Kinetics of multiple ionization of rare-gas atoms in a circularly polarized laser field

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The multielectron tunneling ionization probabilities have been calculated for Ar and Kr atoms in circularly polarized laser fields with the pulse durations of 50 fs and 5 fs. The channels of sequential, nonsequential ionization, and ionization with ionic core excitation (inelastic tunneling) have been taken into account. The calculated results demonstrate that many-body effects in tunneling, connected mostly with ionic core excitation, are very important for creation of Ar^{4+}, Ar^{5+}, Kr^{4+}, and Kr^{5+} ions.

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I. INTRODUCTION

The multielectron ionization of atoms by laser radiation was first observed by Suran and Zapesochnyi [1] in alkaline-earth-metal atoms (for review of relevant papers see Ref. [2]). At present such investigations have become one of the basic areas in physics of atom interaction with strong laser radiation [3,4].

The experimental data for linearly polarized laser field conprise the rescattering model [5,6], in which the multicharged ion formation is a result of inelastic collisions of previously emitted electrons with the parental ion (see recent papers [7,8] and review [9]). However, it is well known that rescattaring processes are negligible for laser field with circular polarization [10–12]. This fact has an evident explanation in quantum electrodynamics. Circularly polarized laser beam is a coherent superposition of photons with definite helicity, e.g., $+1$. In absorption of every photon the projection of the electron orbital momentum onto the beam propagation direction increases by $+1$. Since the tunneling ionization according to Keldysh [13] corresponds to absorption of a great number of photons, the continuum electron will have very large orbital momentum projection and hence a large orbital momentum. As a result, the centrifugal repulsion will not allow the free electron to come close to the residual ion. Therefore, the interaction of the free electron with the bound electrons may be neglected.

So, for describing the multicharged ion formation by circular polarized laser radiation the theoretical models should be used taking into consideration the straightforward action of the laser field on atoms. Several theoretical models based on direct impact of laser radiation on atomic electrons were proposed (see, e.g., Refs. [14–23]), in which the shake-off model [16] is the most widely known.

At the same time it is known that creation of singley charged ions in a laser field by tunneling is described well by the Ammosov-Deloné-Kraĭnov (ADK) model [24–26]. In Ref. [27] the empirical generalization of the ADK equations was proposed by Eichmann *et al.* to describe creation of multicharged ions. The existing theory of the atomic tunneling effect for the case of nonsequential emission of several electrons caused by laser radiation was developed by Zon [28]. Formulas in the theories of Refs. [27] and [28] differ in a number of significant aspects. For example, theory [28] contains dependence of residual ion on the charge, whereas in Ref. [27] this parameter is not incorporated in explicit form. However, the formula obtained in Ref. [28] cannot be applied to experiment directly since along with the direct N-charged ion formation, there exist numerous cascade channels, namely, tunneling ionization accompanied by the atomic core excitation. In Ref. [29] the generalized ADK model with core excitation for creation of the doubly-charged ions for the atoms with two s electrons in the outer shell was described. This generalized model is based on the Carlson [30] approach to the one-photon two-electron transitions in atoms. Shake-off model follows from Ref. [29] as limiting case.

In the present paper the role of core excitation accompanying the tunneling multicharged ion formation is considered for an atom with N equivalent electrons in the outer shell. Also we present a set of kinetic equations, describing the dynamics of multicharged ion formation as a result of laser pulse impact on gas targets. The equations are derived and solved numerically for ionization of Ar and Kr up to Ar^{6+} and Kr^{6+}, respectively. We also take into account the following channels of ion creation: sequential ionization, nonsequential tunneling of several electrons, and ionization with atomic core excitation. The influence of laser pulse duration on the yield of multiply charged ions is discussed in detail.

II. ADK THEORY

A. Linearly polarized field

The rate of single-electron tunneling with quantum numbers nlm is defined by ADK equation [24–26]:

$$W_{1\mathrm{lin}}^{(nlm)}=\frac{\sqrt{6\pi}\hbar Z^2}{a^2 m_e \nu^2} C_{\nu l}^2 Q^2 \frac{(2l+1)(l+|m|)!}{2^{|m|}|m|!(l-|m|)!} \times\left(\frac{2F_a}{F}\right)^{2\nu-|m|-3/2}\exp\left(-\frac{2F_a}{3F}\right), \qquad (1)$$

where m_e is electron mass, $a=\hbar^2/m_e e^2$ is Bohr radius, Z is residual ion charge, F is amplitude of light wave electric field,

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$$F_a = \frac{e}{a^2}\left(\frac{Z}{\nu}\right)^3 \tag{2}$$

is electric field of the atomic core, e is absolute value of the electron charge, and ν is effective principal quantum number defined by the binding energy of the emitted electron E_{nl} according to the following formula:

$$\nu = (Z^2 e^2 / 2aE_{nl})^{1/2}. \tag{3}$$

Constant $C_{\nu l}$ defines the behavior of asymptotic electron wave function at $r \to \infty$. In WKB approximation it is given by the following expression [28]:

$$C_{\nu l} = (2\pi\nu)^{-1/2}(2/\nu)^{\nu}\left(\frac{1-\varepsilon}{1+\varepsilon}\right)^{(l+1/2)/2}(1-\varepsilon^2)^{-\nu/2}. \tag{4}$$

Here $\varepsilon = (l+1/2)/\nu$ corresponds to the eccentricity of the classical elliptic orbit of the electron. Expression (4) is obtained for $\varepsilon < 1$. For $\varepsilon > 1$, WKB approximation appears to be invalid and numerical methods are required to calculate constant $C_{\nu l}$.

Q in Eq. (1) is the overlap integral between wave functions of core electrons and their wave functions in initial state of the neutral atom or the parental ion (see Appendix B).

The validity of Eq. (1) is restricted to a small value of the Keldysh parameter [13]

$$\gamma = \frac{\sqrt{2m_e E_{nl}}}{eF}\omega \ll 1, \tag{5}$$

where ω is the laser field frequency, which corresponds to the tunneling regime, and by small strength of the external field as compared to the atomic field:

$$F \ll F_a. \tag{6}$$

Condition (6) corresponds to the WKB approximation which was used in Refs. [13,24] for the derivation of Eq. (1). Note that for highly excited states, such as Rydberg states, characterized by high values of ν, condition (6) may be replaced with a stronger condition due to occurrence of the above-threshold ionization (see Ref. [28] for more details).

B. Circularly polarized field

The absolute value of electric field is time independent in case of circular polarization. Therefore, in Ref. [25] it was concluded that in the circularly polarized field the rate of tunneling is the same as in the dc field. This conclusion, however, is not quite correct [31], since the rate of tunneling depends on magnetic quantum number of the tunneling electron. Then, in the circularly polarized field the projection of electron angular momentum is conserved only in the direction of the light wave propagation, and not in the direction of the electric field. The latter in this case is time dependent.

Considering this fact, we should define the electrical field of the circularly polarized wave propagating along the z axis. In the dipole approximation this field is now determined by the equation

$$\boldsymbol{F}(t) = F(\boldsymbol{e}_x \cos\omega t + \eta \boldsymbol{e}_y \sin\omega t). \tag{7}$$

Here $\boldsymbol{e}_{x,y}$ are unit vectors along respective axes, $\eta = \pm 1$ for the right (left) circular polarization. The electron-field interaction may be expressed as

$$V = e\,\boldsymbol{r}\boldsymbol{F} = erF\sin\theta\cos(\varphi - \eta\omega t), \tag{8}$$

where θ, φ are the polar and azimuthal angles of the vector $\boldsymbol{r}$.

Then, the unitary transformation of the electron wave functions is

$$\phi = \exp(-i\,\eta\omega t\mathcal{L}_z)\tilde{\phi}, \tag{9}$$

where $\mathcal{L}_z$ is the operator of z projection of the orbital momentum. Transformation (9) in classical physics corresponds to the transition to the coordinate frame rotating around the z axis with a frequency ω. This transformation leads to a corresponding transformation of the one-electron Hamiltonian, in accordance with the well-known quantum mechanics equations:

$$\tilde{H} = \exp(i\,\eta\omega t\mathcal{L}_z)(H_0 + V)\exp(-i\,\eta\omega t\mathcal{L}_z) - \eta\omega\mathcal{L}_z. \tag{10}$$

The last term in Eq. (10) is due to the differentiation of wave function (9) with time, and corresponds to the known Larmor theorem assuming that the transition to the noninertial rotating coordinate system is equivalent to the magnetic-field inclusion in an inertial coordinate system.

In the absence of the laser field, the one-electron Hamiltonian H_0 commutates with $\mathcal{L}_z$. Therefore, it is not transformed by unitary transformation (9). Calculation of the transformed expression for V could be easily performed by using the well-known operator relation

$$\exp(A)\exp(B) = \exp(A + B + \tfrac{1}{2}[A,B]),$$

which is valid, when the commutator of operators $[A,B]$ is c number. Hence,

$$\tilde{V} = exF \tag{11}$$

is time independent.

Thus, in the rotating coordinate frame the electrical field is directed along the x axis, and the electron wave functions have the definite projection of the angular momentum onto the z axis. Therefore, these functions should be expanded over other functions, for which the projection of the momentum onto the x axis is the integral of motion. This expansion is based on Wigner angular matrices:

$$\tilde{\phi}_{nlm_z} = \sum_{m_x} D^l_{m_x m_z}(0, \pi/2, 0)\,\tilde{\phi}_{nlm_x}, \tag{12}$$

where m_x, m_z are orbital momentum projections onto respective axes.

The energy of states (12) depends on the value m_z due to Larmorian term in Eq. (10). Since the energy remains unchanged at tunneling, an electron must have the same value of m_z in both initial and final states [44]. Hence, similar to Eq. (12) we have to expand functions ϕ_{nlm_z} over ϕ_{nlm_x}. Therefore this problem resembles a process of passing of the wave packet through the potential barrier [32].

The resulting expression connecting the rates of the tunneling effect at linear and circular field polarizations is the following:

$$W_{1\mathrm{circ}}^{(nlm)}=(\pi F_a/3F)^{1/2}\sum_{m'}|D^l_{m',m}(0,\pi/2,0)|^4 W_{1\mathrm{lin}}^{(nlm')}; \tag{13}$$

$m\equiv m_z$, $m'\equiv m_x$.

Formula (13) is identical to the one derived in Ref. [25] if D function is replaced by the Kronecker symbol $\delta_{mm'}$ in the right-hand side of Eq. (13).

III. INELASTIC TUNNELING

The ADK model could be easily modified to allow to take into account the possibility of formation of excited atoms after tunneling of one of the electrons. Unlike ion formation in the ground state, a process like this indeed occurs at much slower rates. However, with regard to the possibility of subsequent electron tunneling, a process like this appears to be important for the reaction as a whole [29]. Similar to inelastic scattering, the problem under consideration may be termed ''inelastic tunneling.''

Let Δ_k, $k=1,2,\ldots$, be the ion excitation energy. Then, the rate of tunneling ionization with simultaneous ion excitation into a state k is defined by the ADK formulas (1) and (13), in which

$$\nu\to\nu_k=[Z^2e^2/2a(E_{nl}-\Delta_k)]^{1/2},\quad Q\to Q_k, \tag{14}$$

where Q_k is the overlap integral of the electron wave function in the initial states of an atom or of an ion with the wave function in the ionic k state.

In expression (14), the Carlson formula [30], defining the one-photon two-electron process, when both electrons are removed or one of them is removed and another one is excited, is generalized over the case of tunneling ionization. For $\Delta_k \ll E_{nl}$ (E_{nl}/e is the corresponding ionization potential for an atom or an ion), $\nu_k \simeq \nu$ and both formula (14) and Carlson formula are proceeded assuming the sudden approximation [33]. This approximation to be used to describe tunneling effects (shake-up and shake-off processes) was first proposed in Ref. [16], and numerical estimation of the given approximation was made in Ref. [8] for He atom.

IV. MULTIELECTRON TUNNELING

The ADK model could be generalized over the case of nonsequential (for one half cycle of the light field) tunneling of several equivalent electrons [28]. If $E_{nl}^{(1)}/e, E_{nl}^{(2)}/e, \ldots$ are the first, the second, and so on ionization potentials of an atom or an ion, then nonsequential N electron tunneling is defined in the linearly polarized field by the formula

$$W_{N\mathrm{lin}}^{(nl\{m\})}=\frac{\sqrt{3\pi}\hbar}{a^2m_e}\frac{M!(2l+1)^N C_{\nu l}^{2N}}{2^{M-3/2}N^{M+3/2}}Q^2\left(\frac{Z}{\nu}\right)^{3N-1}\times\prod_{j=1}^{N}\frac{(l+|m_j|)!}{(|m_j|!)^2(l-|m_j|)!}\left(\frac{2F_a}{F}\right)^{2N(\nu-1)-M+1/2}\times\exp\left(-\frac{2NF_a}{3F}\right). \tag{15}$$

Here

$$\nu=(NZ^2e^2/2aE_{nl}^{(N)})^{1/2} \tag{16}$$

is the effective principal quantum number for each of the tunneling electrons.

Comparison with formula (3) shows that for the one-electron tunneling effect $\nu\sim(E_{nl}^{(1)})^{-1/2}$, for N-electron tunneling $\nu\sim(E_{nl}^{(N)}/N)^{-1/2}$. In other words, formula (15) assumes that all tunneling electrons have equal energies in both initial and final states.

Index $\{m\}$ in Eq. (15) means the set of magnetic quantum numbers of tunneling electrons $m_1, m_2, \ldots, m_N$, so that

$$M=\sum_{j=1}^{N}|m_j|.$$

The validity of formula (15) is determined by the inequality (6), in which the parameter ν is calculated by Eq. (16), and inequality (5) where the one-electron Keldysh parameter is replaced by the N-electron one:

$$\gamma_N=\frac{\sqrt{2m_eE_{nl}^{(N)}/N}}{eF}\omega\ll 1.$$

It is obvious, that for $N=1$ Eq. (15) turns into Eq. (1).

The connection between rates of N-electron tunneling in linearly and circularly polarized fields is determined by the formula analogous to Eq. (13):

$$W_{N\,\mathrm{circ}}^{(nl\{m\})}=(\pi F_a/3F)^{1/2}\sum_{m_1',\cdots,m_N'}|D^l_{m_1'm_1}(0,\pi/2,0)\ldots\times D^l_{m_N'm_N}(0,\pi/2,0)|^4 W_{N\mathrm{lin}}^{(nl\{m'\})}. \tag{17}$$

Our model will also be valid for the case with N-electron tunneling when the ion remains in an excited state. However, now in Eqs. (15) and (17) parameter ν shall be obtained with the following expression instead of Eq. (16):

$$\nu\to\nu_k=[NZ^2e^2/2a(E_{nl}^{(N)}-\Delta_k)]^{1/2}. \tag{18}$$

V. KINETIC EQUATIONS

The kinetics of multicharged ion formation was considered as multichannel multi-cascaded reactions, incorporating both single-electron and multielectron cascading transitions, and ionization processes accompanied by excitation of

TABLE I. Ionization and excitation energies of Ar^{X+} and Kr^{X+} ions according to the data published by the NIST.

X	Outer subshell		Energies of ionization (italicized) and excitation [46] (cm^{-1}) Ar	Kr
0	p^6	$^1S_0$	0.0	0.0
1	p^5	$^2P_{3/2}$	127109.8	112914.4
		$^2P_{1/2}$	1431.5831	5370.10
2	p^4	$^3P_2$	222848.2	196475.4
		$^3P_1$	1112.175	4548.4
		$^3P_0$	1570.229	5312.9
		$^1D_2$	14010.004	14644.3
		$^1S_0$	33265.724	33079.6
3	p^3	$^4S_{3/2}$	328550.0	298020.0
		$^2D_{3/2}$	21090.4	17036.8
		$^2D_{5/2}$	21219.3	18699.9
		$^2P_{1/2}$	34855.5	31055.2
		$^2P_{3/2}$	35032.6	33404.9
4	p^2	$^3P_0$	481400.0	423400.0
		$^3P_1$	765.23	3742.86
		$^3P_2$	2028.80	7595.34
		$^1D_2$	16298.9	19722.93
		$^1S_0$	37912.0	39203.92
5	p^1	$^2P_{1/2}$	606000.0	521800.0
		$^2P_{3/2}$	2207.1	8108.0
6	p^0	$^1S_0$	734000.0	633100.0

atomic and ionic cores. It is essential that the number of cascading channels of the process dramatically rises as a function of multiplicity of the output ion. Besides, emitted electrons may have various values of magnetic quantum number m_q, which, in turn, results in a further increase of the number of various ways for the multiple atomic ionization process (or, in branching of cascading channels).

The set of kinetic equations, defining the formation of N-charged ions, in our case looks that in like Refs. [16,19]:

$$\frac{dC_f}{dt} = \sum_{f'=0}^{f-1} W_{f\to f'} C_{f'} - \sum_{f'=f+1}^{f_{\rm tot}} W_{f'\to f} C_f, \qquad (19)$$

$$f = 0,1,\ldots,f_{\rm tot},$$

$$C_0(t_0) = 1; \quad C_f(t_0) = 0.$$

Here indices f, f' enumerate ionic states $|p^k(LS)JM\rangle$, where $f=0$ defines a neutral inert gas atom $|p^6(00)00\rangle$; $f_{\rm tot}$ is the total number of all ionic states involved; $W_{f'\to f}$ is the ionic transition rate from state f' to state f in accordance with the tunneling mechanism; and t_0 is the moment of laser pulse delivery. C_f can be treated as a ratio of the f-type-ion concentration n_f to the initial concentration of neutral atoms $n_{\rm tot}$:

$$C_f = n_f/n_{\rm tot}, \quad n_{\rm tot} = \sum_{f=0}^{f_{\rm tot}} n_f.$$

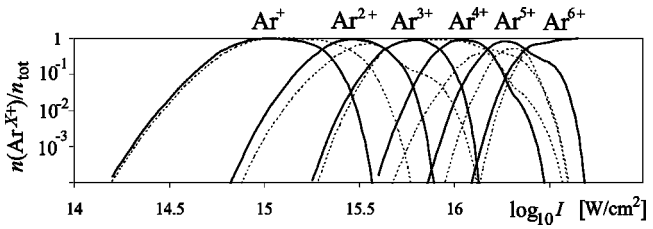

FIG. 1. Concentration of multiply charged Ar ions $n(Ar^{X+})$, defined according to Eq. (20), up to Ar^{6+}, as a function of an intensity of a circularly polarized laser field for a pulse duration of $T=50$ fs in a spatially uniform laser beam with infinitely large focus diameter; $n_{\rm tot}$ is the initial concentration of neutral atoms. Solid lines: all channels involved. Dotted lines: the "pure" ADK model.

Quantities $W_{f'\to f}$ are calculated using Eq. (15) or (17), and overlap integrals are derived analytically in Appendix B [see Eq. (B3)].

We would like to emphasize that the use of quantities $W_{f'\to f}$ in kinetic equations (19) assumes that the laser pulse contains at least three optical cycles [34]. This condition is not fully satisfied in the data below for the pulse duration of 5 fs in the case of Ti:sapphire laser ($\lambda=800$ nm). Obtained results are valid for shorter wavelengths, which satisfy the Keldysh condition (5).

VI. RESULTS AND DISCUSSION

The current section presents results obtained from the integration of the kinetic equations (19), defining the process of multicharged Ar- and Kr-ion formation, up to Ar^{6+} and Kr^{6+}. The analysis was performed for circularly polarized laser field. We deliberately left aside the case of linear polarization as this type of polarization is likely to be affected by the rescattering processes. The envelope of the laser pulse was assumed to be Gaussian: $e^{-t^2/2T^2}$. All rates of cascading transitions were calculated with Eqs. (15), (17), and (19) at the moment $t=+T$ [in Eq. (19) $t_0=-T$] as a function of the laser intensity I. In our calculations we have taken into account all valid transitions between ionic states as defined in Table I.

In the course of analysis the yield of multiply charged ions of argon as a function of the laser radiation intensity I is obtained for a pulse durations of 50 fs and 5 fs, respectively. Let us introduce the concentration of ions A^{X+} in all states:

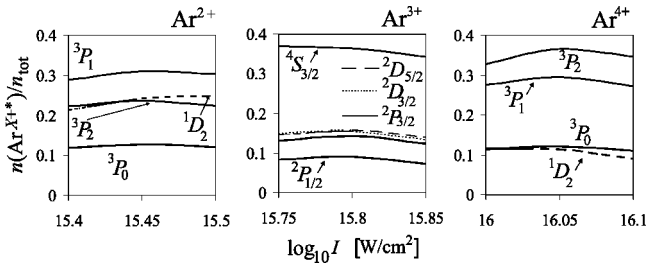

FIG. 2. Populations of ground and excited states of ions Ar^{2+}–Ar^{4+}, as a function of the intensity of a circularly polarized laser field for a pulse duration of $T=50$ fs.

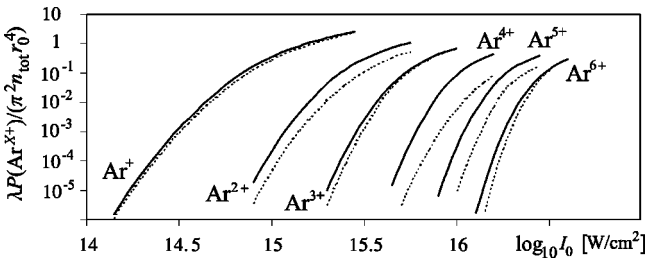

FIG. 3. Spatially averaged integral yield of multiply charged Ar ions $P(\mathrm{Ar}^{X+})$, up to Ar^{6+}, in a focused Gaussian beam (21), as a function of the absolute intensity I_0 of a circularly polarized laser field for a pulse duration of $T=50$ fs. λ is the wavelength, r_0 is the radius of the beam waist, n_{tot} is the initial concentration of neutral atoms. Solid lines: all channels involved. Dotted lines: the "pure" ADK model.

$$n(A^{X+})=\sum_f n_f. \tag{20}$$

The yield curves of multiply charged Ar ions, up to Ar^{6+}, as a function of the laser radiation intensity for a pulse duration of 50 fs are presented in Fig. 1. We compared two groups of results obtained in calculations for the yield of multiply charged Ar ions: first, considering all valid channels, described above, and second, considering only sequential single-electron cascade reactions with inclusion of ionic ground states (pure ADK model). Figure 1 demonstrates that results obtained for both models differ significantly, especially for ions Ar^{2+}–Ar^{5+}.

The populations of ground and excited states of ions Ar^{2+}–Ar^{4+} are presented in detail in Fig. 2 as a function of intensity. Apparently, the population of ionic excited states is comparable with the population of Ar^{2+} and Ar^{3+} ground state and is larger than the former for some Ar^{4+} states. Therefore, the examination of the obtained curves undoubtedly demonstrates the necessity of taking into account the excited states of ionic cores (inelastic tunneling) in studies of tunneling atomic ionization.

However, the obtained results are correct only if the laser beam is spatially uniform and has an infinitely large focus diameter. Now let us consider a focused beam with the Gaussian intensity distribution over the diameter

$$I(\boldsymbol{r})=I_b(z)\exp\left[-\frac{2r^2}{r_b^2(z)}\right], \tag{21}$$

where

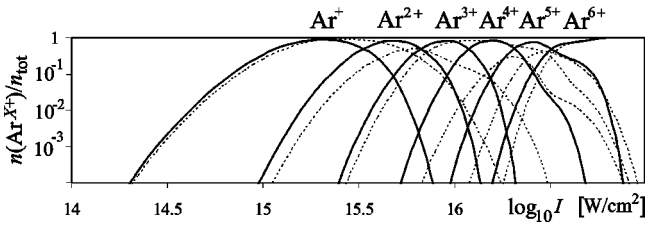

FIG. 4. Same as Fig. 1 but for a pulse duration of $T=5$ fs.

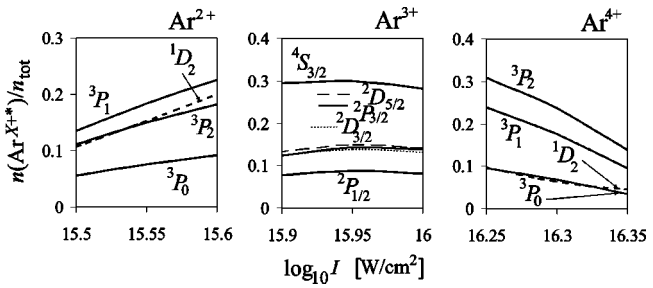

FIG. 5. Same as Fig. 2 but for a pulse duration of $T=5$ fs.

$$r_b(z)=r_0[1+(z/z_0)^2]^{1/2},$$

$$I_b(z)=I_0[1+(z/z_0)^2]^{-1},$$

r_0 is the beam waist radius, I_0 is the intensity of the beam axis in the waist (the absolute intensity), z_0 is the Rayleigh range given by $z_0=\pi r_0^2/\lambda$, and λ is the laser wavelength. If we integrate $C_f(I)$ over the beam volume, we will obtain the following integral spatially averaged ionic yield which is more convenient for comparison with experimental data:

$$P_f(I_0)=n_{\mathrm{tot}}\int C_f[I(r)]d\boldsymbol{r}$$
$$=\frac{n_{\mathrm{tot}}}{\lambda}(\pi r_0^2)^2\int_0^\infty d\zeta(1+\zeta^2)\int_0^{I_0/(1+\zeta^2)}C_f(I)\frac{dI}{I}, \tag{22}$$

where $\zeta=z/z_0$. Let us also introduce the spatially averaged yield $P(A^{X+})$ of A^{X+} ions in all states by analogy of Eq. (20).

The dependence of the integral ionic yield $P(\mathrm{Ar}^{X+})$ on the absolute intensity I_0 is presented in Fig. 3. Similar to Fig. 1, again the results obtained by considering all possible channels are compared with those obtained in the pure ADK model.

Figures 4–6 present results for argon similar to Figs. 1–3 but for a pulse duration of 5 fs. The obtained results explicitly demonstrate the distinct dependence of the yield curves for multiply charged ion creation on the duration of laser pulse.

Figure 3 demonstrates that curves taking into account all possible ionization channels are shifted towards the lower intensities in comparison with the pure ADK theory. Qualitatively these results agree with the experimental data ob-

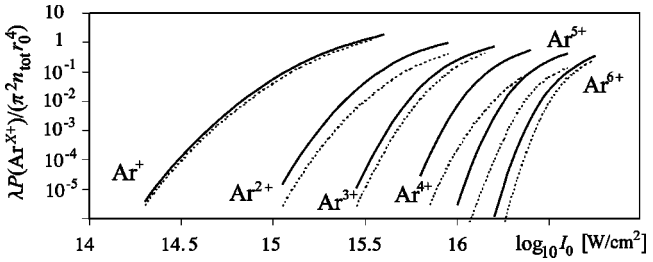

FIG. 6. Same as Fig. 3 but for a pulse duration of $T=5$ fs.

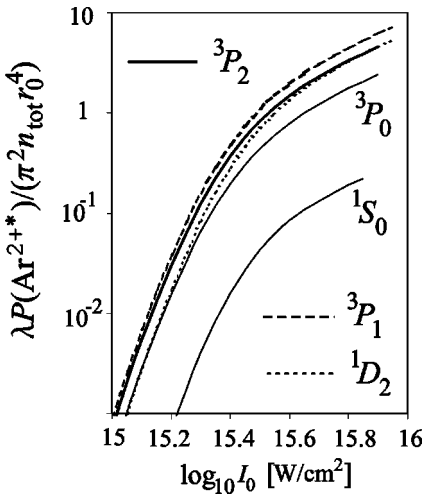

FIG. 7. Spatially averaged integral yield of Ar^{2+} ions in excited states $P(Ar^{2+*})$ in a focused Gaussian beam (21), as a function of the absolute intensity I_0 of a circularly polarized laser field for a pulse duration of $T=50$ fs. All notations are same as in Fig. 3.

tained by Fittinghoff *et al.* [16] for Ne^{2+} in a circularly polarized laser field with $\lambda=614$ nm.

In Fig. 7 we present as illustration of spatially averaged yield for Ar^{2+} ions in excited states. In this case the ground state $^3P_2$ is less populated than the excited state $^3S_2$. This phenomenon can be explained by the difference in overlap integrals (B3) as well as by the relatively low excitation energy (0.56 eV).

We also investigated multiple ionization of krypton for the pulse duration of 50 fs (see Figs. 8–10). Results exhibit similar characteristics of the phenomena. Note that for multiple ionization of krypton, the populations reach maxima and the spatially averaged yield becomes saturated at lower laser field intensities in comparison with argon.

Our numerical calculations demonstrate that the contribution of nonsequential multiple ionization (NSMI) *during a single optical half cycle* determined by Eqs. (15) and (17) does not exceed 0.1% in any of the cases that we have considered. However, for a single optical half cycle, the multiple ionization cannot be obtained experimentally. Therefore, the final conclusion with regard to contribution of the NSMI can be made no sooner than the agreement on the term ''NSMI'' is reached between theorists and experimentalists.

Recent paper by Becker and Faisal [8] presents a theoretical investigation of double ionization of He atoms performed within the *S*-matrix theory and proposes correlated

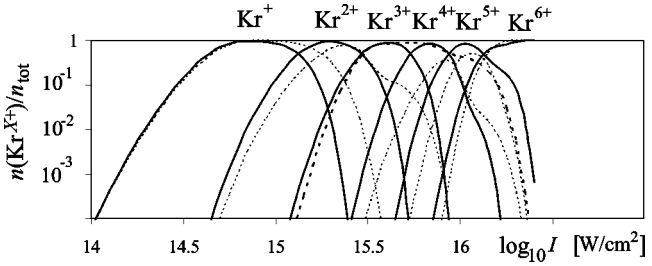

FIG. 8. Same as Fig. 1 but for Kr ions.

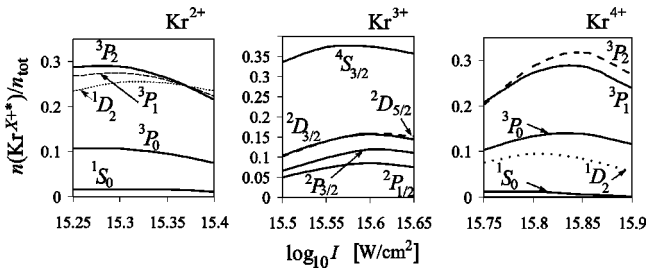

FIG. 9. Same as Fig. 2 but for Kr ions.

energy sharing mechanism to explain experimental data. However, our results are hardly comparable with those obtained in Ref. [8] since both are presented numerically and correspond to different (i) atoms; (ii) intensities; (iii) polarizations. The latter is the most significant.

Guo *et al.* [35] experimentally investigated tunneling formation of Ar^+ and Ar^{2+} ions for the case of circularly polarized field of Ti:sapphire laser and pulse duration of 30 fs. The intensity-selected scanning (ISS) method [36–39] was used. In this method created ions are extracted through a pinhole from a small part of the focal volume lying beside the focal vicinity, not from the entire focal volume. In this case the number of detected ions cannot be obtained by averaging over the focal volume as is required by Eq. (22). In particular, in the ISS method, ionic yield does not follow the usual $I_0^{3/2}$ dependence in the region of saturation [40]. Unfortunately, there is not a simple relation between $C_f(I)$ and the number of ions extracted by the ISS method, while an example of this relation is presented in Ref. [38].

Experimental curves obtained with the ISS method for Ar^+ ions are reproduced by the ADK formulas averaged for the ISS method only in the region of saturation. At the same time, far away from saturation, these experimental curves are higher than what the ADK model. Since our results for single-charged ions are close to the ADK theory, the comparison with the experimental data obtained in Ref. [35] with the ISS method is not possible at present. Let us recall that the experimental data obtained by extracting Ar^+ ions from the entire focal volume agree with the ADK theory very well (see, e.g., Ref. [41]).

VII. SUMMARY

The results we have just dicussed show the significance of the inelastic tunneling effect in formation of multiply charged ions. It should be of interest to verify the obtained data in experiment.

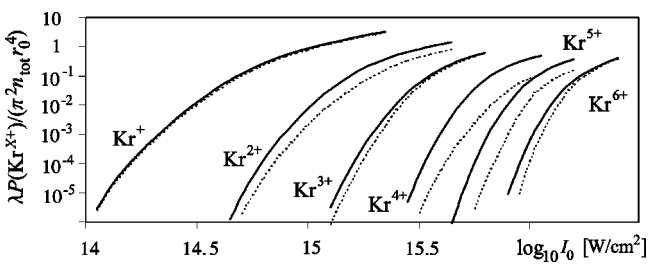

FIG. 10. Same as Fig. 3 but for Kr ions.

Experimental observation of multiply charged ions in excited states should also be interesting since it may clarify the significance of inelastic tunneling. Such an experiment can be performed, for instance, using the probe radiation method [42].

The large contribution of inelastic tunneling to the multielectron Ar and Kr ionization is due to low excitation energy ($\Delta_k \lesssim 4$ eV) of their ions. The influence of inelastic tunneling is decreased with the ion excitation energy, as it takes place for He ($\Delta_k \sim 40$ eV). In this case other many-particle effects (e.g., the correlated energy sharing mechanism from Ref. [8]) may be of greater significance.

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APPENDIX A: ATOMIC AND IONIC WAVE FUNCTIONS

Considering the ionization of inert gas atoms with a p^6 outer shell, we should take into consideration that for all of these atoms and their ions, up to five fold charged ones, the LS coupling occurs. Let $\boldsymbol{r}_q$ be the q's electron radius vector; $\xi_q=(\boldsymbol{r}_q\sigma_q)$ the set of its spatial and spin coordinates; J, M_J the total momentum of an atom or an ion and the projection onto the allocated direction, respectively (in this case along the direction of the electrical field of the light wave, $\boldsymbol{J}=\boldsymbol{L}+\boldsymbol{S}$, where $\boldsymbol{L}$, $\boldsymbol{S}$ are total orbital momentum and spin).

The wave function of five fold charged ion in the p^1 state is

$$\Phi_{JM_J}(\xi;L=1,S=1/2)=R_{np}(r)\sum_{m\mu} C^{JM_J}_{1m1/2\mu}Y_{1m}(\hat{\boldsymbol{r}})\chi_\mu(\sigma). \tag{A1}$$

Here R_{np} is the radial wave function, n is the principal quantum number, χ is the spin function, and the other designations are standard.

For the p^2 state

$$\Phi_{JM_J}(\xi_1,\xi_2;LS)=\sum_{M_LM_S}\sum_{m_1m_2}\sum_{\mu_1\mu_2} C^{JM_J}_{LM_LSM_S}C^{LM_L}_{1m_11m_2} \times C^{SM_S}_{1/2\mu_1 1/2\mu_2}\prod_{q=1}^{2} R_{np}(r_q)Y_{1m_q}(\hat{\boldsymbol{r}}_q) \times\chi_{\mu_q}(\sigma_q), \tag{A2}$$

here, as known, allowed levels are

$$\{JLS\}=\{000\},\{220\},\{011\},\{111\},\{211\}. \tag{A3}$$

For the p^3 state

$$\Phi_{JM_J}(\xi_1,\xi_2,\xi_3;LS) =\sum_{L'S'}\sum_{M_LM_S}\sum_{\text{all }m\mu}\langle p^2(L'S')p|p^3LS\rangle C^{JM_J}_{LM_LSM_S} \times C^{L'M_{L'}}_{1m_11m_2}C^{S'M_{S'}}_{1/2\mu_1 1/2\mu_2}C^{LM_L}_{L'M_{L'}1m_3}C^{SM_S}_{S'M_{S'}1/2\mu_3} \times\prod_{q=1}^{3} R_{np}(r_q)Y_{1m_q}(\hat{\boldsymbol{r}}_q)\chi_{\mu_q}(\sigma_q), \tag{A4}$$

where $\langle p^2(L'S')p|p^3LS\rangle$ is the coefficient of fractional parentage [43]. Here, the allowed levels are

$$\{JLS\}=\left\{\frac{3}{2}0\frac{3}{2}\right\},\quad \left\{\frac{1}{2}1\frac{1}{2}\right\},\quad \left\{\frac{3}{2}1\frac{1}{2}\right\},\quad \left\{\frac{3}{2}2\frac{1}{2}\right\},\quad \left\{\frac{5}{2}2\frac{1}{2}\right\}.$$

For the p^4 state

$$\Phi_{JM_J}(\xi_1,\xi_2,\xi_3,\xi_4;LS) =\sum_{L'S'}\sum_{L''S''}\sum_{M_LM_S}\sum_{\text{all }m\mu}\langle p^2(L''S'')p|p^3L'S'\rangle \times\langle p^3(L'S')p|p^4LS\rangle C^{JM_J}_{LM_LSM_S}C^{L''M_{L''}}_{1m_11m_2}C^{S''M_{S''}}_{1/2\mu_1 1/2\mu_2} \times C^{L'M_{L'}}_{L''M_{L''}1m_3}C^{S'M_{S'}}_{S''M_{S''}1/2\mu_3}C^{LM_L}_{L'M_{L'}1m_4}C^{SM_S}_{S'M_{S'}1/2\mu_4} \times\prod_{q=1}^{4} R_{np}(r_q)Y_{1m_q}(\hat{\boldsymbol{r}}_q)\chi_{\mu_q}(\sigma_q), \tag{A5}$$

and the same levels are allowed, Eq. (A3).

For the p^5 state

$$\Phi_{JM_J}(\xi_1,\xi_2,\xi_3,\xi_4,\xi_5;LS) =\sum_{L'S'}\sum_{L''S''}\sum_{L'''S'''}\sum_{M_LM_S}\sum_{\text{all }m\mu}\langle p^2(L'''S''')p|p^3L''S''\rangle \times\langle p^3(L''S'')p|p^4L'S'\rangle\langle p^4(L'S')p|p^5LS\rangle \times C^{JM_J}_{LM_LSM_S}C^{L'''M_{L'''}}_{1m_11m_2}C^{S'''M_{S'''}}_{1/2\mu_1 1/2\mu_2}C^{L''M_{L''}}_{L'''M_{L'''}1m_3} \times C^{S''M_{S''}}_{S'''M_{S'''}1/2\mu_3}C^{L'M_{L'}}_{L''M_{L''}1m_4}C^{S'M_{S'}}_{S''M_{S''}1/2\mu_4}C^{LM_L}_{L'M_{L'}1m_5} \times C^{SM_S}_{S'M_{S'}1/2\mu_5}\prod_{q=1}^{5} R_{np}(r_q)Y_{1m_q}(\hat{\boldsymbol{r}}_q)\chi_{\mu_q}(\sigma_q). \tag{A6}$$

Similar to the p^1 state, here the allowed levels are

$$\{JLS\}=\left\{\frac{1}{2}1\frac{1}{2}\right\},\quad \left\{\frac{3}{2}1\frac{1}{2}\right\}. \tag{A7}$$

Eventually, for the completely filled p^6 subshell,

$$\Phi_{00}(\xi_1,\xi_2,\xi_3,\xi_4,\xi_5,\xi_6;00)$$

$$=\sum_{L'S'}\sum_{L''S''}\sum_{L'''S'''}\sum_{\text{all } m\mu}\frac{(-1)^{m_5+\mu_5-m_6-\mu_6}}{\sqrt{(2L'+1)(2S'+1)}}$$
$$\times\langle p^2(L'''S''')p|p^3L''S''\rangle\langle p^3(L''S'')p|p^4L'S'\rangle$$
$$\times\langle p^4(L'S')p|p^5 1,1/2\rangle C^{L'''M_{L'''}}_{1m_1 1m_2}C^{S'''M_{S'''}}_{1/2\mu_1 1/2\mu_2}C^{L''M_{L''}}_{L'''M_{L'''}1m_3}$$
$$\times C^{S''M_{S''}}_{S'''M_{S'''}1/2\mu_3}C^{L'M_{L'}}_{L''M_{L''}1m_4}C^{S'M_{S'}}_{S''M_{S''}1/2\mu_4}C^{L'M_{L'}}_{1m_5 1m_6}$$
$$\times C^{S'M_{S'}}_{1/2\mu_5 1/2\mu_6}\prod_{q=1}^{6}R_{np}(r_q)Y_{1m_q}(\hat{\mathbf{r}}_q)\chi_{\mu_q}(\sigma_q). \quad \text{(A8)}$$

We are taking into account that $\langle p^5(1,1/2)p|p^6 00\rangle=1$.

APPENDIX B: OVERLAP INTEGRALS

The many-particle structure of atomic or ionic wave functions is revealed in rates for tunneling through overlap integrals Q.

Let us introduce the notation $\langle\xi^{(k)}|p^kJM(LS)\rangle$ for the state of an atom (or an ion) with k outer equivalent p electrons with total momentum J, its projection M, and intermediate orbital and spin momenta L and S, respectively. The designation ξ^k is used for the set of all spin-spatial variables:

$$\xi^{(k)}\equiv\mathbf{r}_1,\sigma_1;\ldots;\mathbf{r}_k,\sigma_k.$$

For a given k, these states are orthogonal and normalized to the unity:

$$\langle p^kJ'M'(L'S')|p^kJM(LS)\rangle=\delta_{J'J}\delta_{M'M}\delta_{L'L}\delta_{S'S}. \quad \text{(B1)}$$

To obtain angular overlap integrals Q for an N-electron tunneling transition from the initial state $|p^kJ_iM_i(L_iS_i)\rangle$ with k electrons to the final state $|p^{k-N}J_fM_f(L_fS_f)\rangle$ with $k-N$ electrons we neglect the variation of the wave functions of the completely filled internal shells [45].

Now, expressions (A1)–(A8) should be combined into a single expression suitable for arbitrary k and N:

$$\langle\xi^{(k)}|p^kJ_iM_i(L_iS_i)\rangle$$
$$=\sum_{J_fM_fL_fS_f}\sum_{\text{all } m\mu}\langle\xi^{(k-N)}|p^{k-N}J_fM_f(L_fS_f)\rangle$$
$$\times Q^{[kN;J_fM_f(L_fS_f),J_iM_i(L_iS_i)]}_{m_1\mu_1,\ldots,m_N\mu_N}\prod_{q=1}^{N}Y_{1\,m_q}(\hat{\mathbf{r}}_q)\chi_{\mu_q}(\sigma_q). \quad \text{(B2)}$$

Expression (B2) is an expansion of initial ionic state $\langle\xi^{(k)}|p^kJ_iM_i(L_iS_i)\rangle$ over a basis of final states $\langle\xi^{(k-N)}|p^{k-N}J_fM_f(L_fS_f)\rangle$ combined with angular wave functions of emitted electrons. Its coefficients $Q^{[kN;J_fM_f(L_fS_f),J_iM_i(L_iS_i)]}_{m_1\mu_1,\ldots,m_N\mu_N}$ turn out to be the desired overlap integrals. They are expressed in terms of the coefficient of fractional parentage and the Clebsch-Gordan coefficients:

$$Q^{[kN;J_fM_f(L_fS_f),J_iM_i(L_iS_i)]}_{m_1\mu_1,\ldots,m_N\mu_N}$$
$$=\sum_{L_1,\ldots,L_{N-1}}\sum_{S_1,\ldots,S_{N-1}}\langle p^{k-N+1}(L_{N-1}S_{N-1})p|$$
$$\times|p^{k-N+2}(L_{N-2}S_{N-2})\rangle\ldots\langle p^{k-1}(L_1S_1)p|$$
$$\times p^k(L_iS_i)\rangle\sum_{M_{L_i}M_{S_i}}\sum_{M_{L_f}M_{S_f}}C^{J_fM_f}_{L_fM_{L_f}S_fM_{S_f}}C^{J_iM_i}_{L_iM_{L_i}S_iM_{S_i}}$$
$$\times\sum_{\text{all } m}\sum_{M_{L_1},\ldots,M_{L_{N-1}}}C^{L_iM_{L_i}}_{L_1M_{L_1}1m_1}C^{L_1M_{L_1}}_{L_2M_{L_2}1m_2}\cdots$$
$$\times\cdots C^{L_{N-1}M_{L_{N-1}}}_{L_NM_{L_N}1m_N}\sum_{\text{all } \mu}\sum_{M_{S_1},\ldots,M_{S_{N-1}}}C^{S_iM_{S_i}}_{S_1M_{S_1}1/2\mu_1}$$
$$\times C^{S_1M_{S_1}}_{S_2M_{S_2}1/2\mu_2}\cdots C^{S_{N-1}M_{S_{N-1}}}_{S_NM_{S_N}1/2\mu_N}. \quad \text{(B3)}$$

Quantities (B3) have the following properties.

(1) According to the Pauli principle, after permutation of any pairs of indices $m\mu$,

$$Q^{[kN;J_fM_f(L_fS_f),J_iM_i(L_iS_i)]}_{m_1\mu_1,\ldots,m_r\mu_r,\ldots,m_s\mu_s,\ldots,m_N\mu_N}$$
$$=-Q^{[kN;J_fM_f(L_fS_f),J_iM_i(L_iS_i)]}_{m_1\mu_1,\ldots,m_s\mu_s,\ldots,m_r\mu_r,\ldots,m_N\mu_N}. \quad \text{(B4)}$$

(2) The following summation rule is satisfied due to the orthogonality condition (B1):

$$\sum_{J_fM_fL_fS_f}\sum_{\text{all } m\mu}[Q^{[kN;J_fM_f(L_fS_f),J_iM_i(L_iS_i)]}_{m_1\mu_1,\ldots,m_N\mu_N}]^2=1. \quad \text{(B5)}$$

Sets of quantities (B3) may be tabulated with any computer algebra system. Properties (B4) and (B5) may be used to verify results.

[1] V.V. Suran and I.P. Zapesochnyi, Sov. Tech. Phys. Lett. **1**, 420 (1975).

[2] N.B. Delone, V.V. Suran, and B.A. Zon, in *Multiphoton Ionization of Atoms*, edited by S.L. Chin and P. Lambropoulos (Academic Press, New York, 1984), p. 235.

[3] L.F. DiMauro and P. Agostini, Adv. At., Mol., Opt. Phys. **35**, 79 (1995).

[4] *Super-Intense Laser-Atom Physics*, Vol. 12 of *NATO Advanced*

Study Institute, Series B: Physics, edited by B. Piraux and K. Rzazewski, (Kluwer, Dordrecht, 2001).
[5] M.Y. Kuchiev, JETP Lett. **45**, 404 (1987); M.Y. Kuchiev, J. Phys. B **28**, 5093 (1995); M.Y. Kuchiev, Phys. Lett. A **212**, 77 (1996).
[6] P.B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[7] V.R. Bhardwaj, S.A. Aseyev, M. Mehendale, G.L. Yudin, D.M. Villeneuve, D.M. Rayner, M.Y. Ivanov, and P.B. Corkum, Phys. Rev. Lett. **86**, 3522 (2001).
[8] A. Becker and F.H.M. Faisal, Phys. Rev. Lett. **89**, 193003 (2002).
[9] R. Dörner, T. Weber, M. Weckenbrock, A. Staudte, M. Hattass, R. Moshammer, J. Ullrich, and H. Schmidt-Böcking, Adv. At., Mol., Opt. Phys. **48**, 1 (2002).
[10] B. Walker, E. Mevel, B. Yang, P. Breger, J.P. Chambaret, A. Antonetti, L.F. DiMauro, and P. Agostini, Phys. Rev. A **48**, R894 (1993).
[11] K. Kondo, A. Sagisaka, T. Tamida, Y. Nabekawa, and S. Watanabe, Phys. Rev. A **48**, R2531 (1993).
[12] D.N. Fittinghoff, P.R. Bolton, B. Chang, and K.C. Kulander, Phys. Rev. A **49**, 2174 (1994).
[13] L.V. Keldysh, Sov. Phys. JETP **20**, 1307 (1964).
[14] K.C. Kulander, Phys. Rev. A **36**, 2726 (1987).
[15] K.C. Kulander, Phys. Rev. A **38**, 778 (1988).
[16] D.N. Fittinghoff, P.R. Bolton, B. Chang, and K.C. Kulander, Phys. Rev. Lett. **69**, 2642 (1992).
[17] F.H.M. Faisal, Phys. Lett. A **187**, 180 (1994).
[18] A. Becker and F.H.M. Faisal, Phys. Rev. A **50**, 3256 (1994).
[19] K.C. Kulander, J. Cooper, and K.J. Schafer, Phys. Rev. A **51**, 561 (1995).
[20] C.A. Ullrich, U.J. Gossmann, and E.K.U. Gross, Phys. Rev. Lett. **74**, 872 (1995).
[21] A. Becker and F.H.M. Faisal, J. Phys. B **29**, L197 (1996).
[22] P. Maragakis and P. Lambropoulos, Laser Phys. **7**, 679 (1997).
[23] P.A. Golovinski, Laser Phys. **7**, 655 (1997).
[24] B.M. Smirnov and M.I. Chibisov, Sov. Phys. JETP **22**, 585 (1965).
[25] A.M. Perelomov, V.S. Popov, and M.V. Terent'ev, Sov. Phys. JETP **23**, 924 (1966).
[26] M.V. Ammosov, N.B. Deloné, and V.P. Kraĭnov, Sov. Phys. JETP **64**, 1191 (1986).
[27] U. Eichmann, M. Dörr, M. Maeda, W. Becker, and W. Sandner, Phys. Rev. Lett. **84**, 3550 (2000).
[28] B.A. Zon, Sov. Phys. JETP **89**, 219 (1999).
[29] B.A. Zon, Sov. Phys. JETP **91**, 899 (2000).
[30] T.A. Carlson, Phys. Rev. **156**, 142 (1967).
[31] E.B. Tulenko and B.A. Zon, Laser Phys. **13**, 450 (2003).
[32] L.A. MacColl, Phys. Rev. **40**, 621 (1932).
[33] L. Shiff, *Quantum Mechanics* (McGraw-Hill, New York, 1955).
[34] A. Becker, L. Plaja, P. Moreno, M. Nurhuda, and F.H.M. Faisal, Phys. Rev. A **64**, 023408 (2001).
[35] C. Guo, M. Li, J.P. Nibarger, and G.N. Gibson, Phys. Rev. A **58**, R4271 (1998).
[36] P. Hansch and L. Van Woerkom, Opt. Lett. **21**, 1286 (1996).
[37] P. Hansch, M.A. Walker, and L.D. Van Woerkom, Phys. Rev. A **54**, R2559 (1996).
[38] M.A. Walker, P. Hansch, and L.D. Van Woerkom, Phys. Rev. A **57**, R701 (1998).
[39] P. Hansch, M.A. Walker, and L.D. Van Woerkom, Phys. Rev. A **57**, R709 (1998).
[40] A. l'Huillier, L.A. Lompre, G. Mainfray, and C. Manus, Phys. Rev. A **27**, 2503 (1983).
[41] S. Augst, A. Talebpour, S.L. Chin, Y. Beaudoin, and M. Chaker, Phys. Rev. A **52**, R917 (1995).
[42] M.P. de Boer and H.G. Muller, Phys. Rev. Lett. **68**, 2747 (1992).
[43] G. Racah, Phys. Rev. **63**, 367 (1943).
[44] It also follows from preservation of the orbital momentum projection onto the direction of the wave propagation in circularly polarized field that was already mentioned above.
[45] This approximation is accepted further everywhere. It means that the internal shells are considered as ''frozen.''
[46] See, http://physics.nist.gov/cgi-bin/AtData