ATI as a source for multiply charged ion production in a laser field

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Abstract

A theory for the many-electron multi-photon process is presented. It is shown that after single-electron excitation into some level in the continuum (ATI) an inelastic collision of the excited electron with the parent atomic particle can result in an excitation of the ion. It may be the continuum state excitation giving the doubly charged ion or the discrete state which also greatly enhances the doubly charged ion production. The probability of these phenomena greatly exceeds that of the direct ionization of a single-charged ion. The single-electron ATI makes possible the two-electron process even in the moderate field. The example of two-electron excitations of He atom in a 780 nm laser field with intensity above $\approx 10^{14}$ W/cm$^2$ is discussed.

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I. INTRODUCTION

In this paper we consider many-electron processes in a low-frequency laser field. One of the most exciting manifestations among these phenomena is the creation of multiply charged ions in a laser field. Since its discovery (Suran and Zapesochnyi’ 1975) it has been intensively studied experimentally for a variety of atoms and charge multiplicity of ions (L’Huillier et al 1982,1983a,1983b, Lompre et al 1984, Luk et al 1983, Boyer et al 1984, Johann et al 1986, Chin et al 1985, Feldmann et al 1982a, Feldmann et al 1982b, Agostini and Petite 1984,1985a,1985b, Dexter et al 1985, Eichmann et al 1987, Camus et al 1989, Zhu et al 1989, DiMauro et al 1988, Bondar’ and Suran 1993, Walker et al 1994). Different experimental conditions such as a wavelength of radiation, intensity of laser field, duration of laser impulse were used. It was recognised that the ionization process strongly depends on both the atomic structure and the experimental conditions. Multiple ionization can proceed either as a sequential process or as a collective atomic response resulting in simultaneous removal of several atomic electrons. Noble gases are shown (L’Huillier et al 1982,1983a,1983b, Lompre et al 1984, Walker et al 1994) to exhibit simultaneous, direct removal of two electrons below the saturation level. After saturation the sequential ionization takes place (L’Huillier et al 1983a). The later statement agrees with the result of (Johann et al 1986) and with the theoretical estimations (Lambropoulos 1985). For alkaline-earth atoms the two-electron mechanism has been recently shown (Bondar’ and Suran 1993) to be predominant for doubly charged ion formation in the infrared range, while in the optical and ultraviolet ranges the sequential mechanism dominates (Feldmann et al 1982a, Agostini and Petite 1985a, Dexter et al 1985, Eichmann et al 1987, Camus et al 1989, Zhu et al 1989, DiMauro et al 1988).

The resonances can in some cases strongly manifest themselves in the formation of multiply charged ions. There is strong experimental evidence showing that creation of doubly charged alkaline-earth ions is strongly influenced by resonances in neutral atoms and single-charged ions (Feldmann et al 1982a, Feldmann et al 1982b, Agostini and Petite 1984, Dexter et al 1985, Eichmann et al 1987, Camus et al 1989, Zhu et al 1989). The calculation for 5-photon double-electron ionization of Ca atom are in agreement with the resonance mechanism (Tang and
Lambropoulos (1987). In contrast, the resonances seem to be much less important for ionization of noble gases where no resonance structure was reported (L’Huillier et al. 1982, 1983a, 1983b, Lompre et al. 1984, Walker et al. 1994).

There were several attempts to theoretically describe the collective mechanism of ionization in the absence of resonances. In (Boyer and Rhodes 1985, Szöke and Rhodes 1986) there was considered the possibility of energy transfer from the outer electron shell driven by the laser field to the inner-atomic shells. In (Kuchiev 1987) “the antenna” mechanism of ionization was suggested. According to this mechanism after single-electron ionization the ionized electron can absorb energy from the field and transfer it to the parent atomic particle in the inelastic collision. Recently the idea of rescattering of the first ionized electron on the ion has been discussed in (Corkum 1993). In (Ostrovsky and Telnov 1987a, 1987b) there was developed an adiabatic theory of the multiphoton processes. In (Fittinghoff et al. 1992) there was considered the possibility that the first ionized electron is taken away from the atom so quickly that the second electron is shaken off. Note, however, that this idea seems to contradict the adiabatic nature of the multiphoton ionization.

In this paper a theoretical approach to describe the direct, collective many-electron process in the laser field is developed. The basic physical idea is the following. Suppose that first single electron ionization takes place. Suppose that then the ionized electron can undergo the inelastic collision with the parent atomic particle. Then one can expect the electron impact to result in the excitation of the ion into a discrete or continuum state. The reason for the excitation is the fact that the ionized electron strongly interacts with the laser field. It can absorb and accumulate high energy needed for ion excitation. The momentum of the ionized electron oscillates due to wiggling in the laser field. Therefore its “energy” is also time-dependent, it oscillates. The time-dependent “energy” of the ionised electron depends on two parameters. One of them is the time-dependent field momentum of the electron \( k = (F/\omega) \sin \omega t \), where \( F, \omega \) are the strength and frequency of the laser field. The other is the constant translational momentum \( K \). The total momentum is \( K + (F/\omega) \sin \omega t \). Thus the kinetic energy of the photoelectron is \( E_K(t) = (K + (F/\omega) \sin \omega t)^2/2 \). We will show that the ion excitation due to electron impact is possible if the ion excitation energy \( E_{\text{exc}} \) is equal or less than the maximal
The kinetic energy of the photoelectron

\[ E_{\text{exc}} = \max(E_K) = \frac{1}{2} \left( K + \frac{F}{\omega} \right)^2. \] (1)

This is an important inequality. It states that if the field is strong enough, \( F^2/(2\omega^2) \geq E_{\text{exc}} \), then we can forget about the translational momentum, considering \( K \approx 0 \). For weaker fields the translational momentum becomes vital. Here is a point where ATI, see (Agostini et al 1979) and references in review (Freeman and Bucksbaum 1991), comes into play. ATI permits the photoelectron to occupy an excited level in the continuum. It is clear that the higher the level in the ATI spectrum is, the larger is the translational momentum. For sufficiently high level an inequality in Eq.(1) can be fulfilled permitting the mechanism to work. It is important that the necessary level in the ATI spectrum should not be as high as the ion excitation energy is, it can be well below the excitation energy. Really, the energy \( E_{\text{ati}} \) absorbed above the single-electron ionization threshold is equal to the averaged kinetic energy of the photoelectron, \( E_{\text{ati}} = \bar{E}_K = K^2/2 + F^2/(4\omega^2) \). The latter is always less than the maximal kinetic energy, \( \bar{E}_K < \max(E_K) \). The stronger the field is, the larger is the difference between \( \bar{E}_K \) and \( \max(E_K) \). As a result \( \max(E_K) \) can exceed the ion excitation energy, while \( E_{\text{ati}} \) can be below it, \( E_{\text{ati}} < E_{\text{exc}} < \max(E_K) \). All this means that absorption of low energy above the single-ionization threshold can make the considered mechanism to work resulting in high energy excitation of the ion. Section VII illustrates this statement using an example of double ionization of He atom. For the case considered absorption of only few (3–6) quanta above the single-ionization threshold is sufficient to allow for double ionization of He which needs very high energy. It is important that ATI takes place with high probability as the numerous experimental data presented in (Freeman and Bucksbaum 1991) and numerical calculations (Potvliege and Robin Shakeshaft 1989) demonstrate. This makes the considered mechanism to be very efficient.

The idea of the process considered is illustrated by the Feynman diagrams in Fig.1.
The solid lines in Fig. 1 describe the behaviour of two electrons. First both of them are in the ground state of the atom. The lines marked $a$ and $b$ describe the corresponding atomic states. Then the electron $a$ is ionised. The beginning of the ionization process is shown by the sloped dashed line representing the absorbed photon. The propagation of the ionized electron in the laser field, when it absorbs another photon, is shown by the double solid lines. Collision of the ionized electron with the parent atomic particle is shown by the vertical dashed line representing the Coulomb interaction between the electrons. The state marked $c$ describes the ion excitation which can belong either to the discrete or to the continuum spectrum. In both cases it can strongly interact with the laser field. The diagram $b$ describes the exchange process.

The central question of the problem is whether this mechanism can work at all. It arises because in the process of ATI the ionized electron could go far outside the atomic particle and therefore inelastic collision with it could seem improbable. To prove that this mechanism works at all we have to fulfil an accurate calculation. It seems very complicated because we are to consider the perturbation theory over the electron interaction when the behaviour of the electrons is strongly influenced by the time-dependent laser field. Fortunately, the adiabatic nature of the problem results in a very important property of the two-electron amplitude: it may be presented as a product of the amplitude of single-electron ionization and the amplitude of electron ion impact in the presence of a laser field. Both latter amplitudes can be reliably calculated. As a result there appears the possibility of reliable ab initio calculation of the two-electron process.

Factorization of the two-electron amplitude is one of the most important results of the paper. The calculations proving it are fully presented because problems of the type considered
are not well-known in literature. We will be forced also to consider briefly two related problems: single-electron ionization and the scattering problem.

Factorization of the amplitude makes it possible to find a reliable estimation for the probability. It permits us to prove that the considered mechanism of two-electron ionization works well. There is a high probability for the first ionized electron to return to the parent atomic particle. The first ionized electron can be considered as a kind of antenna, it absorbs energy from the external field and transfers it to the ion. This physical idea was first considered in (Kuchiev 1987) but the corresponding calculations were not published.

The electric field is assumed to be linearly polarized

$$F(t) = F \cos \omega t.$$  \hspace{1cm} (2)

The frequency $\omega$ of the field is supposed to be low, $\omega \ll I$, where $I$ is the ionization potential. The Keldysh adiabaticity parameter $\gamma$

$$\gamma = \frac{\omega\sqrt{2I}}{F}$$  \hspace{1cm} (3)

is considered as arbitrary. The first ionised electron, which plays so crucial role in the considered picture, will be described neglecting the static Coulomb field created by the parent atomic particle. A similar description was used in the Keldysh theory of single-charged ion formation (Keldysh 1965, Reiss 1980,1987). One can consider it a reasonably good approximation when the non-resonant processes are important, for example for the two-electron ionization of noble gases. This approximation should be even more reliable for the problem of two-electron detachment from negative ions.

In Section II the general formulae describing the single-electron ionization problem as well as the two-electron problem are presented. Section III contains the prove of factorization of the two-electron amplitude. Section IV presents the necessary consideration of the scattering problem. In Section V a qualitative physical discussion of the physical ideas governing the process of two-electron ionization is given. Section VI presents the estimation of the probability of the two-electron process for different regions of adiabatic parameter. ATI is shown to play the major role when $\gamma > 1$. Section VII gives an illustrative example: the double ionization of He atom by a 780 nm laser field is briefly considered.
II. MULTI-PHOTON PROCESSES

In this section a formalism to describe the multi-photon processes when an atom is placed in a laser field is developed. The general expression for the amplitude \( A_{fi} \) of an adiabatic process in the field is

\[
A_{fi} = \frac{1}{T} \int_{0}^{T} \langle \Psi_f(t) | V_F(t) | \psi_i(t) \rangle .
\] (4)

The wave function \( |\psi_i(t)\rangle \) describes the initial state of the atom taking no account of the interaction of the atom with the laser field. This interaction is described by the operator \( V_F(t) \)

\[
V_F(t) = -r F \cos \omega t ,
\] (5)

where \( r = \sum r_i \) is the dipole moment. We use the \( r \)-form for interaction with the laser field. The wave function \( \langle \Psi_f(t) | \) describes the final state of the reaction taking full account of the interaction of the atom with the laser field. It makes the amplitude (4) describe the multi-photon, nonlinear process. Eq.(4) looks similar to the usual expression for stationary processes except for the fact that in (4) there is the nontrivial integration over the period of time \( T = 2\pi/\omega \), where \( \omega \) is the frequency of the laser field.

A. Single-electron ionization

First let us consider shortly the well-known Keldysh problem of multi-photon single-electron ionization (Keldysh 1965). focusing our attention on those points which will be used in the following consideration of many-electron processes. The initial wave function for the single-electron process is taken as

\[
\psi_i(t) = \Phi_a(r, t) = \phi_a(r) \exp (-iE_a t) ,
\] (6)

where \( \phi_a(r) \) is the wave function of the ground state of the atom, and \( E_a \) is the atomic binding energy. The wave function of the final state must include the interaction of the photo-electron with the external field. If one neglects the static ion field in the final state then this wave function is equal to the Volkov wave function (Volkov 1935) \( \Psi_f(t) = \Phi_p(r, t) \),
\begin{align}
\Phi_p(r, t) &= \exp\left\{i \left[(p + k_t)r - \int_0^t E_p(\tau)\,d\tau + \frac{pF}{\omega^2}\right]\right\}; \quad (7) \\
k_t &= \frac{F}{\omega} \sin \omega t, \quad E_p(\tau) = \frac{1}{2}(p + k_\tau)^2, \quad (8)
\end{align}

which satisfies the Schrödinger equation

\begin{equation}
i \frac{\partial}{\partial t} \Phi_p(r, t) = \left(-\frac{1}{2} \Delta - rF \cos \omega t\right) \Phi_p(r, t). \quad (9)
\end{equation}

The additional phase $pF/\omega^2$ in Eq.(7) is chosen to provide a convenient property

\begin{equation}
\Phi_p(r, t + T/2) = \Phi_{-p}(-r, t) \exp\{-i\bar{E}_p T/2\}, \quad (10)
\end{equation}

which is essential for consideration of the parity conservation law, see Eq.(14) below. Substituting (3),(4) in (4) one gets the known expression for the amplitude $A^{(e)}(l, p)$ of single-electron ionization when $l$ quanta are absorbed and the final-state momentum of the electron is $p$:

\begin{equation}
A^{(e)}(l; p) = \frac{1}{T} \int_0^T \langle \Phi_p(t) | V_F(t) | \Phi_a(t) \rangle = \frac{1}{T} \int_0^T \langle p + k_t | V_F(t) | \phi_a \rangle \exp\left\{i \omega S(\omega t)\right\}. \quad (11)
\end{equation}

Here the usual definition $|p\rangle = \exp\{ipr\}$ is used and $S(x)$ is

\begin{equation}
S(x) = \int_0^x \frac{1}{2}(p + \frac{F}{\omega} \sin x)^2 \,dx - E_a x - \frac{pF}{\omega}. \quad (12)
\end{equation}

The energy conservation law for the process of ionization reads

\begin{equation}
E_a + l\omega = \bar{E}_p. \quad (13)
\end{equation}

The parity conservation law for the ionization process manifests itself as a condition for the amplitude

\begin{equation}
A^{(e)}(l; -p) = (-1)^l P_a A^{(e)}(l; p), \quad (14)
\end{equation}

where $P_a = \pm 1$ is the parity of the atomic state $a$. It is easy to verify using Eq.(14) that the amplitude Eq.(11) satisfies this condition.

The integrand in Eq.(11) contains a large phase $\sim 1/\omega$ and therefore the steepest descent method is applicable. The saddle points, which must be taken in the upper semiplane of the complex plane $x = \omega t$, $\text{Im } x > 0$, in the region $0 \leq \text{Re } x \leq 2\pi$ satisfy the equation
\[ S'(x) = \frac{1}{2} (p + \frac{F}{\omega} \sin x)^2 - E_a = 0 , \]  

which results in the following condition

\[ \sin x = \frac{\omega}{F} \left[ -p_\| \pm i (\kappa^2 + p_\perp^2)^{1/2} \right] , \]

where \( p_\| \) and \( p_\perp \) are the longitudinal and transverse components of the vector \( p \) in respect to the field \( F \) and \( \kappa = \sqrt{2|E_a|} \). It follows from Eq.(15) that there are two saddle points \( x_1, x_2 \) in the region of interest which we will label in such a way that

\[ \text{Re} \cos x_1 > 0, \text{Re} \cos x_2 < 0; \text{Im} \ x_1, \text{Im} \ x_2 \geq 0, 0 \leq \text{Re} \ x_1, \text{Re} \ x_2 \leq 2\pi . \]

Calculating the integral in Eq.(11) by the saddle-point method one finds

\[
A^{(e)}(l, p) = \sum_{\sigma=1,2} A^{(e)}_\sigma (l, p) ,
\]

\[
A^{(e)}_\sigma (l; p) = \frac{1}{T} \int_{C_\sigma} \frac{i}{2\pi} \frac{\omega}{S''(x_\sigma)} \exp \left\{ \frac{i}{\omega} S(x_\sigma) \right\} \left\{ \langle p + k|V_F(t)|\phi_a \rangle \right\} (t=x_\sigma/\omega) ,
\]

where \( A^{(e)}_\sigma (l, p), \sigma = 1,2, \) is the contribution to the amplitude of the saddle point \( x = x_\sigma \) satisfying Eqs.(15), (17) and \( C_\sigma \) is the part of the integration contour in the \( t \) plane which crosses the saddle point \( x_\sigma \). (Note that evaluation of \( \langle p + k|V_F(t)|\phi_a \rangle \) needs accuracy because of the singular nature of this matrix element for complex \( t \).) It is easy to verify that the amplitudes \( A^{(e)}_\sigma (l; p) \) satisfy the condition

\[ A^{(e)}_2 (l; -p) = (-1)^l P_a A^{(e)}_1 (l; p) , \]

which shows explicitly that the amplitude Eq.(18) obeys the parity conservation law Eq.(14). The absolute values of \( A^{(e)}_1 (l; p) \) and \( A^{(e)}_2 (l; p) \) are equal. In contrast, their phases differ substantially and this phase difference depends on the field \( F \). In order to check it out consider small \( p, p^2 \ll \kappa^2 \). For this case the phase difference is easily found to be \( 2\sqrt{1 + \gamma^2} Fp_\|/\omega^2 \). The estimation for the smallest \( p_\| \) is \( p_\| \sim \sqrt{\omega} \). Therefore the estimation for the phase difference is \( \sim 2(F^2/\omega^2 + \kappa^2)^{1/2}/\sqrt{\omega} \gg 1 \). The large phase difference makes interference between \( A^{(e)}_1 (l; p) \)
and $A_2^{(e)}(l; p)$ not important for many cases. Then calculating the probability one can suppose that

$$|A^{(e)}(l; p)|^2 = 2|A_1^{(e)}(l; p)|^2 .$$  \hspace{1cm} (21)

**B. Many-electron processes**

Let us develop an approach to the two-electron processes in a way similar to the single-electron ionization. The main purpose of this section is to obtain the analytical expression describing the Feynman diagrams in Fig.1. The initial two-electron wave function may be chosen as a product of the single-electron wave functions

$$\psi_i(r_1, r_2, t) = \phi_a(r_1)\phi_b(r_2) \exp\{-i(E_a + E_b)t\} .$$  \hspace{1cm} (22)

This form of the wave function neglects the initial-state correlations, but they do not play a role. The most interesting for our purpose is the final-state interaction. To simplify the presentation we do not take into account explicitly the symmetry of the wave function with respect to permutation of electrons though it can be restored as discussed below. At the moment let us consider the two electrons as if they are distinguishable. The electron whose coordinates are $r_1$ will be considered as an atomic electron. It will also be referred to as “the first” electron. The electron with coordinates $r_2$ will represent the degrees of freedom of the single-charged ion. It will be called “the second” electron. The wave function $\phi_a(r_1)$ describes the ground state of the atom, $E_a$ is the binding energy. The function $\phi_b(r_2)$ is to be considered as the wave function of the ground state of the single-charged ion, $E_b$ being the binding energy of this state. Note that the removal of the first electron is an adiabatic process, see Section A. Hence the ion remains mainly in the ground state after single-electron ionization. We are interested in the interaction of the atom with the laser field. Therefore we consider the interaction of the first electron with the field. In particular we take into account the excitation of this electron into continuum states. In contrast, the direct interaction of the ion in the ground state with the laser field is strongly suppressed due to a high ionization potential of the ion. That is why
we will neglect direct interaction of the second electron with the laser field when it occupies the ground state. Certainly this interaction will be considered for the excited states. Thus the only possibility to be considered for the excitation of the second electron comes from its interaction with the first one.

If we neglect the two-electron interaction then the final state wave function can be chosen as a product of the Volkov wave function for the photoelectron and the ion ground state wave function

$$\Psi_{fb}(\mathbf{r}_1, \mathbf{r}_2, t) = \Phi_p(\mathbf{r}_1, t)\phi_b(\mathbf{r}_2) \exp\{-iE_b t\}. \quad (23)$$

Substituting Eqs.\((22),(23)\) into Eq.\((11)\) we reproduce Eq.\((11)\) for the single-electron process.

Consider now the excitations of the single-charged ion into discrete or continuum states. The later case describes the doubly charged ion formation. The simplest wave function describing the excitation is given by the product of the wave function of the ionized electron and the wave function of the excited ion

$$\Psi_{fc}(\mathbf{r}_1, \mathbf{r}_2, t) = \Phi_p(\mathbf{r}_1, t)\Phi_c(\mathbf{r}_2, t) . \quad (24)$$

Here we describe the wave function of the first electron in the continuum by the Volkov wave function \((7)\). The function \(\Phi_c(\mathbf{r}_2, t)\) describes the excited state \(c\) of the ion interacting with the laser field. It satisfies the time-dependent Schrodinger equation

$$i\frac{\partial}{\partial t} \Phi_c(\mathbf{r}_2, t) = (H_{ion}(\mathbf{r}_2) - \mathbf{r}_2 F \cos \omega t) \Phi_c(\mathbf{r}_2, t) , \quad (25)$$

where \(H_{ion}\) is the Hamiltonian of the single-charged ion. We will consider the discrete as well as the continuum state excitations. The wave function of the discrete state, if it is well separated from the other states, may be found in an adiabatic approximation

$$\Phi_c(\mathbf{r}_2, t) = \phi_c(\mathbf{r}_2, t) \exp\{-i \int_0^t E_c(\tau)d\tau\} . \quad (26)$$

Here \(\phi_c(\mathbf{r}_2, t)\) is the wave function satisfying the stationary Schrodinger equation

$$E_c(t)\phi_c(\mathbf{r}_2, t) = (H_{ion}(\mathbf{r}_2) - \mathbf{r}_2 F \cos \omega t)\phi_c(\mathbf{r}_2, t) , \quad (27)$$
with the time-dependent “energy” $E_c(t)$. When the second order of perturbation theory over
the field is applicable then

$$E_c(t) = E_c - \frac{1}{2} \alpha_c(\omega) F^2 \cos^2 \omega t,$$

(28)

where $E_c$ is the energy of the stationary ion state $c$ and $\alpha_c(\omega)$ is the dynamical polarizability
of this ion state. The excitation of the ion into the continuum will be described with the help
of the Volkov wave functions (7). It is convenient to consider the discrete state excitations
and the excitations into the continuum on equal ground. Note that if we neglect the ion static
field in Eq.(27) then this equation is reduced to Eq.(9) describing the continuum states of the
ion. Therefore we can consider the discrete states of the ion and its continuum states using
the same set of Eqs.(26),(27) for the wave function and keeping in mind that $H_{ion}(r_2) \approx -(1/2)\Delta_2$
if a state in the continuum is considered. We will suppose that the phase in the definition of
$\Phi_c(r, t)$ is chosen in such a way that

$$\Phi_c(r, t + T/2) = \Phi_{P(c)}(-r, t) \exp\{-i\bar{E}_c T/2\},$$

(29)

$$\bar{E}_c = \frac{1}{T} \int_0^T E_c(t) dt ,$$

where $\Phi_{P(c)}$ is the wave function of the state with the quantum numbers obtained by applying
the inversion operator $P$ to the quantum numbers of the state $c$. The definition Eq.(29) agrees
with Eq.(11). The possibility to satisfy Eq.(29) follows from the Schroedinger Eq.(25).

The excitation of the ion takes place due to a mixing of the wave function (24) describing
the excitation of the ion with the wave function (23) describing its ground state. In order
to find this mixing consider the time-dependent Schrodinger equation for the final state wave
function

$$-i \frac{\partial}{\partial t} \Psi_f^{*}(r_1, r_2, t) = \left( H_0(r_1, r_2, t) + \frac{1}{r_{12}} \right) \Psi_f^{*}(r_1, r_2, t) .$$

(30)

Here

$$H_0(r_1, r_2, t) = -\frac{1}{2} \Delta_1 - r_1 F \cos \omega t + H_{ion}(r_2) - r_2 F \cos \omega t$$

(31)

is the Hamiltonian describing the considered two-electron system when the interaction between
the electrons is neglected. This interaction is accounted for in Eq.(30) by the term $1/r_{12}$. 

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Consider this interaction in the first order of perturbation theory. We are looking for the final state wave function which in the future, \( t \to \infty \), gives the excited ion described by the wave function Eq.(24). Therefore the later wave function gives the zero approximation

\[
\Psi_f(r_1, r_2, t) \approx \Psi_f^{(0)}(r_1, r_2, t) = \Phi_p(r_1, t)\Phi_c(r_2, t) .
\] (32)

Then the correction \( \delta \Psi_f^*(r_1, r_2, t) \) to the wave function caused by the interaction between the electrons satisfies the equation

\[
\left( -i\frac{\partial}{\partial t} - H_0(r_1, r_2, t) \right) \delta \Psi_f^*(r_1, r_2, t) = \frac{1}{r_{12}} \Psi_f^{(0)*}(r_1, r_2, t) .
\] (33)

We are to find \( \langle \delta \Psi_f \rangle \) from Eq.(33) and use it in Eq.(4) for the amplitude. This procedure is simplified due to the fact that we neglect the interaction of the ion in the ground state with the laser field. This means that we neglect the transitions from the state described by the wave function \( \phi_b(r_2) \). It makes the operator \( V_F(t) \) in Eq.(4) independent on the coordinate \( r_2 \), \( V_F(t) = -r_1 F \cos \omega t \). Thus the integration over \( r_2 \) in (4) is reduced to the projection of the final state wave function on \( \phi_b(r_2) \) which is convenient to evaluate in advance. Let us denote it as \( \Psi_f^*(r_1, t) \)

\[
\Psi_f^*(r_1, t) = \int \delta \Psi_f^*(r_1, r_2, t)\phi_b(r_2) \exp \{-iE_b t\} d^3r_2 .
\] (34)

From Eq.(33) we find that it satisfies the equation

\[
\left( -i\frac{\partial}{\partial t} + \frac{1}{2} \Delta_1 + r_1 F \cos \omega t \right) \Psi_f^*(r_1, t) = V_{cb}(r_1, t)\Phi_p^*(r_1, t) ,
\] (35)

where \( V_{cb}(r_1, t) \) is

\[
V_{cb}(r_1, t) = \exp \left\{ i \left( \int_0^t E_c(\tau) d\tau - E_b t \right) \right\} \int \phi_c^*(r_2, t) \frac{1}{r_{12}} \phi_b(r_2) d^3r_2 .
\] (36)

The solution of Eq.(33) reads

\[
\Psi_f^*(r_1, t) = \int_t^\infty dt' \int \Phi_p^*(r_1', t')V_{cb}(r_1', t')G(r_1', t'; r_1, t)d^3r_1' .
\] (37)

Here we introduce the time-dependent retarding Green function \( G(r_1', t'; r_1, t) \) describing the electron propagation in the laser field. It satisfies the equations
\[-i\frac{\partial}{\partial t} + \frac{1}{2} \Delta + rF \cos \omega t\] 
\[\left( -i \frac{\partial}{\partial t} + \frac{1}{2} \Delta + rF \cos \omega t \right) G(r', t' ; r, t) = \delta(r' - r)\delta(t' - t), \quad (38)\]
\[G(r', t' ; r, t) = 0, \quad \text{if} \quad t' < t . \]

Now from Eqs.(37),(4) we find the amplitude \(A^{(2e)}(n; p, c)\) describing the two-electron process when \(n\) quanta are absorbed, one electron occupies the final state \(p\) in the continuum and the other occupies the discrete or continuum state \(c\)

\[A^{(2e)}(n; p, c) = \frac{1}{T} \int_0^T dt \int_{t'}^\infty dt' \langle \Phi_p(t')|V_{cb}(t')G(t', t)V_F(t)|\Phi_a(t)\rangle . \quad (39)\]

Integration over the variables \(r_1, r'_1\) is presented in (39) in the symbolic form, integration over the variable \(r_2\) is performed evaluating \(V_{cb}\), see Eq.(36). Note that one can restore the symmetry with respect to permutation of the electrons if along with the matrix element \(V_{cb}\) the exchanged matrix element is considered as shown in Fig.1 b.

The energy conservation law for the process considered reads

\[E_a + E_b + n\omega = \bar{E}_p + \bar{E}_c , \quad (40)\]

where \(n\) is the total number of absorbed quanta. Eq.(11) is similar to Eq.(13) for the single-electron process and can be justified in a similar manner, see Section III.

Eq.(39) describes the ionization of the atom which is accompanied by the excitation of the ion into the discrete or continuum state. It gives the analytical expression for the diagram a in Fig.1. Let us formulate the main assumptions made in its derivation.

1.The transitions from the ion ground state caused by the laser field are neglected.
2.The wave functions in the continuum are described by the Volkov wave functions.
3.Interaction between the electrons is considered in the first order of perturbation theory.

III. FACTORIZATION OF THE AMPLITUDE

Direct calculation of the amplitude (39) is complicated due to the adiabatic nature of the process under consideration which makes the integrand exhibit a strong variation with respect to the time variables \(t, t'\). Fortunately the same adiabatic nature provides a very useful property
of the amplitude: the amplitude may be considered as a product of the single electron photo-
ionization amplitude and the amplitude of electron-ion inelastic scattering. We will call this
property factorization. For the first time this idea was considered in (Kuchiev 1987).

In order to prove factorization let us first rewrite Eq.(39) in the more convenient form given
below by Eqs.(50),(51),(53). With this purpose consider representation for the Green function
through the Volkov wave functions

\[ G(r', t'; r, t) = -i\theta(t' - t) \int \Phi_q(r', t')\Phi^*_q(r, t) \frac{d^3q}{(2\pi)^3} . \]  

(41)

This equality can easily be justified with the help of Eqs.(38),(7). Using it one can rewrite
Eq.(39)

\[ A^{(2e)}(n, p, c) = (-i)\frac{1}{T} \int_0^T dt \int dt' \int \frac{d^3q}{(2\pi)^3} \langle \Phi_p(t')|V_{cb}(t')|\Phi_q(t')\rangle\langle \Phi_q(t)|V_F(t)|\Phi_a(t)\rangle . \]  

(42)

It is important to distinguish in Eq.(42) the periodic functions of \( t' \) from the factors having
the form \( \exp\{iE't'\} \) which give the monotonic contribution to the phase. The function \( \phi_c(r_2, t') \)
defined in Eq.(27) is periodic. The last factor in the right-hand side of (26) can be presented
as

\[ \exp \left\{ i \int_0^t (E_c(\tau) - \bar{E}_c)d\tau \right\} = \exp \left\{ i\bar{E}_c t \right\} \exp \left\{ i \int_0^t (E_c(\tau) - \bar{E}_c)d\tau \right\} . \]  

(43)

Then the matrix element \( V_{cb}(r_1, t) \), see Eq.(38), can be presented as

\[ V_{cb}(r, t') = \exp \left\{ i(\bar{E}_c - E_b)t' \right\} U_{cb}(r, t') , \]  

(44)

where

\[ U_{cb}(r_1, t') = \exp \left\{ i \left( \int_0^{t'} (E_c(\tau) - \bar{E}_c)d\tau \right) \right\} \int \phi^*_c(r_2, t') \frac{1}{r_{12}} \phi_b(r_2)d^3r_2 \]  

(45)

is a periodic function of \( t' \). Similar representation for the Volkov functions (7) reads

\[ \Phi_p(r, t) = \chi_p(r, t)\exp \left\{-i\bar{E}_p t\right\} , \]  

(46)

\[ \chi_p(r, t) = \exp \left\{ i \left[ (p + k_t)r - \int_0^t (E_p(\tau) - E_p) \, d\tau + \frac{pF}{\omega^2} \right] \right\} . \]  

(47)

From (42),(44),(46),(47) we find

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\[ A^{(2e)}(n; \mathbf{p}, c) = (-i) \frac{1}{T} \int_{0}^{T} dt \int_{0}^{T} dt' \int \frac{d^3q}{2\pi^3} \langle \chi_p(t') | U_{cb}(t') | \chi_q(t') \rangle \langle \chi_q(t) | V_F(t) | \phi_a \rangle \times \exp \left\{ i \left[ (\bar{E}_p + \bar{E}_c - E_b - \bar{E}_q) t' + (\bar{E}_q - E_a) t \right] \right\} . \] (48)

Now it is useful to take into account the identity
\[ \int_{t}^{\infty} dt' \exp \left\{ iEt' \right\} f(t') = \frac{1}{T} \sum_{n=-\infty}^{\infty} \frac{1}{T} \int_{0}^{T} dt' f(t') \exp \left\{ i \left[ (E - m\omega) t + m\omega t' \right] \right\} \left[ \frac{1}{E - m\omega + i0} \right] , \] (49)
which is valid for any periodic function \( f(t) = f(t + T) \). The identity can easily be checked out with the help of Fourier expansion \( f(t) = \sum_{m} f_m \exp \left\{ -im\omega t \right\} \). Using Eq.(49) we find from Eq.(48)
\[ A^{(2e)}(n; \mathbf{p}, c) = \sum_{l} A^{(2e)}(n, l; \mathbf{p}, c) , \] (50)
where
\[ A^{(2e)}(n, l; \mathbf{p}, c) = \frac{1}{T^2} \int_{0}^{T} dt dt' \int \frac{d^3q}{2\pi^3} \frac{\langle \chi_p(t') | U_{cb}(t') | \chi_q(t') \rangle \langle \chi_q(t) | V_F(t) | \phi_a \rangle}{E_p + E_c - E_b - \bar{E}_q - (n-l)\omega + i0} \times \exp \left\{ i \left[ (E_p + E_c - E_b - E_a - (n-l)\omega) t + (n-l)\omega t' \right] \right\} . \] (51)

This formula looks similar to the usual representation for amplitudes of stationary processes. It is clear that the factor \( \langle \chi_q(t) | V_F(t) | \phi_a \rangle \) in Eq.(51) describes the matrix element of the transition of atomic electron into the continuum, while \( \langle \chi_p(t') | U_{cb}(t') | \chi_q(t') \rangle \) is the matrix element of the inelastic scattering of the photoelectron on the ion. The non-stationary nature of the process manifests itself - there are nontrivial integrations over the time variables in Eq.(51). The integration over \( t \) results in particular in the energy conservation law \( E_a + E_b + n\omega = \bar{E}_p + \bar{E}_c \), Eq.(49). To simplify notation in the following consideration the index of summation \( l \) in Eqs.(50),(51) is chosen differently from Eq.(49), \( l = n - m \). The physical interpretation of the number \( l \) comes from integration over \( t \) which gives the \( l \)-th harmonic of the matrix element of single-electron ionization. Therefore \( l \) is to be considered as the number of quanta absorbed during the single-electron ionization. Similarly, the integration over \( t' \) shows that \( n - l \) is the number of quanta absorbed in the process of inelastic collision of the photoelectron with the ion. At this stage of consideration this interpretation is not of great importance, but later, when we prove factorization, it will become essential.
Our next step is to simplify Eq.(51). With this purpose note the following. First, integration over \( t \) describes an adiabatic transition and therefore can be evaluated by the steepest descent method at the saddle points situated in the complex plane \( t \). Second, the wave function \( \chi_q(r, t) \), see Eq.(17), depends on \( q \) in a very simple way. It is an exponent of a linear form of \( q \), \( \chi_q(r, t) = \exp\{i[qR + \alpha]\} \), \( R \) and \( \alpha \) being \( q \)-independent. This allows us to reduce integration over \( d^3q \) to the residue at the pole arising due to a node of the denominator \( \bar{E}_q = \bar{E}_p + \bar{E}_c - E_b - (n - l)\omega \).

These facts make it convenient to deform the contour of integration over \( t \) into the upper semiplane of the complex plane \( t \) to bring it close to the saddle points

\[
\int_0^T \exp\{i\omega t\} \langle \chi_q(t)|V_F(t)|\phi_a\rangle dt = \int_{C_2+C_1+C_0} \exp\{i\omega t\} \langle \chi_q(t)|V_F(t)|\phi_a\rangle dt .
\] (52)

The integrand here is similar to one in the single-electron ionization problem, see Section II A. From its analysis we know that there are two saddle points of interest \( \omega t = x_1, x_2 \), see Eq.(17).

We call \( C_\sigma, \sigma = 1, 2 \) that part of the contour of integration which is situated in the vicinity of the point \( t = x_\sigma/\omega \). The integrand on the contour \( C_1 \) is the function obtained by analytical continuation from those regions of the real axes \( t \) where \( \cos\omega t > 0 \). Similarly, the integrand on the contour \( C_2 \) is the analytical function obtained by continuation from those regions of the real axes where \( \cos\omega t < 0 \).

The contour \( C_0 \) in Eq.(52) describes those parts of the contour of integration which are well separated from the saddle points and therefore give negligible contribution to the integral, \( \int_{C_0} \approx 0 \). There is no problem with the end-points \( t = 0, T \) as their contributions compensate for each other due to the periodic nature of the integrand. The saddle points \( \omega t = x_1, x_2 \) depend on \( q \), but it is convenient to choose \( C_1, C_2 \) as \( q \)-independent contours situated in the vicinity of the pole \( \bar{E}_q = \bar{E}_p + \bar{E}_c - E_b - (n - l)\omega \). We can do it because then for any given \( q \) situated close to the pole we can deform \( C_1, C_2 \) in such a way that these contours pass through the saddle points. As mentioned, the vicinity of the pole is the only important region of integration over \( q \).

Substituting Eq.(52) in Eq.(51), using the coordinate representation given in Eq.(17) for the wave functions and integrating over \( d^3q \) we find
\( A^{(2e)}(n, l; p, c) = \frac{1}{T^2} \int_{c_1+c_2}^{T} dt \int_{0}^{T} dt' \int d^3r d^3r' \chi^*_p(r', t') U_{cb}(r', t') (-rF \cos \omega t) \phi_a(r) \times \)

\[
( -\frac{1}{2\pi R} ) \exp \left\{ i \left[ K_l R + l \omega t + (n - l) \omega t' + k_r r' - k_r r - \int_{t}^{t'} \left( \frac{1}{2} k_r^2 - \frac{F^2}{4\omega^2} \right) \right] \right\},
\]

where

\[
K_l = \left[ 2 \left( l \omega - \frac{F^2}{4\omega^2} + E_a \right) \right]^{1/2}
\]

is the momentum of the photoelectron after absorption of \( l \) quanta. \( R = \sqrt{R^2} \) in Eq.\((53)\) is the function of all the variables of integration

\[
R = R(r, r'; t, t') = \int_{t}^{t'} k_r d\tau - r' + r = \frac{F}{\omega^2} (\cos \omega t - \cos \omega t') - r' + r.
\]

Eq.\((53)\) is very convenient for evaluating parameters governing the process. First of all note that integration over the variables \( r, r' \) is localised in the vicinity of the atom. Really, integration over \( r \) describes the matrix elements responsible for the ionization process, while integration over \( r' \) describes the matrix elements of inelastic collision of the photoelectron with the ion. These \( r, r' \) should be compared with the amplitude of wiggling of the electron in the laser field \( F/\omega^2 \). The latter becomes large even for quite moderate electric fields

\[
\frac{F}{\omega^2} \gg 1.
\]

Note that this inequality may be fulfilled for a big, \( \gamma > 1 \), as well as for small , \( \gamma < 1 \), adiabatic parameter. Therefore, for the fields satisfying \((56)\) we can assume that

\[
\frac{F}{\omega^2} \gg r, r'.
\]

This permits us to simplify \( R \)

\[
R \approx R_0 - \frac{R_0 \cdot (r' - r)}{R_0},
\]

\[
R_0 = R_0(t, t') = \sqrt{R^2_0},
\]

\[
R_0 = R_0(t, t') = \frac{F}{\omega^2} (\cos \omega t - \cos \omega t') .
\]

As a result the Green function in \((53)\) becomes simpler
\[
\frac{1}{R} \exp \left\{ i K_l R \right\} \approx \frac{1}{R_0} \exp \left\{ i \left( K_l R_0 + K_l \cdot (r' - r) \right) \right\}, \quad (61)
\]

\[
K_l = -K_l \frac{R_0}{R_0}. \quad (62)
\]

Substituting Eq. (61) in (53) and using Eq. (54) to rewrite the exponent we find

\[
A^{(2e)}(n,l; p, c) = -\frac{1}{2\pi T^2} \int_{C_1+C_2} dt \int_0^T dt' \langle \chi_p(t')|U_{cb}(t')|K_l + k_l|V_F(t)|\Phi_a(t) \rangle \times \exp \left\{ i \left[ K_l R_0(t,t') + \frac{1}{2} K_l^2 t + \left( \bar{E}_p + \bar{E}_c - E_b - \frac{1}{2} K_l^2 \right) t' - \int_t^{t'} \frac{1}{2} k_l^2 dt \right] \right\}. \quad (63)
\]

Let us remember that integration over \( C_1 \) is in the region where \( \text{Re}(\cos \omega t) > 0 \), while integration over \( C_2 \) is in the region where \( \text{Re}(\cos \omega t) < 0 \). For inelastic collision the main contribution to the integral over \( t' \) comes from the vicinities of the points where

\[
|\cos \omega t'| \approx 0, \quad (64)
\]

see Eq. (67) in Section IV. (This equality has a clear physical meaning. If it is fulfilled then the wiggling energy of the photoelectron is maximal, \( F^2/(2\omega^2) \), providing the best opportunity for the ion impact excitation.) Therefore we can assume \( \text{Re}(\cos \omega t) - \cos \omega t' > 0 \) when \( t \in C_1 \), and \( \text{Re}(\cos \omega t) - \cos \omega t' < 0 \) when \( t \in C_2 \). The function \( R_0(t,t') \) for complex \( t \) is to be considered as analytical continuation from the real axes. For real \( t \) the inequality \( R_0(t,t') > 0 \) is fulfilled. As a result we find

\[
R_0(t,t') = \frac{F}{\omega^2} \times \begin{cases} \cos \omega t - \cos \omega t', & \text{if } t \in C_1, \\ \cos \omega t' - \cos \omega t, & \text{if } t \in C_2. \end{cases} \quad (65)
\]

Substituting Eq. (65) in Eq. (63) we come to the final expression for the amplitude

\[
A^{(2e)}(n,l; p, c) = \frac{1}{R_l} \sum_{\sigma=1,2} A^{(2e)}_\sigma(n,l; p, c), \quad (66)
\]

\[
A^{(2e)}_\sigma(n,l; p, c) = A^{(e)}_\sigma(l; Q_{\sigma,l}) A^{(e-2e)}(n-l; Q_{\sigma,l}; p, c). \quad (67)
\]

Here \( \sigma = 1,2 \) numerates contributions of the two contours \( C_1, C_2 \) of integration over \( t \). The quantities \( A^{(e)}_\sigma \) are

\[
A^{(e)}_\sigma(l; Q_{\sigma,l}) = \frac{1}{T} \int_{\tilde{E}_\sigma} \langle \Phi_{Q_{\sigma,l}}(t)|V_F(t)|\Phi_a(t) \rangle. \quad (68)
\]
Comparing this expression with Eq. (19) we see that $A_{\sigma}^{(e)}$ are identical with the two terms in the amplitude of the single-electron ionization, see Eq. (18). The momenta $Q_{\sigma,l}$ are defined in accordance with Eqs. (60), (62), (65)

$$Q_{\sigma,l} = (-1)^{\sigma} K_l \frac{F}{\omega}, \quad \sigma = 1, 2.$$  

The quantity $R_l$ in Eq. (67) is equal to

$$R_l = \frac{F}{\omega^2} \sqrt{1 - (\beta_l + i\gamma)^2}, \quad \beta_l = \frac{K_l \omega}{F}.$$  

Evaluating this expression one can suppose that the non-exponential factor $R_0(t, t')$ in Eq. (63) is

$$R_0(t, t') \approx (-1)^{\sigma+1}(F/\omega^2)\cos x_\sigma, \quad \sigma = 1, 2,$$  

see Eqs. (64), (65). The quantity $\cos x_\sigma$ is to be found from Eq. (16) in which $p_\parallel = (-1)^{\sigma} K_l$, $p_\perp = 0$, and the sign in the right-hand side is $\pm 1 = (-1)^{\sigma+1}$ in accordance with Eq. (17). As a result the non-exponential factor is the same for both saddle points: $R_0(t = x_1/\omega, t') = R_0(t = x_2/\omega, t') = R_l$; Re $R_l > 0$, Im $R_l < 0$.

It follows from the definition of $K_l$ (54) that both amplitudes $A_{1}^{(e)}$ and $A_{2}^{(e)}$ in Eq. (67) describe the ionization process with absorption of $l$ quanta. Note however, that in the considered two-electron problem they manifest themselves differently from the single-electron problem see Eq. (18). First, each of them depends on its own argument, which is the photoelectron momentum. It is $Q_{\sigma,l}$ for $A_{\sigma}^{(e)}$, $Q_{1,l} = -Q_{2,l}$. Second, they appear in Eq. (66) with different coefficients.

The second factor in Eq. (67) is

$$A^{(e-2e)}(n - l; Q_{\sigma,l}; p, c) = -\frac{1}{2\pi T} \int_0^T \langle \Phi_p(t)| V_{cb}(t)| \Phi_{Q_{\sigma,l}}(t) \rangle dt.$$  

(71)

It can be recognized as the amplitude of electron impact on the single-charged ion in the presence of the laser field, see Eq. (78) in the next section. The initial and final momenta of the electron are $Q_{\sigma,l}$ and $p$. The collision results in the excitation of the ion into the state $c$ and absorption of $n - l$ quanta. The inequality (56) provides the following restriction on $l$

$$l \geq l_0 = \left[ \frac{F^2}{2\omega^3} \left( \gamma^2 + \frac{1}{2} \right) \right]_>, \quad \text{Eq. (72)}$$

where $[z]_>$ is a minimal natural number greater or equal to $z$, $z \leq [z]_>$. Eq. (72) has a clear physical meaning: after absorption of $l$ quanta the photoelectron must be in the continuum.
state. Otherwise, if this inequality is broken, the amplitude would be suppressed. There will appear the additional suppressing factor \( \exp\left\{-\sqrt{(l_0 - l)F/\omega^2}\right\} \) in the amplitude as it follows from Eq.(63). This factor is governed by the parameter \( \frac{F}{\omega^{3/2}} > 1 \) .

The result of this section is the following representation for the amplitude of two-electron process

\[
A^{(2e)}(n; p, c) = \sum_{l \geq l_0} \frac{1}{R_l} \sum_{\sigma = 1, 2} A^{(2e)}_\sigma(n, l; p, c).
\]  

(74)

Here every term under summation over \( l \) describes the process when first \( l \) photons are absorbed during the single-electron photo-ionization process and then \( n - l \) quanta are absorbed during inelastic collision of photo-electron with the ion. The term with \( l = l_0 \) describes the process which starts from the near-threshold single-electron ionization. The other terms with \( l > l_0 \) describe the processes when above-threshold single-electron ionization takes place. It will be shown in Section V that the role of the terms with \( l = l_0 \) and those with \( l > l_0 \) depends on the strength of the field: for a sufficiently strong field the term with \( l = l_0 \) dominates, while for a weak field the ATI processes become extremely important.

Every amplitude \( A^{(2e)}_\sigma(n, l; p, c) \) in Eq.(74) is presented in the factorization form: it is the product of the amplitudes of photo-ionization and inelastic collision, see Eq.(67). Due to largeness of the phases of the amplitudes \( A^{(e)}_\sigma(n; Q_{\sigma,t}) \) it is more convenient to take into consideration only one of them evaluating the other from the parity conservation law Eq.(20). Then Eq.(74) may be presented as

\[
\sum_{\sigma = 1, 2} A^{(2e)}_\sigma(n, l; p, c) = A^{(e)}_1(l; Q_l, c) \left[ A^{(e-2e)}(n - l; Q_l; p, c) + (-1)^l P_a A^{(e-2e)}(n - l; -Q_l; p, c) \right] = A^{(e)}_1(l; Q_l, c) \left[ A^{(e-2e)}(n - l; Q_l; p, c) + (-1)^n P_a P_b A^{(e-2e)}(n - l; Q_l; -p, P(c)) \right].
\]

(75)

Here \( Q_l = Q_{1,l} = -K_l F/F \), and \( P_b \) is the parity of the ion state \( b \). The last line in Eq.(75) is evaluated taking account of Eq.(81) which presents the parity conservation law for the inelastic scattering amplitude discussed in the next section. Eqs.(74),(75) are valid when the amplitude of the electron wiggling in the laser field is large, see Eqs.(59),(54).
IV. INELASTIC SCATTERING

Consider the inelastic electron-ion impact in the laser filed restricting attention to the aspects of the problem which are most closely related to the many-electron excitations of the atom by the laser field. The general expression for the amplitude of inelastic collision in the first order of perturbation theory over electron-ion interaction is

\[ A^{(e-2e)} = -\frac{1}{2\pi} \frac{1}{T} \int_0^T \langle \Psi_f(t)|\frac{1}{r_{12}}|\Psi_i(t) \rangle, \]  

(76)

where \(\Psi_i(t), \Psi_f(t)\) are the initial and final-state wave functions. Note that Eq.(76) differs from Eq.(4) describing the general process. In Eq.(76) both \(\Psi_i(t)\) and \(\Psi_f(t)\) account for the interaction of the system with the laser field, while in Eq.(4) only the wave function \(\Psi_f(t)\) takes this interaction into account. The difference results from the fact that Eq.(76) from the very beginning describes the perturbation theory over \(1/r_{12}\). Note also that the factor \(-1/(2\pi)\) in Eq.(76) is the usual one for a collision problem.

The wave functions for the collision problem should be chosen as

\[ |\Psi_i(t)\rangle = \Phi_p(r_1, t)\psi_b(r_2) \exp\{-iE_b t\}, \]  

(77)

\[ |\Psi_f(t)\rangle = \Phi'_p(r_1, t)\Psi_c(r_2, t). \]

Here \(p, p'\) are the momenta of the electron before and after collision and \(\psi_b, \Psi_c\) are the wave functions of the ground and excited states of the ion. Substituting Eq.(77) in (76) we find

\[ A^{(e-2e)}(m, p, p', c) = -\frac{1}{2\pi} \frac{1}{T} \int_0^T \langle \Phi_{p'}(t)\rangle V_{cb}(t)|\Phi_p(t) \rangle dt = \]  

(78)

\[-\frac{1}{2\pi} \frac{1}{T} \int_0^T \langle p', \phi_c(t) \frac{1}{r_{12}} p, \phi_b(t) \rangle \exp \left\{ \frac{i}{\omega} S_{sc}(\omega t) \right\} dt, \]

where

\[ S_{sc}(x) = \int_0^x \left[ \frac{1}{2} \left( p' + \frac{F}{\omega} \sin x \right)^2 + E_c(x/\omega) - \frac{1}{2} \left( p + \frac{F}{\omega} \sin x \right)^2 - E_b \right] dx + \frac{(p - p')F}{\omega}. \]  

(79)

Eq.(78) demonstrates that the previously considered in Eq.(71) quantity is the amplitude of inelastic collision indeed. The energy conservation law for the case under consideration reads

\[ \frac{p^2}{2} + E_b + m\omega = \frac{p'^2}{2} + E_c, \]  

(80)
where $|m|$ is the number of quanta which are absorbed, if $m > 0$, or emitted, if $m < 0$, during the scattering. The parity conservation law for the inelastic scattering reads

$$A^{(e-2e)}(m, -p; -p', P(c)) = (-1)^m P_b A^{(e-2e)}(m, p; p', c),$$  \hspace{1cm} (81)

where $P_b$ is the parity of the state $b$, and $P(c)$ is the state with the quantum numbers obtained by applying the inversion operator $P$ to the quantum numbers of the state $c$. It is easy to verify using Eqs.(10),(29) that the amplitude Eq.(78) satisfies Eq.(81).

As is usual in adiabatic processes there appears the large phase $\sim 1/\omega$ in the integrand in Eq.(78). Therefore the saddle points give the major contribution to $t$-integration. They satisfy the condition

$$S_{sc}'(\omega t) = \frac{1}{2} \left( \frac{p' + F}{\omega} \sin \omega t \right)^2 + E_c(t) - \frac{1}{2} \left( \frac{p + F}{\omega} \sin \omega t \right)^2 - E_b = 0.$$  \hspace{1cm} (82)

It is fulfilled for those moments of $t$ when the time-dependent “energy” of the system in the initial state coincides with the “energy” in the final state. If there is a real saddle point $t$ then the process proceeds with a high probability. If it is in the complex plane, $\text{Im} \ t > 0$, then there is a strong suppression originated from the adiabatic nature of the process. This statement can be checked out for special cases when $t$-integration in Eq.(78) can be fulfilled analytically. One of them is the excitation into the discrete ion state $c$ having small polarizability. Then one can neglect the time-dependence of the energy $E_c(t)$ and the corresponding wave function $\phi_c(t)$, $E_c(t) \approx E_c$, $\phi_c(t) \approx \phi_c$ and find from Eq.(78) the simple representation

$$A^{(e-2e)}(m, p; p', c) = f(p; p', c) J_m(qF/\omega^2) i^m,$$  \hspace{1cm} (83)

$$f(p; p', c) = -\frac{1}{2\pi} \langle p', \phi_c | \frac{1}{r_{12}} | p, \phi_b \rangle,$$  \hspace{1cm} (84)

where $f(p; p', c)$ is the scattering amplitude in the absence of the laser field (considered outside the mass-shell), $J_m(z)$ is a Bessel function and $q = p - p'$ is the transferred momentum. Representation similar to Eq.(83) is well-known for the case of elastic scattering considered in (Bunkin and Fedorov 1966). A Bessel function is large when its argument is greater than its index. Thus the amplitude Eq.(83) is not suppressed if $|qF|/\omega^2 \geq |m|$. The latter condition is equivalent to one given in Eq.(82) for the considered case $E_c(t) = E_c$. 

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Consider now how much energy can be transferred to the ion from the electron and the field during collision. Suppose that the initial-state momentum $p$ is given. The final-state moment $p'$ will be considered as a free parameter and we will choose it to make the energy level $E_c$ as high as possible. From Eqs.(80),(82) we find that to make the process probable the following inequality is to be fulfilled for some real moment of time $t$

$$\frac{1}{2}(p + k_t)^2 \geq E_c(t) - E_b.$$  (85)

The quantity in the left-hand side of Eq.(85) may be considered as a time-dependent “energy” of the impact electron. To clarify the meaning of this equation consider first the discrete-state excitations of the ion. For the discrete state $c$ we can use Eq.(28) to estimate its energy. Then from Eq.(85) we find the maximum energy gap between the ion ground state $b$ and its excited state $c$ which can be populated in the collision

$$\max(E_c - E_b) = \frac{1}{2} \left( |p||| + \frac{F}{\omega} \right)^2 + \frac{1}{2} p_{\perp}^2.$$  (86)

Evaluating Eq.(86) it was assumed that $\alpha_c(\omega)\omega^2 \leq 1$. Note that in the left-hand side of Eq.(86) there stands the non-shifted value of the energy $E_c$. The moments of time $t$ for which the transition into the state $c$ satisfying Eq.(86) takes place are those for which the wiggling energy is maximal

$$|\sin \omega t| = 1.$$  (87)

Then we find that final-state momentum $p'$ is to be

$$|p'|| \approx \frac{F}{\omega}, \quad p_{\perp}' \approx 0.$$  (88)

Consider now the excitations of the ion into continuum state, i.e. creation of a doubly charged ion. For this case $E_c(t) = (p_1 + k_t)^2/2$, where $p_1$ is the momentum of the knock-out electron. It is interesting to find the maximum binding energy of the ion $E_b$ which would still allow the collision to result in double ionization. From Eq.(85) we find

$$\max|E_b| = \frac{1}{2} \left( |p||| + \frac{F}{\omega} \right)^2 + \frac{1}{2} p_{\perp}^2.$$  (89)
which agrees with Eq.(86) if we put in the latter one $E_c = 0$. Conditions (87),(88) are valid for this case as well. Eq.(87) was used previously to evaluate the quantity $R_l$ in Eq.(70).

Note that according to Eq.(86) the maximum possible excitation energy of the ion is equal to the maximum of the “energy” of the impact electron. The latter is always higher, and can be much higher, than the averaged energy which stands in the energy conservation law: $\max(\mathbf{p} + \mathbf{k}_t)^2/2 > \bar{E}_p$. This fact will be important in the following estimation of the probability of many-electron processes given in Section VI.

V. QUALITATIVE DISCUSSION

In the previous sections we considered the collective response of two atomic electrons to the laser field and found that the amplitude of the process may be presented as a product of the amplitude of single-electron ionization and the amplitude of inelastic scattering, see Eq.(67) or (75). This means that at the beginning of the process single-electron ionization takes place. After that the ionized electron undergoes inelastic collision with the parent atomic particle. The general reason for factorization of the amplitude is quite clear. It follows from the existence of two different scales for electron localization. One is the radius of the atom. The other is the radius of the wiggling of the electron in the laser field which is supposed to greatly exceed the atomic radius.

The simple physical meaning of the mathematical description presented above is the following. The single-electron ionization may be considered as a quasiclassical movement of the atomic electron under the non-stationary, time-dependent barrier. When the tunneling is finished the electron appears from under the barrier at some particular point separated from the atom. After that the electron propagates in a classically allowed region. It strongly interacts with the laser field exhibiting the wiggling. The propagation of the electron wave function in the classically allowed region may proceed in any direction because it starts from the point. In particular it propagates in the direction of the parent atomic particle. This makes possible the return of the electron to the single-charged ion. As a result the collision of this electron with the ion is very probable, the probability being proportional to the cross section and inversely
proportional to the square of the distance which separates the electron from the atom after finishing the under-barrier part of the trajectory. This distance is equal to the absolute value of $R_l$ given in Eq. (70)

$$|R_l| = \frac{F}{\omega^2} \left\{ ((\beta_l + 1)^2 + \gamma^2)((\beta_l - 1)^2 + \gamma^2) \right\}^{1/2}; \quad \beta_l = \frac{K\omega}{F}, \quad \gamma = \frac{\kappa\omega}{F}. \quad (90)$$

It depends on the amplitude of the electron wiggling $F/\omega^2$ as well as on $\gamma$ and the parameter $\beta_l$ which is the ratio of the photoelectron momentum to the field momentum. The photoelectron momentum depends on the number of absorbed quanta, which makes $|R_l|$ depend on it as well. This separation $|R_l|$ can exceed the amplitude $F/\omega^2$ of the wiggling. For this case wiggling itself is not sufficient to prove that the electron can return to the parent atomic particle. That is why the discussion of all mathematical aspects of the problem given in the previous section is vital.

There are two paths for the under-barrier electron escape from the atom. Roughly speaking, one of them is along the direction of the field $\mathbf{F}$ and the other is in the opposite direction. (The trajectory is described by complex time and coordinates. Therefore, it is more accurate to say that the real part of the trajectory is either along the field or in the opposite direction.) Every term $A_{\sigma}^{(e)}$, $\sigma = 1, 2$ in the amplitude of single-electron ionization, see Eq. (18), describes the escape along one of these two paths. If we consider the pure single-electron ionization problem then after the tunneling the momentum of the electron should be the same for both routes of the escape. In contrast, for the problem of two-electron ionization the electron must return to the parent atomic particle and have the collision with it. The return route is in the direction opposite to the escape path, being different for the events described by $A_1^{(e)}$ and for those described by $A_2^{(e)}$. This explains why in Eq. (67) the momentum $Q_{1,l}$, which is the argument of $A_1^{(e)}$, is opposite to the field direction while the momentum $Q_{2,l}$, which is the argument of $A_2^{(e)}$, is along this direction.

**VI. ESTIMATION OF THE PROBABILITY OF MANY-ELECTRON PROCESS**

There are different ways for the process of two-electron excitation with absorption of $n$ quanta. During ionization of the first electron there may be absorbed some number $l$ of quanta.
The remaining number of quanta $n - l$ is absorbed in the collision. The two-electron amplitude is the sum of the amplitudes of these independent events, see Eq. (74). The interference of the terms with different $l$ should be suppressed by the large and field-dependent phase of the amplitude $A_1^{(e)}$, see discussion in Section II A. Therefore as a first approximation we can neglect this interference. Then we find from Eq. (74) the simple estimation for the probability $W^{(2e)}(c)$ of the two-electron process when one electron in the final state is in the continuum and the other is excited into the state $c$ belonging to the discrete or continuum spectrum

$$W^{(2e)}(c) = \sum_n W^{(2e)}(n; p_n, c),$$

$$W^{(2e)}(n; p_n, c) = \sum_{l \geq l_0} dW^{(e)}(l, \theta = 0) \frac{\sigma(n - l; K_l; p_n, c)}{|R_l|^2}. \quad (92)$$

Here the total probability $W^{(2e)}(c)$ is presented as a sum of the probabilities with absorption of a given number $n$ of quanta $W^{(2e)}(n; p_n, c)$. The quantity $dW^{(e)}(l, \theta = 0)/d\Omega$ is the differential probability of the single-electron ionization with absorption of $l$ quanta, and transition of the electron into the state with momentum $K_l$ whose absolute value is given in Eq. (54), the condition $\theta = 0$ shows that the considered momentum direction is along the field, $K_l = K_l F/F$. The quantity $\sigma(n - l; K_l; p_n, c)$ in Eq. (92) is the cross section of the electron-ion inelastic scattering in the presence of the laser field, $K_l$ and $p_n$ are the momenta of the electron in the initial and final states for the scattering problem. The scattering is accompanied by the absorption of $n - l$ quanta and excitation of the ion into the state $c$. Evaluating Eq. (92) we took into account Eq. (21) for the probability of the single-electron event and for the sake of simplicity neglected the interference of the two terms in the square brackets in Eq. (73). The distance $|R_l|$ is given in Eq. (90).

Consider now how high is the excitation energy provided by the considered mechanism. The general rules showing that the excitation is probable are given by Eqs. (86), (89) in which the impact electron momentum for the considered case is $|p|| = K_l$, $p_\perp = 0$. As a result we come to Eq. (1)

$$\frac{1}{2} \left( K_l + \frac{F}{\omega} \right)^2 \geq E_{exc}, \quad (93)$$

which it is convenient to present as
\[ \beta_l + 1 \geq \gamma_{exc}, \quad \gamma_{exc} = \frac{\omega \sqrt{2E_{exc}}}{F}, \quad (94) \]

where the quantity \( \gamma_{exc} \) may be called the adiabatic parameter for the considered ion excitation.

The energy \( E_{exc} \) is introduced to consider simultaneously the ion excitations into discrete and continuum spectra: \( E_{exc} = E_c - E_b \) for discrete excitations and \( E_{exc} = |E_b| \) for excitations into the continuum, compare Eqs.(86),(89).

Eq.(93) demonstrates that the first ionized electron must possess “enough energy” to excite the ion. Due to wiggling in the laser field the “energy” of the electron is not conserved, it oscillates. Condition (93) states that at the most favourable moment of time, when the field momentum is big, \( k_t = F/\omega \), and parallel to the translational momentum \( K_l \), this energy must be equal to the excitation energy. Note that the averaged energy of the photoelectron is always lower than the maximum one: \( (K_t + F/\omega)^2/2 > E_{K_c} = K_l^2/2 + F^2/(4\omega^2) \). For further consideration it is convenient to distinguish the case of high wiggling energy from the case of low wiggling energy. Consider them in turn.

1. High wiggling energy.

Suppose that the energy of the electron wiggling is so high that it exceeds the ion excitation energy for the considered state \( c \), \( F^2/(2\omega^2) \geq E_c - E_b \), i.e.

\[ \gamma_{exc} \leq 1. \quad (95) \]

Then it is sufficient to take into account only one first term \( l = l_0 \) in the sum over \( l \) in Eq.(92). It describes the process when first the near-threshold single-electron ionization takes place. The momentum of photoelectron is small \( K_{l_0} \approx 0 \). The electron wiggling provides enough energy to excite the ion during collision as is seen from the conditions (94),(95). The probability of the process may be estimated as

\[ W^{(2e)}(c) \approx \frac{dW^{(e)}}{d\Omega} (l_0, \theta = 0) = \frac{\sigma(n - l_0; K_{l_0}; p_n, c)}{|R_n|^2}. \quad (96) \]

Eq.(90) for the considered case reduces to

\[ |R_{l_0}| = \frac{F}{\omega^2} \sqrt{1 + \gamma^2}. \quad (97) \]
2. Low wiggling energy.

Suppose that the wiggling energy is small compared with the excitation energy, \( F^2/2\omega^2 < E_c - E_b \),

\[
\gamma_{\text{exc}} > 1 .
\]  \( (98) \)

Then the wiggling energy \( F^2/(2\omega^2) \) is insufficient to provide the ion excitation. Therefore in this case we are to consider ATI during the single-electron ionization described by the terms \( l > l_0 \) in the sum in Eq.(92). Let us call \( l_1 \) the minimum number of quanta the absorption of which satisfies Eq.(93). From Eqs.(93),(74) we find

\[
l_1 = \left[ \frac{F^2}{2\omega^3} (\gamma_{\text{exc}} - 1)^2 + \gamma^2 + \frac{1}{2} \right] > l_0 = \left[ \frac{F^2}{2\omega^3} (\gamma^2 + \frac{1}{2}) \right].
\]  \( (99) \)

The inequality (94) shows that for a very weak field when \( \gamma_{\text{exc}} \gg 1 \), the above threshold energy must be equal to the ion excitation energy: \( K_{l_1}^2/2 \approx E_c - E_b \). In this case \( l_1 \) can substantially exceed \( l_0 \). With increase of the field the necessary above threshold energy rapidly decreases. Really, it is seen from Eq.(93) that if \( \gamma_{\text{exc}} \sim 1 \), then \( l_1 \sim l_0 \). In this case the purpose of ATI is to supply the ionized electron with the momentum \( K_l \) which combined with the large field momentum will give the necessary high maximum “energy” for the photoelectron, see Eq.(93). At the same time its averaged energy may be much lower than the ion excitation energy. The example of two-electron process in He considered in Section VII illustrates this possibility. We come to a conclusion that there are two reasons for increase of the probability of the two-electron process with increase of the field in the region Eq.(98). First, the total probability of single-electron ionization grows. Second, the stronger the field the lower becomes the necessary level in the ATI spectrum.

The estimation for the probability of two-electron process is

\[
W^{(2e)}(c) \approx \frac{dW^{(e)}(l_1, \theta = 0)}{d\Omega} \sigma(n - l_1; K_{l_1}; p_n, c) \left| R_{l_1} \right|^2 ,
\]  \( (100) \)

where \( R_{l_1} \) is given in Eq.(90).

The total number of absorbed quanta \( n \) in the processes described by Eqs.(90),(100) must be large enough to provide the excitation of the level \( c \) as well as a sufficiently large final state.
momentum $p_n$ of the photoelectron: $p_n \approx F/\omega$, see Eq.(88). Therefore, the averaged energy of the scattered electron is $E_{p_n} \approx 3F^2/(4\omega^2)$. Estimation for the number of absorbed quanta $n$ follows from the conservation law

$$n \approx n_{\min} + \left[ \frac{F^2}{2\omega^3} \right], \quad n_{\min} = \left[ \frac{1}{\omega} \left( E_c - E_a - E_b + \frac{F^2}{4\omega^2} \right) \right] = \left[ \frac{F^2}{2\omega^3} \left( \gamma_{\text{exc}}^2 + \gamma^2 + \frac{1}{2} \right) \right].$$

(101)

VII. DOUBLE-IONIZATION OF HE ATOM BY 780 NM LASER FIELD

In order to illustrate the ideas discussed in the previous section consider the recent experimental results (Walker et al 1994) on precision measurement of double ionization of He atom by the 780 nm laser field in the intensity region from 0.15 to 5.0 PW/cm$^2$. The data show the high ratio of He$^{++}$ to He$^+$ yield in this region of intensities. For the lowest intensity 0.15 PW/cm$^2$ the field momentum is $F/\omega = 1.2$ atomic units. Therefore the maximal wiggling energy $F^2/(2\omega^2) = 19.3$ eV is well below the lowest excitation energy of He$^+$ which is 41 eV. Absorption of only 3 quanta above the single-electron ionization threshold changes the situation drastically. It gives the translational momentum $K = 0.58$. As a result the maximal energy of photoelectron becomes $(K + F/\omega)^2/2 = 42.8$ ev. The above consideration, see Eq.(108) or (1), shows that this maximal energy of photoelectron can be used for ion excitation. Thus we see that absorption of only 3 above-threshold quanta during the single-electron ionization makes the excitation of He$^+$ possible. The excitation might result in the double charged ion formation due to field ionization from the excited state. Note that the above-threshold excitation energy itself in the considered example is sufficiently low. After absorption of 3 quanta it is only $\sim 5$ eV which is far below the excitation energy. This illustrates that the role of ATI is to supply the ionized electron with the momentum, rather than with above-threshold energy in ATI spectrum. Similarly, the absorption of 6 above-threshold quanta makes possible the direct excitation of the ion into the continuum, i.e. double-ionization. The probability of absorbing several quanta above the threshold is high for the considered region of intensities of the laser field, see for example the recent work (Schumacher et al 1994) where ATI for the first and sec-
ond harmonics of 1064 nm laser field with total intensity of $4 \times 10^{13}$ and $8 \times 10^{13}$ W/cm$^2$ was examined. Thus the proposed mechanism qualitatively agrees with the experimental results.

**VIII. CONCLUSION**

We considered the mechanism of the many-electron process when first single-electron ionization takes place and then the ionized electron absorbs energy from the laser field and transfers it to the parent atomic particle. We proved by explicit calculations that this mechanism is very probable and provides the high energy necessary for ion excitation. The amplitude of the two-electron process is presented as a product of the amplitude of single-electron ionization and the amplitude of inelastic scattering. Both later quantities can be calculated *ab initio* as discussed in the Sections II A and IV. Therefore the presented results provide a possibility for accurate calculations of many-electron processes in the laser field. The simple estimations for the probability of the process are also possible as discussed in Sections VI,VII. ATI proves to be very important for the mechanism considered. It supplies the ionised electron with an additional translational momentum. The latter gives sharp rise to the maximal energy of the photoelectron resulting in the possibility of excitation of the high-energy levels of the ion.

The results obtained can be developed to cover related phenomena. One of them is ATI itself. We have seen that the scattering of the ionised electron on the parent atomic particle results in the transformation of its maximum energy into the excitation of the ion. Another possibility is to transform the maximum total energy into the energy of translational movement. This would strongly increase the probability of the population of high-energy ATI levels. For the first time this idea was discussed in (Kuchiev 1987). Another related problem is the high-energy gamma-quanta production by an atom in a laser field.

In this paper we described the continuum states with the help of the Volkov functions. This description needs improvement, as the static Coulomb field in some cases is important, see for example (Reiss and Krainov 1994). One can expect this improvement to modify the amplitude of single-electron ionization and the scattering amplitude. It can also modify the parameter $R_l$ describing the separation between the ion and first ionised electron. However, factorization as a
general property should remain valid. The reason for this comes from the fact that factorization is based on the parameter which is the large amplitude of classical wiggling of the electron in the laser field. For a sufficiently strong field this quantity can remain large enough even if one takes into account the ion Coulomb field.

The other approximation made in the paper is the consideration of only the first order of perturbation theory over the electron interaction. This approximation can be insufficient if we are interested in the absorption of the very large number of quanta. The reason for this is that the electron impact transforms electron energy into the ion excitation, but there is always a possibility for the scattered electron to absorb more energy from the field and again transfer it to the ion during one more collision with it.

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