Quantum Theory of a High Harmonic Generation as a Three-Step Process

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Abstract

Fully quantum treatment explicitly presents the high harmonic generation as a three-step process: (i) above threshold ionization (ATI) is followed by (ii) electron propagation in a laser-dressed continuum. Subsequently (iii) stimulated (or laser assisted) recombination brings the electron back into the initial state with emission of a high-energy photon. Contributions of all ATI channels add up coherently. All three stages of the process are described by simple, mostly analytical expressions that allow a detailed physical interpretation. A very good quantitative agreement with the previous calculations on the harmonic generation by H$^-$ ion is demonstrated, thus supplementing the conceptual significance of the theory with its practical efficiency. The virtue of the present scheme is further supported by a good accord between the calculations in length and velocity gauges for the high-energy photon.

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I. INTRODUCTION

Under the influence of an intensive electromagnetic field an atom can emit electrons and photons. The number of photons absorbed from the field in the first process generally can exceed the minimum necessary for ionization resulting in distribution of the photoelectrons over the above threshold ionization (ATI) channels. The photon production manifests itself as the harmonics generation (HG) for the incident monochromatic laser radiation. Both ATI and HG are capable of populating the channels with remarkably high energy, as has recently been registered in experiments (see, e.g., Ref. [1–5]) and tackled by the theory [6–23] (the list of references is unavoidably incomplete, for more bibliography see the reviews [24,25]).

An idea that the two processes referred above are interrelated was articulated long ago. Since in the HG process an active electron ends up in the initial bound state, it is appealing to represent it as ionization followed by recombination. This mechanism presumes a strong interaction between the emitted electron and the core that is omitted in the standard Keldysh [26] model of multiphoton ionization. The importance of this interaction was first pointed out by Kuchiev [28], who predicted several phenomena for which the electron-core interaction plays a crucial role. The related mechanism was named “atomic antenna”.

Specifically for HG, the simple relation between this process and ATI was suggested by Eberly et al [6] but proved to be non-realistic, see below. The hybrid classical-quantum model due to Corkum [8] (see also the paper by Kulander et al [10]) casts HG as a three-step process: tunneling ionization and subsequent propagation in the continuum is completed by recombination. This intuitive model has influenced many research in experiment and theory. The simplicity of the model is due to some drastic presumptions. Usually it is emphasized that the intermediate electron propagation in the laser field is described by the Corkum [8] model classically. Probably less attention is paid to the fact that neither the tunneling ionization through the time-dependent barrier, nor the laser-stimulated recombination receive a genuine quantum treatment as well. Being successfully applied to the comparison with some experimental data, the model resorts to such a loosely defined free parameter as the ‘transverse spread of the electron wave function’. From the conceptual side the Corkum [8] model does not appeal to ATI process just because the discrete ATI channels do not appear within the classical framework. The subsequent theoretical developments were based on more sophisticated approaches and led to important advancements [11–13,15], but apparently abandoned a perspective to establish a quantitative relation between ATI and HG. The ATI characteristics merely do not emerge in the papers devoted to HG theory, with few exceptions [14,16]. For instance, Ref. [20] establishes what the authors think to be ‘the most general formal relation between ionization and high harmonic generation’ that contains functional derivative of ‘the ground state persistence amplitude’ $Z$ but not ATI amplitude. As the authors recognize, this relation is ‘of limited practical use’ since it requires knowledge of $Z$ for arbitrary electric field $F(t)$.

Theoretically HG is governed by the Fourier components in the field-dresses wave function. The proper description of the high-order components is a challenging task for the theory. Although some important features could be understood in classical calculations, the significance of fully quantum quantitative theory could not be overestimated. The computer-intensive numerical studies achieved substantial progress [7,9], but the available computer facilities often limit them to the one-dimensional models [8,21,22]. They could
be favorably complemented by analytical studies capable of providing an important physical insight. However, with the two different analytic quantum approaches being developed recently \[12,15\], the problem could not yet be considered as closed. For instance, as far as we know, these two approaches have never been applied to the same system in order to provide quantitative comparison of their results, albeit the differences in the formulation were indicated by Becker et al \[15\]. The only exception known to us is provided by Ref. \[20\] where in Fig. 6 some comparison is presented, albeit not for such a basic characteristics as HG rates, but for harmonic ellipticity and offset angle that reflect rather subtle peculiarities of HG process. For the offset angle a severe disagreement between the two theories is demonstrated. The lack of comparison looks quite typical to the current state of theory when the attention is mostly directed towards the qualitative aspects of the problem such as description of the pattern of emitted harmonic spectrum.

The principal objective of the present study is to derive a fully quantum formulation for the HG amplitude in terms of the ATI amplitude and the amplitude of electron laser assisted recombination (LAR) in the laser field. Importantly, all the amplitudes are physical, i.e. no off-energy-shell entities appear. This circumstance adds to the conceptual appeal of the present theory its significance as a true working tool. In our approach, briefly described in the Letter to Editor \[27\], we base on the ideas outlined \[28\] and later developed in more detail \[29,30\] by Kuchiev. We successfully test its efficiency by comparison with the benchmark calculations by Becker et al \[15\] for HG by H\(^-\) ion. It should be emphasized that we test and achieve quantitative agreement for absolute values of HG rates, in contrast to many theoretical works which concentrate mostly on the qualitative issues. The quantitative character of our development is further illustrated by comparative calculations within the dipole-length and dipole-velocity gauges for the emitted high-energy photons: the good agreement testifies in favor of the method. In the broader perspective it should be emphasized that our theoretical technique is directly applicable to other processes of current key interest, such as multiple ionization by laser radiation or enhanced population of high ATI channels due to the photoelectron rescattering on the atomic core.

We start (Sec.II) with exposure of the general relations and outline our basic approximations. In Section III we cast the harmonic generation amplitude as a sum over contributions coming from different ATI channels. The form and interpretation of this representation becomes particularly transparent when we resort to the adiabatic approximation (Sec.IV). The general theory is illustrated by the quantitative results in Section V that is followed by the concluding discussion (Sec.VI).

II. FORMULATION OF THE PROBLEM AND BASIC APPROXIMATIONS

A. General relations

The generation of a harmonic with the frequency \(\Omega\) is governed by the Fourier transform of the dipole transition matrix element

\[
D(\Omega) = \int_{-\infty}^{\infty} dt \ \exp(i\Omega t) \ d(t) ,
\]

\[
d(t) = \int d^3r \ \Psi_f(r, t)^* \hat{d}\epsilon \ \Psi_i(r, t) , \quad \hat{d}\epsilon = \epsilon \cdot r ,
\]
where $\Psi_i$ and $\Psi_f$ are initial and final states of the atomic system dressed by the laser field (atomic units are used throughout the paper).

We construct the initial field-dressed state $\Psi_i$

$$
\Psi_i(r, t) = \Phi_a(r, t) + \int_{-\infty}^{t} dt' \int d^3r' G(r, t; r', t') V_F(r', t') \Phi_a(r', t'),
$$

(2.3)
developed out the initial field-free stationary state $\Phi_a$

$$
\Phi_a(r, t) = \varphi_a(r) \exp(-iE_at),
$$

(2.4)
$$
H_a \varphi_a(r) = E_a \varphi_a(r),
$$

(2.5)
using the retarded Green function $G(r, t; r', t')$ which obeys equation

$$
\left[i \frac{\partial}{\partial t} - H_a - V_F(t)\right] G(r, t; r', t') = \delta(t - t') \delta(r - r') \quad (t > t'),
$$

$$
G(r, t; r', t') = 0 \quad (t < t'),
$$

(2.6)
where $H_a = \frac{1}{2} \hat{\mathbf{p}}^2 + V_a(r)$ is the Hamiltonian of an atomic system in the single active electron approximation, $V_a(r)$ is the interaction with the core, $V_F(r, t)$ is the interaction with the laser field that generally includes the field-switching effects [below we presume that $V_F(T)$ is real].

We consider harmonic generation when the atomic system ends up in the initial state. Other final states are also feasible, but till now this possibility have not been explored neither in the experiment, nor in the theory. In this case the final state tends to $\Phi(t)_a$ for $t \to \infty$ being presented similarly to (2.3) as

$$
\Psi_f(r, t)^* = \Phi_a^*(r, t)^* + \int_{t}^{\infty} dt' \int d^3r' \Phi_a(r', t')^* V_F(r', t') G(r', t'; r, t),
$$

(2.7)
Employing (2.3) and (2.7) we transform $d(t)$ to

$$
d(t) = \langle \Phi_a(t) | \hat{d}_\varepsilon | \Phi_a(t) \rangle + \int_{-\infty}^{t} dt' \langle \Phi_a(t) | \hat{d}_\varepsilon G(t, t') V_F(t') | \Phi_a(t') \rangle + \\
+ \int_{t}^{\infty} dt' \langle \Phi_a(t') | V_F(t') G(t', t) \hat{d}_\varepsilon | \Phi_a(t) \rangle + \\
+ \int_{t}^{\infty} dt'' \int_{-\infty}^{t} dt' \langle \Phi_a(t'') | V_F(t''; t') G(t', t) \hat{d}_\varepsilon G(t, t') V_F(t') | \Phi_a(t') \rangle,
$$

(2.8)
where $\langle \ldots | \ldots | \ldots \rangle$ notation is employed to represent integration over the space variables. This formula could be compared with Eq. (2.15) in Ref. 20 obtained under presumption $\Psi_f \equiv \Psi_i$. Although the general structure is similar, the important difference lies in the range of temporal integration. In formula (2.8) the first term in the right hand side turns zero for the inversion-invariant (for instance, spherically symmetric) potentials. Physically the second term describes the process when the high harmonic photon is emitted after the interaction with the laser field, $t \geq t'$, whereas the third term describes the 'time-reversed' event in which the radiation precedes the absorption of laser quanta. We denote the second and the third terms respectively as $d^+(t)$ and $d^-(t)$,
\[
d^+(t) = \int_{t'}^t dt' \langle \Phi_a(t) | \hat{d}_\epsilon G(t, t') V_F(t') | \Phi_a(t') \rangle ,
\]
\[
d^-(t) = \int_t^{\infty} dt' \langle \Phi_a(t') | V_F(t') G(t', t) \hat{d}_\epsilon | \Phi_a(t) \rangle .
\]

The last term in (2.8) includes the effect usually referred to as \textit{continuum-continuum transitions} \[11,12,20\]. In the present outlook it corresponds to the “mixed” picture when a part of laser photons is absorbed prior to emission of the harmonic followed by absorption of missing low-energy quanta.

In this paper the continuum-continuum transitions are omitted as a rather standard approach, apparently assumed originally in Refs. \[11,12\], albeit never scrutinized. There exists a simple physical reason why this term should not be important. The absorption of large number of low-frequency quanta happens when the active electron is well separated from an atom, see discussion of the role of large distances in \[33,34\]. In contrast, one should anticipate that emission of the high-energy quantum occurs when the electron is localized close to the atom. A transition of the electron from the outer region to the vicinity of the atom inevitably produces a suppression factor that describes the electron propagation. Later on it will be considered in detail, see the factor \(1/R\) in (4.14). For a \textit{natural sequence} of events, when electron first absorbs \(N\) laser quanta and then emits the high-harmonic this suppression factor appears only once. For continuum-continuum transitions the electron has to go from the outer region into the vicinity of the atom thrice, that induces appearance of three suppression factors, thus substantially reducing the amplitude. Technically the explicit calculation of the continuum-continuum contribution means substantially higher level of difficulty, since it contains two Green functions; it is not pursued in the present study. An alternative, albeit indirect justification for omission of continuum-continuum contribution could be seen in the fact that our calculations of HG rates within this approximation are in a good \textit{quantitative} agreement (Sec. V) with the results obtained by Becker \textit{et al} who apparently do not rely on it. One more argument in favor of this approximation stems from the fact that it provides a very good agreement between the results in length and velocity gauges, as detailed in Sec. V.

Further we presume that the monochromatic laser field

\[
V_F(r, t) = \mathbf{r} \cdot \mathbf{F} \cos \omega t
\]

is switched on adiabatically at some remote time (\(\mathbf{F}\) is the amplitude of the electric field strength in the laser wave). In this case one can easily check that \(d^-(t) = d^+(t)\). After dropping in the right hand side of (2.8) the last term, that describes the continuum-continuum transitions, we arrive to the formula

\[
d(t) = d^+(t) + d^+(t) .
\]

One should remember that in our calculations according to (2.3) and (2.7) we employed \textit{different wave functions} \(\Psi_i\) and \(\Psi_f\). Unfortunately, there is a strong trend in the literature \[6,11,12,18,31–34\] (to cite only part of references) to presume that \(\Psi_i \equiv \Psi_f\) that leads to replacement of the relation (2.11) by

\[
d(t) = d^+(t) + (d^+(t))^* ,
\]
and, consequently, to the real-valued dipole momentum \(d(t)\).

Contrary to this almost universal delusion the expression (2.12) is incorrect for description of harmonic emission by a single atom and the dipole momentum \(d(t)\) is not real-valued, both in the exact formulation (2.8) and within the approximation neglecting the continuum-continuum transitions (2.11). The recent paper on the unified theory of harmonic generation by Becker et al. \([20]\) describes two different approaches. The calculations of the dipole-moment expectation presume \(\Psi_f \equiv \Psi_i\) and lead to the real-valued \(d(t)\) [see Eqs. (2.15) and (2.16) in \([20]\)], whereas the \(S\)-matrix approach accounts for the distinction between \(\Psi_f\) and \(\Psi_i\) and provides a similar formula (2.30) but with a different range of temporal integration [cf. our discussion below Eq. (2.8)]. The mentioned formula (2.30) gives complex-valued results. We agree with the authors comment that the \(S\)-matrix approach is to be employed when the harmonic emission of a single atom is considered; namely this process is the subject of our present study. However, Ref. \([20]\) as well as Refs. \([13,14]\) carry out all calculations for the dipole moment expectation presuming that just this quantity is required as a source term for the integration of the Maxwell equations when the emission by the medium is considered. We believe that the theory of collective emission should be ultimately based on the proper description of a single atom process, but further discussion of this issue is beyond the scope of the present paper.

For the high harmonic generation the 'time-reversed' process described by \(d^-(t) = d^+(−t)\), i.e. emission of high harmonics followed by absorption of a large number of the laser quanta, is strongly suppressed as compared with the "natural" sequence of events represented by \(d^+(t)\), i.e. when at first a large number of laser quanta is gained and subsequently one high-frequency photon is emitted. Therefore the further approximation of Eq. (2.11)

\[
d(t) = d^+(t)
\]

should work well. Below (Sec.\(\text{V}\)) we demonstrate by explicit calculation of the Fourier components that indeed the term \(d^-(t)\) gives negligible contribution.

For monochromatic laser filed (2.10) \(d(t)\) is a periodic function of time with the period \(T = 2\pi/\omega\) where \(\omega\) is the laser frequency. Introducing the Fourier transform

\[
d_N = \frac{1}{T} \int_0^T dt \exp(iN\omega t) d(t)
\]

we see that

\[
D(\Omega) = 2\pi \sum_N \delta(\Omega - N\omega) d_N .
\]

This stationary picture does not account for the depletion of the initial state by the laser-induced transitions. To describe this effect one could employ for \(\Psi_i\) and \(\Psi_f\) the quasi-energy states with complex-valued quasienergies; however we do not resort to this complication below (another method to account for the depletion effects was suggested by Lewenstein et al. \([12]\)).

Thus the problem under consideration is reduced to calculation of the finite Fourier transform \(d_N\) (2.14). One readily notices that

\[
d^+_N = d^-_{-N} ,
\]
and therefore
\[ d_N = d_N^+ + d_N^- , \] (2.17)
whereas (2.12) leads to the distinct result
\[ d_N = d_N^+ + (d_N^-)^* . \] (2.18)
As discussed above, for high harmonic generation we anticipate that \( |d_N^-| \ll |d_N^+| \). If the term \( d_N^- \) is negligible, then the formulae (2.17) and (2.18) agree. The quantitative assessment for the quality of this approximation could be found in Sec.V.

B. Keldysh-type approximation

Our basic approximation is to neglect the effect of the atomic core potential \( V_a \) in the time-propagator \( G(t, t') \). A similar assumption underlies the Keldysh [20] model, whose recent adiabatic modification [35–37] gives very reliable quantitative results for photodetachment. A useful extension of the Keldysh model accounts for the Coulomb electron-core interaction [39,40]. The Green function within this approximation is straightforwardly represented via the standard Volkov wave functions \( \Phi_p(r, t) [\theta(x) = 1, (x > 0); \theta(x) = 0, (x < 0)]:\n\]
\[ G(r, t; r', t') = -i\theta(t - t') \int \frac{d^3q}{(2\pi)^3} \Phi_q(r, t) \Phi_q^*(r', t') . \] (2.19)

Explicit expression for the Volkov functions is conveniently cast as
\[ \Phi_p(r, t) = \chi_p(r, t) \exp \left( -i\hat{E}_p t \right) , \] (2.20)
\[ \chi_p(r, t) = \exp \left\{ i \left[ (p + k_t)r - \int_0^t (E_p(\tau) - \hat{E}_p) d\tau + \frac{pF}{\omega^2} \right] \right\} , \] (2.21)
where the factor \( \chi_p(r, t) \) is time-periodic with the period \( T = 2\pi/\omega \),
\[ k_t = \frac{F}{\omega} \sin \omega t , \] (2.22)
\[ E_p(t) = \frac{1}{2} (p + k_t)^2 , \] (2.23)
\[ \hat{E}_p = \frac{1}{T} \int_0^T E_p(\tau) d\tau = \frac{1}{2}p^2 + \frac{F^2}{4\omega^2} . \] (2.24)

Due to the property (2.16) it is sufficient to restrict subsequent analysis to the \( d_N^+ \) component. Using (2.9a), (2.14) and (2.19) one can rewrite Eq.(2.14) as
\[ d_N = -\frac{i}{T} \int_0^T dt \int_{-\infty}^t dt' \int \frac{d^3q}{(2\pi)^3} \langle \Phi_a(t) | e^{i\Omega t} \hat{\epsilon} | \Phi_q(t')V_F(t')|\Phi_a(t') \rangle = \]
\[ = -\frac{i}{T} \int_0^T dt \int_{-\infty}^t dt' \int \frac{d^3q}{(2\pi)^3} \langle \varphi_a | \hat{\epsilon} | \chi_a(t) \rangle \langle \chi_q(t')V_F(t')|\varphi_a \rangle \times \]
\[ \times \exp \left[ i \left( E_a - \hat{E}_q + \Omega \right) t + i \left( \hat{E}_q - E_a \right) t' \right] \] (2.25)
with \( \Omega = N\omega \). In the latter representation the phase factors with the phases linear in \( t \) and \( t' \) are explicitly singled out; the remaining factor in the integrand is \( T \)-periodic both in \( t \) and \( t' \).

### III. GREEN FUNCTION REPRESENTATION

#### A. Factorization technique

In the present section we use the theoretical technique elaborated by Kuchiev [29,30]. It allows us to transform the right hand side of (2.25) to the form convenient for the analysis. This transformation amounts to some special representation of the time-dependent Green function, since (2.25) can be considered as its generalized matrix element.

Generally speaking the correct description of the high Fourier components \( d_N \) represents a formidable theoretical task. Its numerical implementation via solving the non-stationary Schrödinger equation requires both a supercomputer and exceptional effort. In representation (2.25) the difficulty lies in the strong variation of the integrand as a function of the time variables \( t, t' \). The application of the asymptotic technique is hindered by the fact that in expression (2.25) the integration variables are not independent: namely, the limit of \( t' \)-integration depends on \( t \). The crucial simplification gained by using the factorization technique [29] allows us to disentangle the integration variables at a price of introducing an extra summation. Very importantly, this summation is physically meaningful as it corresponds to the contributions of different ATI channels. The integration over the intermediate momenta \( q \) [coming from (2.19)] is carried out in closed form. It should be emphasized that we do not use the so called pole approximation [43] applied recently by Faisal and Becker in their model of non-sequential double ionization by laser field [41,42].

In order to implement this program we transform the integral over \( t' \) using the identity

\[
\int_{-\infty}^{t} dt' \exp(iEt') \ f(t') = -i \sum_{m=-\infty}^{\infty} \frac{1}{T} \int_{0}^{T} dt' \ f(t') \exp \{i [(E - m\omega)t + m\omega t']\} \frac{1}{E - m\omega - i0},
\]

that is valid for any periodic function \( f(t) = f(t + T) \). The identity can be easily derived with the help of the Fourier expansion \( f(t) = \sum_{m} f_{m} \exp(-im\omega t) \). Employing it we rewrite Eq.(2.25) in the form of the series

\[
d_{N}^{\pm} = \sum_{m} d_{N,m}^{\pm},
\]

where

\[
d_{N,m}^{\pm} = -\frac{1}{T^2} \int_{0}^{T} dt \int_{0}^{T} dt' \int \frac{d^3q}{(2\pi)^3} \ \frac{\langle \varphi_a | \hat{d}_{\epsilon} | \chi_{q}(t) \rangle \langle \chi_{q}(t') | V_{F}(t') | \varphi_a \rangle}{E_{q} - E_{a} - m\omega - i0} \times \exp[i(\Omega - m\omega)t + im\omega t'].
\]

The next step is to carry out the integration over \( q \). To this end we note that the wave function \( \chi_{q}(r, t) \) [see Eq.(2.21)], depends on \( q \) in a very simple way. Namely, it is an exponent
of a linear form of \( q \):
\[
\chi_q(r, t) = \exp \{ i q [r + (F/\omega^2) \cos \omega t] + \alpha \},
\]
where \( \alpha \) is \( q \)-independent phase. The other source of \( q \) dependence in the integrand is its denominator that contains term \( \frac{1}{2} q^2 \). The calculation of three-dimensional integrals of this type is standard. The integration over \( |q| \) might be carried out as a contour integration in the complex plane in order to specify the following validity conditions:
\[
\begin{align*}
\text{Re} R &> 0 , \\
\text{Im} K_m &> 0 ,
\end{align*}
\]
where
\[
K_m = \sqrt{2 \left( m\omega - \frac{F^2}{4\omega^2} + E_a \right)}
\]
is the photoelectron momentum after absorption of \( m \) laser quanta (see also Sec. III B), \( R = \sqrt{R^2} \) is the function of all the variables of integration
\[
R = R(r, r', t, t') = \int_t^{t'} k \tau \, d\tau - r' + r = \frac{F}{\omega^2} \left( \cos \omega t - \cos \omega t' \right) + r - r'.
\]
Note that below we shift the integration to the complex-valued time \( t' \), and hence complex-valued \( R \). After integration over \( q \) the expression for \( d_{Nm}^+ \) reads
\[
d_{Nm}^+ = -\frac{1}{T^2} \int_0^T dt \int_0^T dt' \int d^3 r \int d^3 r' \varphi_a(r) (\epsilon \cdot r)(r' \cdot F \cos \omega t') \varphi_a(r') \times \\
\times \frac{1}{2\pi R} \exp \left\{ i \left[ K_m R + (\Omega - m\omega)t + m\omega t' + k_t r - k_t r' + \int_t^{t'} \frac{k^2}{2} \tau - \frac{F^2}{4\omega^2} \right] \right\} .
\]
Eq. (3.8) is convenient for evaluating the parameters governing the process. First of all note that integration over the variables \( r, r' \) is localized in the vicinity of the atom. The characteristic atomic dimensions should be compared with the amplitude \( F/\omega^2 \) of the electron wiggling in the laser field (the latter motion being described by \( \int k \tau \, d\tau \)). This amplitude becomes large even for quite moderate electric fields
\[
\frac{F}{\omega^2} \gg 1 .
\]
Note that this inequality may be satisfied both for large \( (\gamma > 1) \) as well as small \( (\gamma < 1) \) values of the Keldysh adiabaticity parameter \( \gamma = \omega \sqrt{2|E_a|}/F \). Therefore, for the fields satisfying (3.9) we can assume that
\[
\frac{F}{\omega^2} \gg r, r'.
\]
Actually the applicability of the resulting approximation is even broader than outlined above since in fact relation (3.9) has to be replaced by its more accurate version
\[
\frac{(-eF)}{\omega^2} | \cos \omega t - \cos \omega t' | \gg 1 .
\]
The main contribution corresponds to the complex values of time when $|\cos \omega t'| \gg 1$, as elaborated in the adiabatic approximation of Sec. [V].

These observations allow us to simplify $R$ by neglecting $r$ and $r'$ in the pre-exponential factor of the integrand in (3.8) and retaining the first order terms in the phase:

$$R \approx R_0 + \frac{R_0 \cdot (r - r')}{R_0},$$  

(3.12)

$$R_0 = R_0(t, t') = \sqrt{R_0^2},$$  

(3.13)

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

(3.14)

As a result the Green function in (3.8) becomes simpler in the 'wave zone':

$$\frac{1}{R} \exp (iK_m R) \approx \frac{1}{R_0} \exp \{i [K_m R_0 + K_m \cdot (r - r')]\},$$  

(3.15)

$$K_m = \sigma K_m \frac{F}{F}, \quad \sigma = \pm 1.$$  

(3.16)

Substituting Eq.(3.15) in (3.8) and using Eq.(3.6) to rewrite the exponent we find

$$d_{Nm}^+ = - \sum \sigma \frac{1}{2\pi T^2} \int_0^T dt \int_0^T dt' \int d^3r \int d^3r' \varphi_a(r) (er) \cdot (r'F \cos \omega t') \varphi_a(r') \times$$  

$$\times \frac{1}{R_0(t, t')} \exp \{i [K_m R_0(t, t') + (\Omega - m\omega)t + m\omega t' + (K_m + k_r) r -$$  

$$- (K_m + k_{r'}) r' + \int_{t'}^t d\tau \left(\frac{1}{2}k_r^2 - \frac{F^2}{4\omega^2}\right)\}.$$  

(3.17)

Here summation over $\sigma = \pm 1$ indicates that one has to take $K_m$ parallel or antiparallel to $F$ depending on the sign of $(\cos \omega t - \cos \omega t')$ in order to satisfy the convergence condition (3.4) (we postpone till the next section the more detailed discussion of this issue). Note that the denominator $R_0$ in (3.17) can turn zero thus challenging convergence of integrals. This problem is circumvented if one presumes that the integration over $t'$ is shifted from the real axis into the upper half-plane of complex-valued time. The integration contour $C$ will be specified more exactly below.

Now we return to the time-dependent bound state wave function $\Phi_a(r, t)$ and Volkov states $\Phi_p(r, t)$ bearing in mind relation (3.6). This gives us an appealing representation:

$$d_{Nm}^+ = - \sum \sigma \frac{1}{2\pi T^2} \int_0^T dt \int_0^T dt' \frac{1}{R_0(t, t')} \times$$  

$$\times \langle \Phi_a(t) \mid \exp(i\Omega t) \hat{d} \mid \Phi_{K_m}(t) \rangle \langle \Phi_{K_m}(t') \mid V_F(t') \mid \Phi_a(t') \rangle.$$  

(3.18)

B. Above Threshold Ionization and Laser Assisted Recombination

An interpretation of formula (3.18) is based on the fact that the integrand is a product of physically meaningful factors. In order to recognize the first of them one should recall
that the amplitude of $m$-photon detachment of electron from the initial state $\Phi_a$ within the Keldysh \cite{26} approximation is given by

$$A_m(p) = \frac{1}{T} \int_0^T dt \, \langle \Phi_p(t) | V_F(t) | \Phi_a(t) \rangle .$$ \hspace{1cm} (3.19)

Generally, the number $m$ of photons absorbed is larger than a minimum necessary for the electron detachment, therefore the process corresponds to the Above Threshold Ionization (ATI). In the right hand side of (3.19) the index $m$ is implicit. It enters via the absolute value of the final electron momentum $p$ which is subject to the energy conservation constraint

$$\frac{1}{2} p^2 = m\omega - \frac{F^2}{4\omega^2} + E_a .$$ \hspace{1cm} (3.20)

Note that $-E_a \equiv \frac{1}{2}\kappa^2 > 0$ is the electron binding energy in the initial state; $F^2/(4\omega^2)$ is the electron quiver energy in the laser field. Comparing (3.20) with (3.6) we confirm that $K_m$ is exactly the physical translational electron momentum in the $m$-th ATI channel.

The second relevant process is that of laser assisted recombination (LAR) when the continuum electron with the momentum $p$ collides with an atom in the laser field. The electron absorbs some extra laser quanta and goes to the bound state $\Phi_a$ emitting single photon of frequency $\Omega$. In the Keldysh-type approximation this LAR process has the amplitude

$$C_{N,m}(p) = \frac{1}{2\pi T} \int_0^T dt \, \langle \Phi_a(t) | \exp(i\Omega t) \hat{d}\epsilon | \Phi_p(t) \rangle .$$ \hspace{1cm} (3.21)

It could be called also laser-induced recombination. The allowed values of the high-energy photon frequency $\Omega$ are

$$\Omega = \frac{1}{2} p^2 + \frac{F^2}{4\omega^2} + m\omega$$ \hspace{1cm} (3.22)

with integer $m$.

One readily notices that the integrand in (3.18) bears a striking resemblance to the product of the integrands in (3.19) and (3.21). This allows us to interpret the amplitude (3.18) as describing the two-step transition: at first the electron goes to the laser-field dressed continuum (Volkov) state $\Phi_{K_m}$ after absorption of $m$ photons; at the second step it returns to the initial state emitting single photon with the frequency $\Omega = N\omega$. Note that the intermediate momentum $p = K_m$ is restricted not only in magnitude (3.6) but also in direction: according to Eq. (3.16) the vector $K_m$ is parallel or anti-parallel to the electric vector $F$ in the laser wave. The physical implications of this circumstance are discussed in the next section.

There is an extra factor $1/R(t,t')$ in the integrand of (3.18) which prevents complete separation of integrations in $t$ and $t'$. In fact its presence is physically well understandable. Indeed, the definition of $R(t,t')$ (3.7) shows that classically it is the distance between the electron positions at the moments $t$ and $t'$ with account for the electron wiggling in the laser field. $1/R$ could be named an expansion factor since in the absence of the laser field ($F \to 0$) it describes conventional decrease of the amplitude in a spherical wave as it expands in 3D.
space ($\sim 1/|\mathbf{r} - \mathbf{r}'|)$). When the laser field is operative, the form of the expansion factor is drastically modified according to the approximation (3.14). Hence the interpretation of the expression (3.18) is that the electron first is transferred to the $m$-th ATI channel, then propagates in space under the influence of the laser wave and finally recombines to the initial state emitting the photon with the frequency $\Omega$. The contribution of each path is labeled by the number of virtually absorbed photons $m$. These contributions add up coherently as shown by Eq. (3.2). For the actual ATI process the momentum $K_m$ should be real. The summation in (3.2) includes also the virtual processes with the imaginary values of $K_m$, but their contribution is anticipated to be small.

It is worthwhile now to outline equivalent but more convenient representation of ATI and LAR amplitudes. Bearing in mind the character of $\mathbf{r}$-dependence of Volkov states (2.20)-(2.21) it is easy to see that the space integration in the formulae (3.19) or (3.21) essentially reduces to the Fourier transformation. For instance, as shown in much detail by Gribakin and Kuchiev [35], the ATI amplitude (3.19) is exactly presented as

$$A_m(p) = -\frac{1}{2T} \int_0^T dt \left[ (p + k_t)^2 + \kappa^2 \right] \tilde{\phi}_a(p + k_t) \exp[iS(t)], \quad (3.23)$$

where $S(t)$ is the classical action

$$S(t) = \frac{1}{2} \int_0^t d\tau (p + k_\tau)^2 - E_a t, \quad (3.24)$$

$\tilde{\phi}_a(q)$ is the Fourier transform of $\phi_a(r)$:

$$\tilde{\phi}_a(q) = \int d^3r \exp(-iqr) \phi_a(r). \quad (3.25)$$

Similarly, for the LAR amplitude one obtains

$$C_{Nm} = -\frac{1}{2\pi T} \int_0^T dt \exp \{ i[\Omega t - S(t)] \} \tilde{\phi}^{(e)}(-K_m - k_t) \quad (3.26)$$

with

$$\tilde{\phi}^{(e)}(q) = i (\mathbf{e} \cdot \nabla_q) \tilde{\phi}_a(q). \quad (3.27)$$

Before concluding this section it is worthwhile to make an important observation. It is well known that the Keldysh approximation in the theory of multiphoton ionization is not gauge invariant. The detailed discussion of this issue can be found in Ref. [44]. Basing on physical grounds we use the dipole-length form (2.10) for the interaction of the electron with the laser field. As thoroughly discussed earlier [35,36], this gauge stresses large separations of the active electron from the core where one-electron approximation is better justified and one can employ the asymptotic form for the initial-state wave function:

$$\phi_a(r) \approx A_a r^{-\nu - 1} \exp(-\kappa r) Y_l(\hat{r}) \quad (r \gg 1/\kappa), \quad (3.28)$$

where $\kappa = \sqrt{2|E_a|}$, $\nu = Z/\kappa$, $Z$ is the charge of the atomic residual core ($\nu = Z = 0$ for a negative ion), $l$ is the active electron orbital momentum in the initial state and $\hat{r} = \mathbf{r}/r$ is
the unit vector. The coefficients $A_a$ are tabulated for many negative ions [45]. The Fourier transform $\tilde{\Phi}_a(q)$ (3.25) is singular at $q^2 = \kappa^2$ with the asymptotic behavior for $q \to \pm i\kappa$ defined by the long-range asymptote (3.28) in the coordinate space

$$
\tilde{\phi}_a(q) = 4\pi A_a(\pm 1)^l Y_m(q) \frac{(2\kappa)^\nu \Gamma(\nu + 1)}{(q^2 + \kappa^2)^{\nu+1}}, \quad (3.29)
$$

with $(\pm 1)^l$ corresponding to $q \to \pm i\kappa$.

At the present stage of our development the interpretation of (3.18) outlined above should be considered only as qualitative, since the two time integrations in Eq. (3.18) are not completely separated due to the factor $1/R_0(t, t')$. The complete factorization is possible under additional approximation which actually is not restrictive possessing a broad applicability range.

IV. ADIABATIC ANALYSIS

A. Adiabatic approach to Above Threshold Ionization

We start with reiterating the basics of the adiabatic approximation in multiphoton detachment theory [35,36] that allows one to carry out analytically integration in expression (3.19) or (3.23) for the amplitude. It is presumed that the laser frequency $\omega$ is small, i.e. that the number of absorbed photons $m$ is large (the practical applicability of this approach proves to be very broad, since it gives reasonable results even for $m = 2$, see, for instance, Ref. [37]). Then the integrand in (3.19) or (3.23) contains large phase factor $\exp\left[iS(t)\right]$ and the integral may be evaluated using the saddle point method [35–38]. The positions of the saddle points in the complex $t$ plane is defined by equation

$$
S'(t_{mu}) = 0 , \quad (4.1)
$$

or, more explicitly,

$$
\left( p + k_{t_{mu}} \right)^2 + \kappa^2 \equiv \left( p + F \frac{\omega}{\sin \omega t_{mu}} \right)^2 + \kappa^2 = 0 . \quad (4.2)
$$

Eqs. (4.1) or (4.2) are to be considered together with the energy conservation constraint Eq. (3.20). Note that according to formula (3.29) the position of the saddle point coincides with the singularity of the bound-state wave function in the momentum space thus stressing the importance to describe correctly the long-range behavior of coordinate-space wave function. The latter could be ensured much more easily than the proper description of the wave function inside the core region. Therefore the use of the adiabatic approximation and characterization of the bound wave function solely by its asymptotic behavior (3.28) constitutes a self-consistent approach.

The result of calculations of the amplitude (3.13) in the stationary phase approximation could be written as a modification of formula (25) in Ref. [35].
\[ A^{(sp)}_{m \mu} (p) = \sum_{\mu} A^{(sp)}_{m \mu} (p) , \quad (4.3) \]

\[ A^{(sp)}_{m \mu} (p) = -\frac{(2\pi)^2}{T} A_a \Gamma(1 + \nu/2) 2^{\nu/2} \kappa^{\nu} Y_{lm}(\hat{p}_{m \mu}) \frac{\exp \left[ iS(t_{m \mu}) \right]}{\sqrt{2\pi [-iS''(t_{m \mu})^{\nu+1}]}}. \quad (4.4) \]

In the plane of the complex-valued time the saddle points \( t_{m \mu} \) lie symmetrically with respect to the real axis. There are four saddle points in the interval \( 0 \leq \text{Re} t_{m \mu} \leq T \), two of them lying in the upper half plane (\( \text{Im} \ t_{m \mu} > 0 \)). The integration contour in the plane of complex time is shifted upwards. Therefore only two saddle points with \( \text{Im} \ t_{m \mu} > 0 \) are operative being included into the summation in (4.3) (\( \mu = \pm 1 \)); \( \hat{p}_{m \mu} \) is a unit vector in the direction of \( p + k_{t_{m \mu}} \).

**B. Adiabatic approach to Harmonic Generation**

Although in the harmonic generation matrix element (3.18) the time integration variables \( t \) and \( t' \) are not fully separated, as indicated above, the large phase factor \( \exp \left[ iS(t') \right] \) in \( t' \) integration is the same as in the multiphoton detachment case (3.19). Therefore we can apply the saddle point approximation to carry out integration over \( t' \). The integration contour is again shifted upwards in the complex \( t' \) plane; this is the contour \( C \) in the formulae (3.17) and (3.18). The saddle points are defined from the same Eq.(4.2). By solving it we find for the \( m \)-dependent saddle points \( t'_{m \mu} \):

\[
\sin \omega t'_{m \mu} = \frac{\omega}{F} \left( -K_m + i\mu \kappa \right) .
\]

\[
\cos \omega t'_{m \mu} = \sqrt{1 - \left( \frac{\omega}{F} \right)^2 \left( -K_m + i\mu \kappa \right)^2} \quad (\text{Im} \cos \omega t'_{m \mu} > 0). \quad (4.5)
\]

For each value of \( m \) there are two essential saddle points, i.e. these with \( \text{Im} t'_{m \mu} > 0 \), labeled by subscript \( \mu = \pm 1 \).

Thus in the stationary phase approximation for the \( t' \) integration we obtain from (3.18)

\[ d^+_N m(\Omega) = \sum_{\sigma} \sum_{\mu} A^{(sp)}_{m \mu}(K_m) B^+_N m \mu , \quad (4.6) \]

\[ B^+_N m \mu = \frac{1}{2\pi T} \int_0^T dt \frac{1}{R_0(t, t'_{m \mu})} \langle \Phi_{a}(t) | \exp(i\Omega t) \hat{d}_\epsilon | \Phi_{K_m}(t) \rangle . \quad (4.7) \]

Similarly to Eq.(3.26) one can conveniently employ the Fourier transform which gives

\[ B^+_N m \mu = \frac{1}{2\pi T} \int_0^T dt \frac{\exp \left\{ i [\Omega t - S(t)] \right\}}{(F/\omega^2) \left( \cos \omega t - \cos \omega t'_{m \mu} \right)} \tilde{\phi}_{a}(\epsilon)(-K_m - k_t) . \quad (4.8) \]

In particular, for a negative ion (\( \nu = 0 \)) with the active electron in an \( s \) state (\( l = 0 \)) we have from (3.29) (\( \hat{q} \equiv q/q \))

\[
\tilde{\phi}_{a}(q) = \sqrt{4\pi} A_a \frac{1}{(q^2 + \kappa^2)} , \quad (4.9)
\]

\[
\tilde{\phi}_{a}(\epsilon)(q) = -i (\epsilon \cdot \hat{q}) \sqrt{4\pi} A_a \frac{2q}{(q^2 + \kappa^2)^2} , \quad (4.10)
\]
and (4.8) simplifies to

$$B_{N \mu} = i \frac{2A_n}{\sqrt{\pi T}} \frac{\omega^2}{F} \int_0^T dt \exp \left\{ i [\Omega t - S(t)] \right\} \frac{\epsilon \cdot (K_m + k_i)}{\left[ (K_m + k_t)^2 + \kappa^2 \right]^2}. \quad (4.11)$$

As noted above Eq. (3.18), the summation over $\sigma = \pm 1$ indicates that in order to satisfy the convergence condition (3.4) one has to choose a sign in $R_0(t, t') = \pm (F/\omega^2) (\cos \omega t - \cos \omega t')$ that makes $R_0(t, t') > 0$. By considering the phase factor in (3.17), we see that the upper sign corresponds to $K_m$ parallel to $F$ whereas the lower sign leads to $K_m$ antiparallel to $F$ (without changing $|K_m|$). This looks natural since there is no reason to prefer one of these directions.

The right hand side of formula (4.6) contains also summation over the saddle points $t_{m \mu}'$ (for the monochromatic laser field two such points are operative being labeled by the index $\mu = \pm 1$). However, as argued by Kuchiev [29], actually only one saddle point (i.e. one value of the label $\mu$) contributes for each choice of $\sigma = \pm 1$, i.e. for $K_m$ parallel to $F$ and for $K_m$ antiparallel to $F$ (as discussed below, this has a simple and clear physical interpretation). Consequently the double summation over $\sigma$ and $\mu$ is effectively replaced by a single summation. Moreover, in the latter sum both terms are equal. To see this one should remember that the preexponential factor $1/R_0(t, t')$ changes sign depending on the value of $\sigma = \pm 1$. Hence the remaining single summation is equivalently replaced by the factor of 2. This allows us to finally rewrite (3.2) using (4.6) as

$$d_N^+ = 2 \sum_m A_{(sp)}^{(m \mu_0)}(K_m) B_{N m \mu_0}, \quad (4.12)$$

where $A_{(sp)}^{(m \mu_0)}(K_m)$ and $B_{N m \mu_0}$ are given respectively by (4.4) and (4.11) for negative ion ($\nu = 0$). Expression (4.12) is to be calculated for the subscript $\mu_0$ corresponding to one of the saddle points $t_{m \mu_0}'$, for instance, that with the smaller value of $\text{Re} t_{m \mu_0}'$ ($\text{Im} t_{m \mu_0}' > 0$).

In formula (4.12) the factor $B_{N m \mu_0}$ describes jointly the 3D-wave expansion and LAR. These two effects could be further factorized using the approximation $|\cos \omega t_{m \mu_0}'| \gg |\cos \omega t|$ [29]:

$$B_{N m \mu_0} = \frac{1}{R_{m \mu_0}} C_{N m}(K_m), \quad (4.13)$$

$$d_N^+ = 2 \sum_m A_{(sp)}^{(m \mu_0)}(K_m) \frac{1}{R_{m \mu_0}} C_{N m}(K_m), \quad (4.14)$$

where $1/R_{m \mu_0} = \omega^2/(F \cos t_{m \mu_0}')$ is the laser-modified expansion factor in its simplest form and $C_{N m}$ (3.26) is LAR amplitude.

The adiabatic approximation could be further applied to carry out integration over $t$ in $B_{N m \mu}$ (4.11) or in $C_{N m}$ (3.26) by the saddle point method. However below we do not pursue this objective and evaluate these integrals numerically.

Eq. (4.12) and its simplified version (4.14) present the main result of this paper. These formulae implement the very simple picture of the HG process as consisting of three successive steps. First, the electron absorbs $m$ laser photons. The amplitude of this event is $A_{m \mu}$. In order to contribute to HG the photoelectron has to return to the parent atomic
core where LAR is solely possible. The amplitude of return is described by the expansion factor $1/R$. It appears explicitly in (4.14), while in (4.12) it is incorporated in the definition of the amplitude $B_{N m \mu_0}$. The propagation of the electron describes the second phase of the event. At the third step the electron collides with the core absorbing $N - m$ photons from the laser field and emitting the single high-frequency quantum $\Omega = N \omega$ as it recombines to the bound state. This LAR process has the amplitude $C_{N m} (3.21)$. The summation over $m$ in the total amplitude $d_N$ (4.12) takes into account interference of the transitions via different intermediate ATI channels.

This appealing physical picture is supplemented by the very simple way to evaluate numerically all the quantities in (4.12). The amplitude of photoionization $A_{m \mu_0}^{(sp)}$ is calculated via plain analytical formulae with the validity well testified before. The LAR process did not attract much attention in the literature and certainly deserves more study that we hope to present elsewhere. Here we emphasize only that since $C_{N m \mu_0}$ as well as closely related amplitude $B_{N m \mu_0}$ are very similar in structure to the amplitude $A_{m \mu_0}^{(sp)}$, one can hope that similar methods of evaluation also produce reliable results.

The nontrivial point in the presented picture is the probability for the ATI electron to return to the core. Intuitively, one could anticipate that such a process is suppressed, because the most natural behavior for the electron would be simply to leave the atom. The proper description of the suppression plays substantial role in the theory. According to the physical image of the ATI process worked out in the adiabatic approach [35], after tunneling through the time-dependent barrier the ATI electron emerges from under the barrier at some point which is well separated from the core. As a result this point becomes the source of an expanding spherical wave. This occurs twice per each cycle of the laser field, at the two moments of time $t'_{m \mu}$ when the source-points lie up and down the field $F$ from the core. The interference of the two spherical waves originating from the two different source-points results in non-trivial patterns in the angular ATI photoelectron distributions obtained from (4.3)-(4.4) [35] [36] in agreement with the available theoretical and experimental data. The probability for the ATI electron to return to the core from the source-point is governed by the expansion factor $1/R$ and by the direction of propagation. At each of the moments $t'_{m \mu}$ only one of the two possible directions of $K_{m}$, labeled in (3.18) by $\sigma = \pm 1$, results in the electron eventually approaching the core. For the opposite direction of $K_{m}$ the electron recedes from the core and does not come back to recombine. In other words, for each direction of $K_{m}$ only one of the two saddle points $t'_{m \mu}$ contributes to HG. Since both values of $\sigma$ give identical contributions, summation over $\sigma$ simply gives an extra factor of 2 in (4.12).

C. Choice of the gauge

Calculation of $d_N$ according to the formulae (2.2), (2.14), with exact wave function $\Psi$ might be equivalently carried out in various forms which correspond to different choice of gauge. As soon as the approximations are employed, the theory loses gauge invariance. Although the length gauge is known to be superior for the description of ATI within the adiabatic approximation [35], the situation is not that straightforward for the high-energy photon. In the derivation above we employed the dipole-length form for the operator $d_e$ (2.2). In the velocity-length form this operator is to be substituted according to the rule ($\mathbf{p} \equiv -i \nabla_{\mathbf{r}}$ is the electron momentum)
\[ d_\epsilon = \epsilon \cdot r \Rightarrow \frac{i}{\Omega} \epsilon \cdot p \]  

(4.15)

when calculation of \( d_N \) is concerned \((\Omega = N\omega)\). It is easy to see that this substitution is equivalent to replacement of \( B_{m\mu} \) \((4.11)\) by

\[
B_{m\mu}^{(v)} = -\frac{1}{\Omega} \frac{A_\alpha}{\sqrt{\pi} T} \int_0^T \frac{dt}{F} \exp \{i [\Omega t - S(t)]\} \frac{\epsilon \cdot (K_m + k_i)}{(K_m + k_i)^2 + \kappa^2}. \]

(4.16)

As compared with the expression \((4.11)\), the latter one differs by the extra factor

\[
\frac{(K_m + k_i)^2 + \kappa^2}{2\Omega} \]

(4.17)

in the integrand.

V. RESULTS OF CALCULATIONS

Within framework of the present theory we calculate the rates of generating the \( N \)-th harmonic radiation \((c \) is the velocity of light)

\[
R_N \equiv \frac{\omega^3 N^3}{2\pi c^3} |d_N|^2
\]

(5.1)

introduced by Becker et al \([15]\) (and denoted by these authors as \( dR_N/d\Omega_K \)) who in particular carried out calculations for the HG by \( \text{H}^- \) ion in the \( \omega = 0.0043 \) laser field. Fig. \([4]\) provides a complete comparison of these results with the results of our calculations. We employ the binding energy of \( \text{H}^- \) (\( \kappa = 0.2354 \)), but replace the true value \( A_\alpha = 0.75 \) \([40]\) by \( \sqrt{2\kappa} = 0.686 \) since this corresponds to the zero-range potential model used by Becker et al \([15]\). Our rates were multiplied by the extra factor \( N_e^2 \), where \( N_e = 2 \) accounts for the presence of two active electrons in \( \text{H}^- \). For the real \( \text{H}^- \) ion the results shown in Fig. \([4]\) are to be scaled by a factor \( A^4/(2\kappa)^2 \).

Fig. \([4]\) shows harmonic spectra for the laser field intensities \( I = 10^{10}, 2 \cdot 10^{10}, 5 \cdot 10^{10}, 10^{11} \) \( \text{W/cm}^2 \) that correspond respectively to the Keldysh adiabatic parameter values \( \gamma \equiv \omega\kappa/F = 1.898, 1.342, 0.849, 0.600 \). Thus the most interesting region of transition from the multiphoton regime \((\gamma \gg 1)\) to the tunneling mechanism \((\gamma \ll 1)\) is covered. Our major results are shown by open circles in Fig. \([4]\). They are obtained using the expression \((4.12)\) for \( d_N \) with the time-integration in \( B_{N m\mu_0} \) \((4.11)\) carried out numerically. Based on physical arguments, we extend the summation in \((4.12)\) only over open ATI channels with the real values of \( K_m \). Generally the HG spectrum is known to consist of the initial rapid decrease, the plateau domain and the rapid fall-off region. The present theory is designed to describe the high HG but not the initial decrease, which in the case considered is noticeable only for one or two lowest harmonics. In the fall-off region, i.e. on the large-\( N \) side, our rates perfectly coincide with those obtained by Becker et al \([15]\) (closed circles in Fig. \([4]\)). The slight difference that could be hardly distinguished in the plot scale lies within uncertainty in retrieving data from the small-size plot published in Ref. \([13]\). Of course, our comparison is carried out in absolute scale without any fitting or normalization.
The deviations increase as $N$ decreases, but within entire plateau region the agreement of rates averaged over structures remains good. Remarkably, the positions of numerous dips and peaks that exist in the plateau region are well reproduced by our calculations, although there exist some, generally not strong, discrepancies in their magnitudes. The structures has not yet received a universally accepted physical interpretation in the current literature with two tentative explanations being available. Becker et al. [14] relate these structures to ATI channel closing whereas Lewenstein et al. [19] suggest that it stems from quantum interferences between the contribution of different electron trajectories. Within the present framework we can say that the origin of these structures lies in the interplay of interfering contributions of various ATI paths, but their precise description would require an additional detailed analysis.

Approximation (4.14) with numerical calculation of the integral in $C_{N,m}$ (3.26) (open squares in Fig. 1) somewhat overestimates HG rate, but still retains the structure, though smoothed. It is worthwhile to emphasize two circumstances. First, to the best of our knowledge, we present here almost unique quantitative comparison of HG rates calculated within different theoretical methods, namely, by the present approach and by that developed by Becker et al. We are aware of only two cases when the quantitative comparison was carried out previously, one refers to some parameters of HG by elliptically polarized light in Ref. [20] (see also discussion in the Introduction), the other is comparison between the approach of Ref. [12] and results of direct numerical integration of the three-dimensional time-dependent Schrödinger approximation [51]. Second, the comparison is presented in the log-scale, as are the results reported by Becker et al, because it is appropriate both for the physics of the problem and for the current state of experiment.

In the summation (4.12) over ATI channels (i.e. over $m$) the coherence is very important, since large number of terms is comparable in modulus, but have rapidly varying phases. Many ATI channels contribute to HG for each $N$. This is in variance with the tentative conclusion by Eberly et al. [6]). The low ATI channels give appreciable contribution even for quite high harmonics. Only for the highest harmonics considered the contribution of low ATI channels becomes negligible.

Although the length gauge is known to be superior for the description of ATI within the adiabatic approximation [35], the situation is not that straightforward for the high-energy photon. Therefore our calculations for the rates were carried out using both the length and velocity gauges. For large $N$ the results obtained are very close, see Fig. 2: the dipole-velocity gauge producing the rates about 10% larger. For smaller $N$ the difference increases and manifests rather irregular $N$-dependence. However, even in the most unfavorable situation the ratio of the length-form to velocity-form results deviates from 1 not more than by 50%, which is rather reasonable, bearing in mind the multiphoton nature of the process.

An evolution of the parameters of the individual $N$-th harmonic radiation with variation of the laser field intensity $I$ is presented in Fig. 3 for some particular values of $N$. We show both $|d_N^+|^2$ (that is proportional to HG rate) and the phase $\Phi_N \equiv \arg d_N^+$. The intensity dependence of the phase $\Phi_N$ is known [17–50, 41] to play an important role in description of the harmonic field propagation in the experimental conditions when the spectral and spatial coherence properties are substantial. Our calculations demonstrate rapid variation of the phase $\Phi_N$ with the intensity $I$: as $I$ increases by an order of magnitude the phase changes by about $10\pi$ (for $N = 9$) or $15\pi$ (for $N = 11$). The dependence of phase on $I$ looks most simple
in the fall-off domain where it is essentially linear, Fig. 3a. As it is well known [14,15], the HG rate manifests spikes (see Fig. 3a, where the spikes are slightly smoothed due to finite step over $I$ used in plotting) at the intensities $I_m$ that correspond to the threshold of $m$-th ATI channels, that is closed for $I > I_m$ due to ponderomotive potential $F^2/(4\omega^2)$.

As discussed in Sec.II, in the Fourier component $d_N$ (2.17) the contribution of $d^+_N = d^-_N$ is anticipated to be negligible as compared with that of $d^+_N$. The reader have to recall that the physical reason for this is that $d^+_N$ describes the natural sequence of events, when electron at first absorbs energy from the laser field and subsequently emits the high harmonic photon, whereas for $d^-_N$ the sequence is inverted. In order to illustrate how strong the preference is we show in Fig. 4 the ratio $|d^-_N/d^+_N|^2$. The ratio rapidly decreases with $N$ being very small in all cases where the present theory applies. It becomes noticeable only for small $N$ in case of intense laser field.

VI. CONCLUSION

The major result of this paper is a quantum mechanical description of the high-harmonic generation problem as a three-step process. In the first step the atomic electron absorbs several laser quanta and populates some ATI channel. Secondly, the electron propagates in the laser field back to the atomic core. The third step is the laser-induced recombination when the high harmonic photon is emitted. This mechanism is nothing more but interpretation of our principal formulae (4.12), (4.14) derived quantum mechanically with minimal approximations and without resort to any classical or intuitive arguments. The distinctive feature of our formulae (4.12), (4.14) is that they include only genuine amplitudes of the three constituent processes, i.e. each amplitude describes true physical fully accomplished process, as HG, ATI or LAR. Here lies a crucial difference between our results and those of previous authors [12] (see also Introduction) who discussed mathematical structure of the matrix elements or integrals considered in their approach in terms of three-step mechanism but failed to present HG amplitude via amplitudes of physically observable accomplished ATI and LAR processes. Even in a more broad context this is a rare and remarkable situation when a complicated process is reduced essentially exactly to the sum over all possible paths with every path described as a sequence of real, fully accomplished physical processes. This conceptual simplicity arises due to multiphoton, adiabatic nature of HG process.

With all three constituent amplitudes available from analytical formulae or simple numerical calculations our theory promises to be an efficient practical tool. This hope is supported by good agreement of our quantitative results for HG by H$^-$ ion with the previous calculations by Becker et al [15] in a wide range of laser intensities and frequencies of the emitted quantum. As the simplest example of possible future extensions we note only that our approach can be straightforwardly applied to negative ions with the outer electron having non-zero orbital momentum, such as halogen ions. These species could be easier accessible for the experimental studies.

The structure of the formulae (4.12), (4.14) is so simple that it is tempting to suggest that they will work well if one substitutes in them ATI or LAR amplitudes obtained in some approximations other than those used in the derivation above. For instance, if ATI amplitude more accurate than that given by the Keldysh approximation is available from
some theory, one can employ it for HG calculations via (4.12), (4.14). This gives a hope to relatively simply improve account for the electron rescattering effects in the HG theory.

The distinctive feature of the present theory of HG is the use of the representation based on the discrete set of ATI channels. To the best of our knowledge this simple and apparently straightforward idea has not been exploited before. It would be underestimate to consider it merely as a detail of theoretical technique. Indeed, if one pursues the objective of the most direct and far-reaching quantum implementation of the three-step mechanism of HG, then the use of ATI channel representation becomes an unavoidable and crucial point. Otherwise, if the amplitudes of ATI do not appear in the theoretical scheme, HG cannot be properly described as a three-step process.

In this paper we do not discuss qualitative features of HG spectra, such as extension of plateau domain etc. Detailed discussion of these issues could be found in other publications [8,11,12], in particular in the paper by Becker et al [15] whose results for HG by H\(^{-}\) ion we reproduce closely within our theory. This implies that the analysis of the numerical results carried out by Becker et al [15] is fully applicable in our case. Concerning the mechanisms and physical interpretation of HG spectra, the present theory, hopefully, can add more to our understanding. However, the related analysis requires further theoretical developments which would overburden the present, already quite a long paper. We hope to present these subsequent developments elsewhere. In a broader perspective, one can expect that modifications of our approach could be applied to a variety of processes such as population of high ATI channels and multiple ionization of atoms.

As a summary, the three-step mechanism of the harmonic generation is ultimately justified. The theory is quantitatively reliable and easy to apply. It gives an important physical insight being a particular realization of the general atomic antenna mechanism.

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FIGURES

FIG. 1. Harmonic generation rates (5.1) (in sec$^{-1}$) for H$^-$ ion in the laser field with the frequency $\omega = 0.0043$ and various values of intensity $I$ as indicated in the plots. Closed circles - results obtained by Becker et al [15], open circles - present calculations in the dipole-length gauge using the expression (4.11) for $B_{Nm\mu}$, open squares - same but with the simplified formula (4.14) for $B_{Nm\mu}$.

FIG. 2. Ratio of rates for HG by H$^-$ ion in the laser field with the frequency $\omega = 0.0043$ calculated within the present approach using the velocity and length gauges. (a) – for the rates calculated using the expression (4.11) for $B_{Nm\mu}$: circles – $I = 10^{10}$; squares – $I = 2 \cdot 10^{10}$; triangles – $I = 5 \cdot 10^{10}$; diamonds – $I = 10^{11}$ W/cm$^2$; (b) - same as in (a) but with the simplified formula (4.14) for $B_{Nm\mu}$.

FIG. 3. Evolution of HG parameters with variation of the laser field intensity $I$. The harmonic intensity parameter $M_N \equiv 2 \log_{10} |d_N^+/d_N^-|$ and the reduced harmonic phase $\Phi_N/\pi$ with $\Phi_N \equiv \arg d_N^+$ are shown for harmonic of various order $N$. The bars with numbers $m$ indicate the threshold intensities $I_m$ such that for $I > I_m$ the $m$-th ATI channel is closed due to ponderomotive potential.

FIG. 4. Parameter $Q_N = 2 \log_{10} |d_N^-/d_N^+|$ as a function of harmonic number $N$ for HG by H$^-$ ion in the laser field with the frequency $\omega = 0.0043$ and various intensities: circles – $I = 10^{10}$; squares – $I = 2 \cdot 10^{10}$; triangles – $I = 5 \cdot 10^{10}$; diamonds – $I = 10^{11}$ W/cm$^2$. 
Fig. 3c

- $M_n$ vs. $I \times 10^{-10}$, W/cm$^2$

Graph with curves for $N=15$, $N=17$, and $N=19$. The y-axis represents $M_n$ and the x-axis represents $I \times 10^{-10}$, W/cm$^2$. The curves show variations with different $N$ values.
