probability density of finding the electron at a radial distance r from its classical trajectory is

$$\Gamma(r) = \frac{1}{\pi a^2(t)}\exp\left[-\frac{r^2}{a^2(t)}\right],$$

where $a(t)$ is the wave-packet width.

An atom interacting with a superstrong field is ionized in the above-threshold regime. If the ionization probability is assumed constant over a half-cycle of the laser field with frequency ω, then the number of electrons emitted per unit time is proportional to ω/π. Therefore, the z component of the current density is

$$j = \frac{q\omega}{\pi^2 a^2(t)},$$

where q is the final charge state of the ion.

Noting that the photoelectron (released at time t_0) has zero velocity in the plane perpendicular to the vec-

[1] The analog of expression (21) given in [12] contains $U_p^{(0)}$ instead of U_p, which explains the difference in the coefficient of the second term.

tor **F**, we use the nonrelativistic expression for the width of a freely evolving wave packet:

$$a^2(t) = a_B^2\left\{1 + \left[\frac{\hbar(t-t_0)}{ma_B^2}\right]^2\right\},$$

where a_B is the Bohr radius.

Taking the current density in (24) at the first recollision time t_1, we obtain the following final expression for the cycle average of the nuclear excitation rate:

$$P_{exc} = q\frac{\ln 2}{\pi^2}\frac{2I_f+1}{2I_i+1}\frac{\hbar\omega^3}{a_B^2\kappa c^2F^2T_{1/2}(1+\alpha)}$$
$$\times\int_{\pi/2}^{\pi} d\phi_0(\cos\phi_1-\cos\phi_0)^{-2}$$
$$\times\left\{1+\left[\frac{\hbar(\phi_1-\phi_0)}{m\omega a_B^2}\right]^2\right\}^{-1} \quad (25)$$
$$\times\int_0^{\pi}\left(\frac{K}{\kappa}\right)^{2\lambda}V(\theta)\sin\theta d\theta,$$

where $\phi_0 = \zeta_0 = \omega t_0$ and $\phi_1 = \omega t_1$.

5. NUMERICAL RESULTS

We have calculated the excitation rates for nuclei driven by a monochromatic laser field at 800 nm as given by formula (25), using the initial and final angular momenta and parities, as well as the γ-decay half-lives of the excited states, as nuclear parameters. Note that the nuclear excitation rate exhibits threshold behavior as a function of laser intensity, due to the existence of a maximum recollision momentum estimated by (19). Indeed, a nucleus can be excited only if

$$\sqrt{p_{max}^2c^2 + m^2c^4} - mc^2 > \Delta E.$$

Figure 2a shows the curves of P_{exc} divided by the charge state q that were calculated in [8] for low-lying excited states in ^{133}Ba and ^{239}Pu in the nonrelativistic limit. Since the corresponding threshold intensities are 6.25×10^{16} and 3.97×10^{16} W/cm^2, respectively, the geometry proposed in [7] should be used for exciting the nuclei. The behavior of the curves near the threshold is determined by the dynamics of the rescattered electron. The nuclear excitation yield saturates with increasing intensity and ultimately depends on nuclear parameters and internal conversion coefficients rather than laser intensity. In particular, the higher yield of excited ^{239}Pu is explained by the shorter lifetime of its excited state, which implies a higher Coulomb excitation rate.

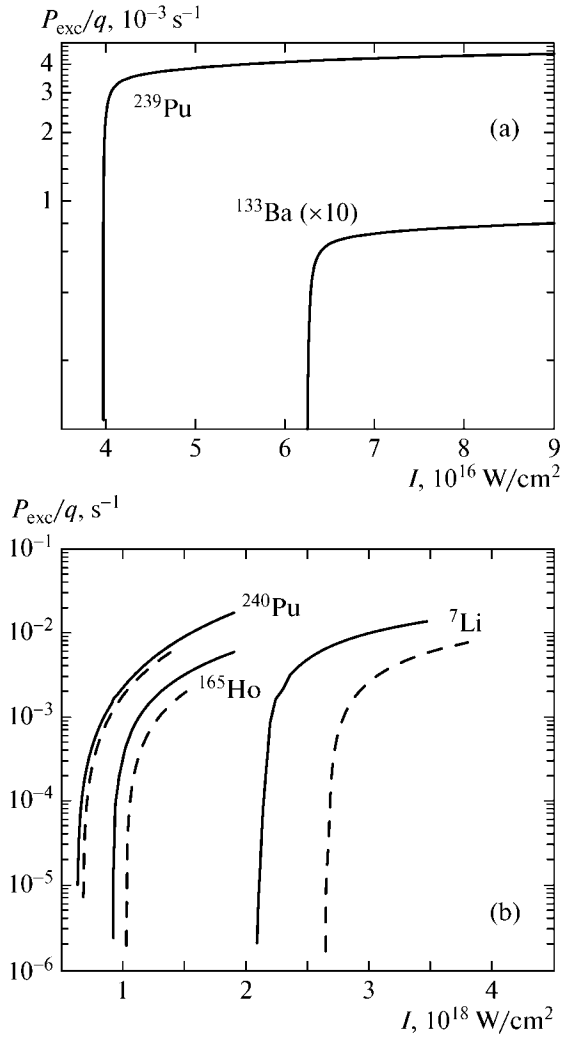

Fig. 2. Nuclear excitation rate divided by charge state q vs. 800-nm laser intensity: (a) nonrelativistic calculations from [8]; (b) comparison between calculations based on relativistic (solid curves) and nonrelativistic (dashed curves) equations of motion. For lithium, q cannot exceed 3.

To estimate the gamma radioactivity generated by laser-induced excitation of the ^{239}Pu isotope in a target, we use a lowest excited state lifetime of 36 ps and assume $q \approx 10$ as reasonable for the laser parameters considered here. Counterpropagating beams with a pulse duration of 10 fs and a pulse energy of 3 J can be focused into a volume of 5×10^{-3} cm^3 to obtain an intensity well above the threshold. The short-lived radioactivity from ^{239}Pu excited by a laser pulse in a gas target of density 10^{20} cm^{-3} can be as high as 10^3 Ci. A similar radioactivity can be obtained by exciting the ^{240}Pu isotope (with lifetime of 164 ps) by a laser pulse with an energy of 45 J, other parameters being equal.

Figure 2b compares the results calculated for other isotopes listed in the table with those obtained in the nonrelativistic limit. It is clear that the role played by

Table

Isotope	$\tau_{1/2}$, ground state	$I_i^{\pi_i}$	ΔE, keV; excited state	$I_f^{\pi_f}$	Type of transition	$T_{1/2}$, ns
^{7}Li	Stable	$3/2^-$	477.612	$1/2^-$	$M1$	7.3×10^{-5}
^{133}Ba	10.52 years	$1/2^+$	12.322	$3/2^+$	$M1$	7.0
^{165}Ho	Stable	$7/2^-$	94.700	$9/2^-$	$M1 + E2$: $\delta = 1.055$	0.022
^{239}Pu	24110 years	$1/2^+$	7.861	$3/2^+$	$M1 + E2$: $\delta = 0.055$	0.036
^{240}Pu	6563 years	0^+	42.824	2^+	$E2$	0.164

relativistic effects increases with nuclear excitation energy, as predicted by expressions (19) and (21).

A comparison between results based on the three-step model of [2] and those obtained by ab initio integration of the time-dependent Schrödinger equation, presented in [14] for above-threshold ionization of an atom, showed that exact treatment of the Coulomb interaction between the photoelectron and the ion core significantly increases the contribution of the first rescattering to the ionization yield (by up to an order of magnitude). Similar results are expected for nuclear Coulomb excitation, leading to excitation rates higher than predicted in this study. Furthermore, it was demonstrated in [14] that higher order rescattering contributions are lower by five orders of magnitude as compared to the first-order one. This finding justifies the assumption above that the first rescattering plays a dominant role in laser-induced nuclear excitation.

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