Quantum Theory of High Harmonic Generation via Above Threshold Ionization and Stimulated Recombination

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Abstract. Fully quantum treatment explicitly presents the high harmonic generation as a three-stage process: above threshold ionization (ATI) is followed by the continuum electron propagation in a laser field and subsequent stimulated recombination back into the initial state. The contributions of all ATI channels add up coherently. All three stages of the process are described by simple, mostly analytical expressions. 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.

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 exceeds the minimum necessary for ionization, i.e. the photoelectrons are characterized by their distribution over above threshold ionization (ATI) channels. The photon production corresponds to 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., L’Huillier et al 1991, Macklin et al 1993, Schafer et al 1993) and tackled by the theory.

An idea that the two processes referred above are interrelated has been articulated quite 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 (1964) model of multiphoton ionization. The importance of this interaction for a variety of processes was first pointed out by Kuchiev

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(1987), 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 (1989) but proved to be non-realistic, see below. The hybrid classical-quantum model due to Corkum (1993) casts HG as a three-step process: tunneling ionization and subsequent propagation in the continuum is followed 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 (1993) 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 (1993) model does not directly appeal to ATI process just because the discrete ATI channels do not appear within the classical framework. The subsequent developments were based on more sophisticated theory and led to important advancements (Lewenstein et al 1994, Becker et al 1994), but apparently abandoned the 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 (Zaretskii and Nersesov, 1996).

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 amplitude of electron stimulated recombination (SR) 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. We successfully test its efficiency by quantitative comparison with the benchmark calculations by Becker et al (1994) for HG by H− ion. 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 photoelectron rescattering.

The rate of the \( N \)-th harmonic generation is proportional to \( |d_N|^2 \), where \( d_N \) is the \( N \)-th Fourier component of the transition dipole momentum \( d(t) \)

\[
d_N = \frac{1}{T} \int_0^T dt \exp(i\Omega t) \int d^3 r \, \Psi_f(r, t)^* \hat{d}_e \Psi_i(r, t) .
\]

Here \( \hat{d}_e = \epsilon \cdot r \) is an operator of the dipole moment (the atomic units are used), \( \Psi_i \) and \( \Psi_f \) are the initial and final states of the atomic system dressed by the laser field with the frequency \( \omega = 2\pi/T, \Omega = N\omega \).

Both experiment and theory concentrate almost exclusively on the case when the
initial and final states coincide. One can employ the exact time-dependent Green function $G(t, t')$ to construct the field-dressed states developed out of the initial (field-free) stationary state $\Phi_a$

$$\Phi_a(\mathbf{r}, t) = \varphi_a(\mathbf{r}) \exp(-iE_at), \quad H_a \varphi_a = E_a \varphi_a,$$

where $H_a = \frac{1}{2} \mathbf{p}^2 + V_a(\mathbf{r})$ is the atomic system Hamiltonian, $V_a(\mathbf{r})$ is the interaction of the active electron with the core. This results in the expression (cf. Becker et al 1997)

$$d_N = \frac{1}{T} \int_0^T dt \int_{-\infty}^t dt' \langle \Phi_a(t) | \exp(i\Omega t) \hat{d}_a G(t, t') V_F(t') | \Phi_a(t') \rangle,$$

where $V_F(\mathbf{r}, t) = \mathbf{F} \cdot \mathbf{r} \cos \omega t$ is the interaction between the active electron and the laser wave with the electric field amplitude strength $\mathbf{F}$ in the dipole-length gauge. Eq.(3) implies that the high harmonic $\Omega$ is emitted after the absorption of several low-frequency laser quanta, $t \geq t'$. Strictly speaking, there are ‘time-reversed’ processes in which the radiation of the high harmonic precedes the absorption of some laser quanta. However, for a large number of the quanta absorbed such a mechanism is suppressed and is therefore omitted in (3) together with the continuum-continuum transitions (the latter approximation is a rather standard one, see, e.g., Lewenstein et al (1994)).

The next basic approximation is to discard the effect of the atomic core potential $V_a$ on the Green function $G$ that allows one to represent it via the standard Volkov wave functions $\Phi_q(\mathbf{r}, t)$

$$G(\mathbf{r}, t; \mathbf{r}', t') = -i \int \frac{d^3q}{(2\pi)^3} \Phi_q(\mathbf{r}, t) \Phi^*_q(\mathbf{r}', t'), \quad t > t'.$$

Similar assumption underlies the Keldysh (1964) model, whose recent adiabatic modification (Gribakin and Kuchiev 1997a,b, Kuchiev and Ostrovsky 1998) gives fully reliable quantitative results for photodetachment. A useful extension of the Keldysh model accounts for the Coulomb electron-core interaction (Ammosov et al 1986, Krainov 1997).

Generally 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 the representation (3) the difficulty lies in the strong variation of the integrand as a function of the time variables $t$, $t'$. The crucial simplification is gained by using the factorization technique Kuchiev (1995) which 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 (4)] is carried out in closed form. Some minor additional approximations [see, for instance, Eq.(11) below; the detailed derivation is to be published elsewhere] brings us to the appealing
representation:

\[ d_N = \sum_{m=-\infty}^{\infty} \sum_{\sigma=\pm1} d_{Nm}^{(\sigma)}, \quad (5) \]

\[ d_{Nm}^{(\sigma)} = -\int_0^T dt \int_0^T dt' \langle \Phi_a(t) \mid \exp(i\Omega t) \hat{d}_e \mid \Phi_{Km}(t) \rangle \times \]

\[ \times \frac{1}{2\pi T^2 R(t, t')} \langle \Phi_{Km}(t') \mid V_F(t') \mid \Phi_a(t') \rangle. \quad (6) \]

Here the vector \( K_m \) has an absolute value

\[ K_m = \left[ 2 \left( m\omega - F^2/(4\omega^2) + E_a \right) \right]^{1/2} \quad (7) \]

and is parallel or antiparallel to \( F \)

\[ K_m = \sigma K_m F/F, \quad \sigma = \pm1. \quad (8) \]

The physical interpretation of (5), (6) is based on the observation that the amplitude of \( m \)-photon detachment of the initial state \( \Phi_a \) within the Keldysh model is

\[ A_m(p) = \frac{1}{T} \int_0^T dt' \langle \Phi_p(t') \mid V_F(t') \mid \Phi_a(t') \rangle. \quad (9) \]

In the right hand side of (9) 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. Namely, the absolute value of \( p \) is given by the right hand side of the formula (7), where \( F^2/(4\omega^2) \) is the electron quiver energy in the laser field. This shows that the vector \( K_m \) entering the representation (6) of HG amplitude component is exactly the physical electron translational momentum in the \( m \)-th ATI channel, but with the specific directions (8).

From the Volkov state \( \Phi_p \) the electron can recombine back to the bound state \( \Phi_a \) with emission of the photon of frequency \( \Omega \). This process with the amplitude

\[ C_{Nm}(p) = -\frac{1}{2\pi T} \int_0^T dt \langle \Phi_a(t) \mid \exp(i\Omega t) \hat{d}_e \mid \Phi_p(t) \rangle \quad (10) \]

is possible only in the presence of the laser field from which the necessary \( N - m \) low-frequency quanta \( \omega \) are gained, that justifies its name stimulated recombination.

One readily notices that the integrand in (6) bears a striking resemblance to the product of the integrands in (9) and (10). However, the complete separation of integrations in \( t \) and \( t' \) variables is prevented by the factor \( 1/R(t, t') \) stemming from the chain of equations

\[ R(r, r'; t, t') = (F/\omega^2) (\cos \omega t - \cos \omega t') + r - r' \approx \]

\[ \approx R(t, t') \equiv (F/\omega^2) (\cos \omega t - \cos \omega t'). \quad (11) \]
The latter approximation is applicable provided $F/\omega^2$ exceeds the typical dimensions of localization of the active electron in the initial state $\Phi_a$, that holds in most practical situations. Classically $R(t, t')$ gives the distance between the electron positions at the moments $t$ and $t'$ due to electron wiggling in the laser field. $1/R$ could be named an expansion factor since in the absence of the laser field it describes conventional decrease of the amplitude in a spherical wave as it expands in 3D space. When the laser field is operative, the form of the expansion factor is drastically modified according to Eq.(11). Hence the interpretation of the expression (6) 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 contributions of all paths labeled by $m$ add up coherently as shown by Eq.(5).

Following the factorization technique (Kuchiev 1995) we simplify Eq.(6) further on by performing the $t'$ integration by the saddle point method. This is justified since the integrand in (6) contains a large phase factor $\exp\left[iS(t')\right]$ with the classical action

$$S(t') = \frac{1}{2} \int_{t'}^{t'} dt \left[p + \left(F/\omega\right) \sin \omega t\right]^2 - E_a t' \ .$$

The saddle point positions $t' = t'_{m\mu}$ in the complex $t'$ plane are defined by the equation

$$S'(t'_{m\mu}) = 0 \ .$$

Similar adiabatic approximation in a simpler case of photodetachment casts the ATI amplitude (9) as

$$A_{mL}(\mathbf{p}) = \sum_{\mu=1,2} A_{m\mu}(\mathbf{p}) \ ,$$

$$A_{m\mu}(\mathbf{p}) = -\frac{1}{\omega} A_a Y_{lm}(\mathbf{p}) \sqrt{\frac{2\pi i}{S''_{\mu}}} \exp\left(iS_{\mu}\right) \ .$$

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 $\text{Im}t_{m\mu} > 0$. Only these two saddle points are operative in the contour integration being included into the summation over $\mu = 1,2$. In (15) $l$ is the active electron orbital momentum in the initial state, $\kappa = \sqrt{-2E_a}$, $A_a$ is the coefficient in the wave function $\phi_a(\mathbf{r})$ asymptote. For more details see the papers by Gribakin and Kuchiev (1997a,b), where the approximation based on (14)-(15) was demonstrated to be very efficient and accurate for multiphoton detachment.

After carrying out the $t'$ integration in (17) by the saddle point method we arrive to our major result

$$d_N = 2 \sum_m A_{m\mu}(\mathbf{K}_m) B_{N_{m\mu}}(\mathbf{K}_m) \ .$$
where the factor

$$ B_{N,m}(K_m) = -\frac{1}{2\pi T} \times $$

$$ \times \int_0^T dt \frac{\langle \Phi_a(t) | \exp(i\Omega t) \hat{d}_e | \Phi_{K_m}(t) \rangle}{(F/\omega^2)(\cos \omega t - \cos \Omega t)} $$

(17)

describes jointly the 3D-wave expansion and SR. These two effects could be further factorized using the approximation $|\cos \omega t_m| \gg |\cos \Omega t|$ (Kuchiev 1995):

$$ B_{N,m}(K_m) = \frac{1}{R_m} C_{N_m}(K_m), $$

(18)

where $1/R_m = \omega^2/(F \cos t'_m)$ is the laser-modified expansion factor in its simplest form.

Now it is worthwhile to comment more on the physics of HG as implemented in Eq.(16). On the first stage of the three-step process the electron absorbs $m$ laser photons with the amplitude $A_{m,\mu}$. In order to contribute to HG the photoelectron has to return to the parent atomic core where SR is solely possible. The amplitude of return is described by the expansion factor $1/R$. At the third step the electron collides with the core virtually absorbing $N - m$ photons from the laser field and emitting the single high-frequency quantum $\Omega$ as it recombines to the bound state. This SR process has the amplitude $C_{N_m}$. The summation over $m$ in the total amplitude $d_N$ (16) takes into account interference of the transitions via different intermediate ATI channels.

The nontrivial point 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 (Gribakin and Kuchiev1997a,b), 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 $t'_m$ 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 (14)-(15) (Gribakin and Kuchiev 1997a,b, Kuchiev and Ostrovsky 1998) 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 $t'_m$ only one of the two possible directions of $K_m$, labeled in (5) 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. In other words, for each direction of $\mathbf{K}_m$ 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 (16).

The practical calculations of $B_{Nm\mu}$ or $C_{Nm}$ can be conveniently performed using the Fourier transform of the bound state wave function $\phi_a$. After that one has to carry out a single numerical integration over the finite interval of time $t$, or, alternatively, to resort to the saddle-point method that provides purely analytical formulæ.

Based on physical arguments, we extend the summation in (16) only over open ATI channels with the real values of $K_m$. We present the rates of generating the $N$-th harmonic radiation

$$R_N \equiv \frac{\omega^3 N^3}{2\pi c^3} |\langle \mathbf{d}_N \rangle|^2$$

as introduced by Becker et al (1994) (and denoted by these authors as $dR_N/d\Omega_K$); $c$ is the velocity of light. Some typical results for the HG by $\text{H}^-\text{ion}$ in the $\omega = 0.0043$ laser field are shown in Fig. 1 for the smallest and largest field intensities considered in the paper by Becker et al (1994). We take the binding energy corresponding to that of $\text{H}^-\text{(}\kappa = 0.2354\text{)}$, but replace the true value $A_a = 0.75$ (Radzig and Smirnov 1985) by unity since this corresponds to the zero-range potential model used by Becker et al (1994). For the real $\text{H}^-\text{ion}$ the results shown in Fig. 1 are to be scaled by a factor $A^4_a N_e$, where $N_e = 2$ accounts for the presence of two active electrons in $\text{H}^-\text{.}$

The HG spectrum is known to consist generally 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). On the large-$N$ side our results employing Eq.(17) coincide with those obtained by Becker et al (1994) within the plot scale. The deviations increase as $N$ decreases, albeit remarkably the structures in $N$-dependence of the rates are well reproduced. The approximation (18) somewhat overestimates HG rate, but still retains the structure, though smoothed.

In the summation over ATI channels (i.e. over $m$) the coherence is very important, since the large number of terms is comparable in modulus, but have rapidly varying phases. Many ATI channels contribute to HG for each $N$ (contrary to tentative conclusion by Eberly et al (1989)).

Although the length gauge is known to be superior for the description of ATI within the adiabatic approximation (Gribakin and Kuchiev 1997a), the situation is not that straightforward for the high-energy photon. Therefore our calculations were reiterated with replacement of the $\hat{d}_e$ operator in (17) by its dipole-velocity counterpart. The agreement between the two forms proves to be very good, see Fig. 1.

As a summary, the three-step mechanism of the harmonic generation is ultimately justified. It is implemented in fully quantum relations expressing its amplitude via
amplitudes of the above-threshold ionization and stimulated recombination. 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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Figure 1. Harmonic generation rates (19) (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 (1994), open circles - present calculations in the dipole-length gauge using the expression (17) for $B_{Nm\mu}$, open squares - same but with the simplified formula (18) for $B_{Nm\mu}$; open triangles - present calculations in the dipole-velocity gauge. The symbols are joined by lines to help the eye.
\[ \frac{z \omega_m}{M} I_{10} = I \]