Tailoring the waveforms to extend the high-order harmonic generation cutoff

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Increase in the cutoff value in the high-order harmonics generation process is demonstrated for a special case of the driving field composed of several harmonics of a given frequency. It is shown that a moderate, of the order of 20%, increase in the cutoff value can be achieved. This result possibly constitutes an upper limit for the increase in the cutoff value, attainable for a class of the waveforms considered in the paper.

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I. INTRODUCTION

High-order harmonic generation (HHG) is a nonlinear atomic process which can be described using a simple classical picture [1–3]. Driven by a strong electromagnetic (EM) field, the atomic electron emerges into the continuum with zero velocity at some particular moment of time. At a later time, the classical electron trajectory returns to the nucleus, where the electron can recombine and emit a photon. The frequency of the emitted photon is determined by the amount of energy acquired by the electron and the atomic ionization potential (assuming that the electron recombines to the ground state). The classical analysis shows that, for the monochromatic EM field of the form $F_0 \cos \Omega t$, the kinetic energy of the electron returning back to the nucleus cannot exceed the value of $3.17U_p$, where $U_p = F_0^2/(4\Omega^2)$ is the ponderomotive potential. This leads to the well-known $I_p + 3.17U_p$ cutoff rule for the maximum harmonic order. Here, $I_p$ is the atomic ionization potential.

The quantum counterpart of the classical model [4] assumes, that the released electron moves only under the action of the EM field neglecting the influence of the atomic potential, the so-called strong-field approximation (SFA) [4,5]. The SFA employs the analytical Volkov states, which makes the problem tractable. The classical returning trajectories emerge as extrema in the saddle-point analysis of the quantum-mechanical amplitudes computed within the SFA [4].

The aforementioned classical and quantum-mechanical results correspond to the pure cosine form of the driving EM field. Available pulse-shaping techniques [6] make it possible to modify the HHG characteristics by suitably tailoring the driving EM field. This problem belongs to a rapidly developing field of the quantum optimal control [7].

Several aspects of the optimal control of the HHG process were addressed in the literature. In the paper [8], the emission intensity of a given harmonic order was optimized by tailoring the laser pulse. In Ref. [9], the emphasis was placed on optimizing the particular high-order harmonics from which single attosecond pulses could be synthesized. Both these works employed the so-called genetic algorithm, which mimicked the natural selection process by introducing the mutation procedure and suitable fitness function emphasizing the desired properties of the target state. Numerically, this procedure requires multiple solution of the time-dependent Schrödinger equation (TDSE), which may constitute a considerable computational task if one is interested in formation of HHG in real atomic systems.

If the desired goal is to increase the harmonics cutoff order, there is a possibility to find the optimum field parameters using a purely classical approach based on the electron trajectory analysis [10,11]. In the paper by Radnor et al. [10] such an analysis, supplemented by the quantum calculation relying on the genetic algorithm, was used to show, that the optimum waveform allowing to maximize the recollision energy is a linear ramp with the DC offset $F(t) = a t + \beta$ for $t \in (0, T)$, $T$ being the period of oscillations. Such a form has been shown to provide an absolute maximum of the kinetic energy of the electron at the moment of its return to the nucleus. This energy was approximately 3 times larger than the corresponding energy for the pure cosine wave with the same period and field intensity [11]. To avoid using a strong DC field in practice, it was suggested in [11], that it could be replaced by an AC field of a lesser $\Omega/2$ frequency, while the linear ramp could be replaced by a combination of the harmonics with frequencies $n\Omega$. Here, $\Omega$ is the frequency corresponding to the oscillation period $T$, $n$ is integer. The overall pulse has thus a period of $2T$, rather than $T$. The weights corresponding to different harmonics constituting the pulse were found by means of the genetic algorithm. Results of the quantum calculation relying on SFA reported in [11] confirmed, that this waveform allowed to achieve considerable increase in the cutoff position.

In the present work, we address a related question: what gain in the HHG cut-off can be achieved if we use the driving EM field with the waveform composed of the harmonics with the multiple frequencies $n\Omega$. In other word, we demand the driving EM pulse to be strictly $T$ periodic and such that its integral over a period is zero (i.e., no DC component is present). It turns out that a moderate increase in the cutoff position is possible in this case. A simpler case of adding the second harmonic $2\Omega$ to the waveform was considered in earlier works [12,13].

We supplement the classical trajectory analysis by a quantum mechanical TDSE calculation of the HHG process in the lithium atom. Choice of this particular target was motivated by the experiments on the laser field ionization of magnetically trapped (MOT) Li atoms [14]. Numerical solution

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of TDSE takes full account of the effect of the atomic potential. Such a calculation ensures, that the effect of the extended HHG cutoff, which we report below, is not an artifact of a simplified treatment.

We shall consider below EM fields for which the field amplitude $F(t)$ is a periodic function of time, having a fixed period $T$. The field intensity for such EM fields can be expressed as $W=\frac{c}{\Omega}f_0^2 F(t)^2 dt$, where $c$ is the speed of light. For the monochromatic EM field $F(t)=F_0 \cos \Omega t$ this gives the well-known relation $W=\frac{F_0^2}{\pi \Omega}$. Throughout the paper, we shall use the atomic units. The unit of the EM field intensity corresponding to the unit field strength $F_0=1\ \text{a.u.}=5 \times 10^9 \ \text{V cm}^{-1}$ is $3.51 \times 10^{16} \ \text{W cm}^{-2}$ [15]. The field intensity of the monochromatic wave $F_0 \cos \Omega t$ can thus be expressed as $W=3.5 \times 10^{16} F_0^2$, if field intensity is measured in W/cm$^2$ and the field strength is expressed in the atomic units. From the expressions above it is clear, that $T$-periodic EM fields with different $F(t)$ but equal values of $\int_0^T F(t)^2 dt$, will have the same intensities. In particular, EM field $F(t)$ will have the same intensity as the monochromatic wave $F_0 \cos \Omega t$ of the same period if $\int_0^T F(t)^2 dt=TF_0^2/2$.

II. THEORY

A. Classical approach

We begin with a purely classical problem of finding returning trajectories of an electron moving in a periodic EM field with a given period $T$, corresponding frequency $\Omega =2\pi/T$, and which does not contain a DC component:

$$F(t) = 2 \Re \sum_{k=1}^{K} a_k e^{i\Omega t}.$$  \hspace{1cm} (1)

The field is assumed to be linearly polarized along the $z$ axis. Our task is to find the set of coefficients $a_k$ in Eq. (1) for which electron returning to the nucleus possesses the highest possible kinetic energy. For this problem to be well defined, we must impose some restrictions on the possible choice of this set. A natural requirement is that only the fields $F(t)$ of the same intensity are to be considered. This implies that $\sum_{k=1}^{K} |a_k|^2 = F_0^2$, where $F_0$ is amplitude of the monochromatic waveform $F_0 \cos \Omega t$ having the same intensity.

As it is customarily done in the classical three-step model of HHG, we neglect the influence of the atomic core on the electron motion. We solve the classical equations of motion of electron in the EM field with the initial conditions $z(t_0)=0, \ z'(t_0)=0$. Here, $t_0$ is the moment of time when the atomic ionization event occurs. In the classical calculation, we do not introduce any envelope function to describe the EM field, i.e., as a driving force in the classical equations of motion, we use the flat envelope pulse of infinite duration. This is permissible, since in the quantum calculation presented in the next section we shall use a pulse long enough, so that all transient effect, as well as all effects due to the finite duration of the pulse (such as dependence on the carrier phase) become unimportant. The results of both calculations can, therefore, be legitimately compared, and, as we shall see, will give qualitatively similar results.

FIG. 1. (Color online) Classical dependence of the quantity $(E + IP)/\Omega$ on the time of electron release within an optical cycle $(E$-electron energy at the moment of return to the nucleus, $IP=0.196 \ \text{a.u.}$-ionization potential of the Li atom). The three sets of curves correspond, respectively, to the pure cosine wave-solid (red) line; odd harmonics in Table I-dashed (green) line; odd and even harmonics in Table I-short (blue) dash.

We are interested only in the returning trajectories for which $z(t_0)=0$ for some $t_0$. For such trajectories, we compute the kinetic energy $E$ at the moment of return.

We use the following field parameters: $I=10^{12} \ \text{W cm}^{-2}$, $F_0=0.0053 \ \text{a.u.}, \ \Omega=0.185 \ \text{eV} \ (6.705 \ \text{mcm})$. In this and the subsequent section we consider the case of the Li atom with the ionization potential $I_p=0.196 \ \text{a.u.}$ for this set of the field and atomic parameters, the value of the Keldysh parameter $\gamma=\sqrt{I_p}/2U_p=0.8$.

Our choice of the field parameters was motivated, primarily, by the following reason. We need to choose a combination of $F_0, \ \Omega$, and $I_p$ such that the picture of the HHG process [4], which establishes the connection of HHG with returning classical trajectories remained valid. Among the conditions of the validity of this picture are the requirements, that depletion of the ground state can be ignored, and that the value of the Keldysh parameter should be less than one [4]. For the lithium atom, with its small ionization potential, we have a rather narrow corridor of the field parameters, which satisfy both these requirements. For the field parameters thus defined we have the value $F_0/\Omega^2 \approx 115$ a.u. for the excursion radius of electron motion in the EM field of the cosine form $F_0 \cos \Omega t$. Similar values for the excursion radius are obtained for all EM fields given by Eq. (1) we consider below. Thus, the electron moves predominantly far from the nucleus, and neglect of the Coulomb potential in the classical equations of motion is legitimate.

For the pure cosine form of the EM field, the classical procedure described above leads to the typical dependence of the kinetic energy at the moment of return on the time of release shown in Fig. 1 by the solid (red) line. For convenience, in Fig. 1 we plot not just the kinetic energy itself, but the quantity $N=(E+IP)/\Omega$, which gives us the order of the harmonic corresponding to given kinetic energy $E$. The solid curve in Fig. 1 shows that, for the parameters we chose, the maximum harmonic order is approximately $N_{\text{max}} \approx 100$, which is a visualization of the well-known $I_p+3.17U_p$ cutoff rule.

For the set of parameters in Eq. (1), defining the EM field different from the pure cosine wave, we proceed as follows.
TABLE I. Coefficients in Eq. (1) for which the highest kinetic energy of the returning electron is maximized. The second column: pure cosine wave; the third column: odd harmonics with $K=7$; the fourth column: odd and even harmonics with $K=5$

<table>
<thead>
<tr>
<th>$k$</th>
<th>Cosine wave ($10^3a_k$)</th>
<th>Odd harmonics ($10^3a_{2k+1}$)</th>
<th>Odd and even harmonics ($10^3a_k$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2.665</td>
<td>2.503−0.076i</td>
<td>2.123−1.033i</td>
</tr>
<tr>
<td>2</td>
<td>0</td>
<td>−0.443+0.566i</td>
<td>0.403+0.754i</td>
</tr>
<tr>
<td>3</td>
<td>0</td>
<td>0.061−0.385i</td>
<td>−0.558+0.271i</td>
</tr>
<tr>
<td>4</td>
<td>0</td>
<td>0.138−0.264i</td>
<td>−0.302−0.358i</td>
</tr>
<tr>
<td>5</td>
<td>0</td>
<td>0</td>
<td>0.224−0.248i</td>
</tr>
</tbody>
</table>

For each set of parameters in Eq. (1), subject to the constraint of the fixed intensity, so that the field intensity had the same value as in the case of the pure cosine wave, we can compute a maximum kinetic energy of the returning electron. This gives us a function defined on the set of the parameters $a_k$ in Eq. (1). We look for the maximum of this function using the gradient ascent method, giving as a starting values of the independent variables some particular set of the parameters in Eq. (1), satisfying the fixed intensity constraint. This procedure is guaranteed to converge to a local maximum. Since the convergence is generally quite fast, and requires only modest computational effort, we can repeat the procedure many times with different starting values, until we can be reasonably sure, that we have found the global maximum.

We perform two calculations of this kind. In the first calculation, we impose an additional restriction that only the terms with odd $k$ values are to be present in Eq. (1). This ensures that the resulting HHG spectrum contains only odd harmonics of the main frequency. In the second calculation, we retain the terms with both odd and even $k$ values in the expansion (1). In this case, the resulting HHG spectrum contains even harmonics as well, since symmetry of the Hamiltonian, which for the case when only odd harmonics are present in (1), leads to only odd harmonics in the HHG spectrum, is broken by superimposition of fields of $\Omega$ and $2\Omega$ frequencies [16].

The first calculation was performed with $K=7$, while in the second we chose $K=5$. The resulting sets of coefficients $a_k$ for which the maximum of the highest kinetic energy of the returning electron is attained, are presented in Table I. Also presented is the set consisting of only $a_1$, which defines the pure cosine wave for the field parameters considered above.

The degree to which this procedure increases the highest energy of the returning electron is illustrated in Fig. 1. Resulting shapes of the driving field $F(t)$, corresponding to the three cases considered above are visualized in Fig. 2.

As one can see, the set of the parameters corresponding to only odd harmonics present in Eq. (1) allows to achieve a 10% gain in the position of the cutoff. The curve representing dependence of the kinetic energy on the time of release remains symmetric with respect to the translation $t \rightarrow t + T/2$, as in the case of the pure cosine wave. This is, in fact, a general property exhibited by the classical solutions of the equations of motion in the EM field with only odd harmonics present in Eq. (1), which leads to essentially the same structure of the classical returning electron trajectories as in the case of the cosine wave. There are two such trajectories per every half cycle of the EM field (the so-called “long” and “short” trajectories) for the plateau region, i.e., for the kinetic energies below the apex of the corresponding curves in Fig. 1. There is one trajectory per every half cycle with the kinetic energy of the returning electron near the apex of the curves (the cutoff harmonics).

Situation is different for the case of even and odd harmonics present in Eq. (1). The kinetic energy curves are no longer symmetric with respect to the half cycle translation $t \rightarrow t + T/2$.

One should note, that increase in the cutoff position shows very little sensitivity to further increase in the number of terms in Eq. (1). If, for example, we used $K=9$ instead of $K=7$ in the case of only odd harmonics included in Eq. (1), we would have gained additional increase in the cutoff position of the order of 1%. Similar observation applies for the case of even and odd harmonics in Eq. (1). This indicates, that the low-order harmonics in the series (1) are primarily responsible for the increase in the cutoff position, and the pulses composed using the coefficients in Table I are optimal in the sense that no further significant increase in the cutoff position is possible as long as we rely on the expansion (1) for the waveform.

The discussion presented so far was purely classical and constituted a simple generalization of the three-step model for the case of the EM field given by Eq. (1). Quantum calculation is needed to confirm the classical results. Such calculation is presented in the next section.

### B. Quantum calculation

In this section, we present results of the HHG calculation for the Li atom for the set of coefficients $a_k$ given in Table I. We use the procedure, which we developed recently in Ref. [17] for the solution of TDSE for realistic atomic targets, which can be described within the single active electron approximation. For completeness, most essential features of this procedure are outlined below.
The field-free atom in the ground state is described by solving a set of self-consistent Hartree-Fock equations [18]. The field-free Hamiltonian \( \hat{H}_{\text{atom}} \) in this model is thus a non-local integrodifferential operator.

The EM field is chosen to be linearly polarized along the \( z \) axis. We describe the atom-EM field interaction using the

\[
\hat{H}_{\text{int}} = \sum_{l} \frac{\alpha l}{\hbar^2} F_{l}^2(r) \frac{\partial^2}{\partial r^2} + \sum_{l} \frac{\beta l}{\hbar^2} F_{l}^2(r) \left( \frac{\partial}{\partial r} \right)^2 \tag{1}
\]

function \( F_{l}^2(r) \) smoothly grows from 0 to 1 on a switching interval \( 0 < t < T_1 \), and is constant for \( t > T_1 \). The switching time is \( T_1 = 5T \).

We represent solution of TDSE in the form of an expansion on a set of the so-called pseudostates:

\[
\Psi(r,t) = \sum_{j} b_{j}(t) f_{j}(r). \tag{2}
\]

This set is obtained by diagonalizing the field-free atomic Hamiltonian on a suitable square integrable basis [19,20]:

\[
\langle f_{j}^{\text{\text{\textsc{n}}}}|\hat{H}_{\text{\text{\textsc{atom}}}^{\text{\text{\textsc{n}}}}}|f_{j}^{\text{\text{\textsc{n}}}} \rangle = E_{i} \delta_{ij}. \tag{3}
\]

Here, the index \( j \) comprises the principal \( n \) and orbital \( l \) quantum numbers, \( E_{i} \) is the energy of a pseudostate and \( N \) is the size of the basis. To construct the set of pseudostates satisfying Eq. (3), we use either the Laguerre basis, or the set of \( B \) splines (for angular momenta \( l > 15 \)), confined to a box of a size \( R_{\text{max}} = 200 \) a.u. \( B \) splines of the order \( k = 7 \) with the knots located at the sequence of points lying in \([0,R_{\text{max}}]\) are employed. All the knots \( t_{i} \) are simple, except for the knots located at the origin and the outer boundary \( R = R_{\text{max}} \) of the box. These knots have multiplicity \( k = 7 \). The simple knots were distributed in \((0,R_{\text{max}})\) according to the rule \( t_{i+1} = \alpha t_{i} + \beta \). The parameter \( \alpha \) was close to 1, so that the resulting distribution of the knots was almost equidistant. For each value of the angular momentum \( l \), the first \( l + 1 \) \( B \) splines and the last \( B \) spline resulting from this sequence of knots were discarded. Any \( B \) spline in the set thus decreases at least as fast as \( r^{l+1} \) and assumes zero value at the outer boundary.

In the present calculation, the system is confined within a box of a finite size which may lead to appearance of spurious harmonics in the spectrum due to the reflection of the wave packets from the boundaries of the box [2]. One can minimize this effect by using a mask function or an absorbing potential. We use the absorbing potential \(-iW(r)\) which is a smooth function, zero for \( r \leq 180 \) a.u. and continuously growing to a constant \(-iW_{0}\) with \( W_{0} = 2 \) a.u. outside this region.

For the EM field parameters, which we employed in the classical treatment of the previous section, the maximum harmonic order is of the order of a hundred. This implies that to describe accurately formation of all harmonics, we have to retain pseudostates with correspondingly high-angular momenta. In the calculation we present below, the pseudostates with angular momenta \( l < 120 \) were retained in Eq. (2).
FIG. 6. Part of the spectrum of Li for odd and even harmonics in Eq. (1) with $K=5$.

FIG. 5. Harmonics spectra of Li for the EM fields from Table I. Odd and even harmonics in Eq. (1) with $K=5$ [red solid line], classical cutoff position marked with the (green) dashed line.

The same intensity. Cutoff position increases yet further for the pulse constructed using the set of the coefficients from the fourth column of Table I [odd and even harmonics with $K=5$ in Eq. (1)]. In this case, the spectrum contains harmonics of both odd and even orders. A magnified fragment of the spectrum illustrating this fact is shown in Fig. 6.

Quantitatively, the TDSE results for the cutoff positions are in agreement with the classical predictions, summarized in Fig. 1. Use of the pulse constructed from all harmonics with $K=5$ in Eq. (1) allows to increase the cutoff position by about 20%, in agreement with the classical analysis given above.

We can, in fact, establish a closer correspondence between classical and quantum results by performing the time-frequency analysis of our data. The techniques used for this purpose, the wavelet transform [8,22–24], or the closely related Gabor transform [22,25], offer possibility to track the process of harmonics formation in time, combining both the frequency and temporal resolution of a signal. By using these techniques, we can try to find, in the quantum domain, the traces left by the classical trajectories. The fact that such traces may be present, follows from the quantum-mechanical treatment of the HHG process given in [4], where the classical trajectories naturally appear in the saddle-point analysis. Such manifestation of the classical trajectories in the HHG spectra was demonstrated, for example, for the hydrogen atom [24].

We perform our analysis of the HHG process by applying the wavelet transform of the dipole operator $d(t)$ in Eq. (4). This transform is defined as [26]

$$T_{\psi}(\omega, \tau) = \int d(t) \sqrt{\alpha}|\Psi^\dagger(\omega t - \omega \tau)|^2 dt.$$  (5)

The transform is generated by the Morlet wavelet $\Psi(x) = x_{\omega_0}^{1/2} \exp(-ix/\omega_0)\exp(-x^2/2\omega_0^2)$.

Figure 7 presents a well-known picture of the harmonics formation in time [24]. For the plateau harmonics, the amplitude of the wavelet transform has four maxima per cycle, corresponding to the two pairs of the so-called long and short trajectories for the harmonics at the plateau. For the near cutoff 101st harmonic, two maxima per cycle are present. Those features agree completely with the classical picture shown in Fig. 1.

For the pulse containing only odd harmonics in Eq. (1), classical picture of the dependence of kinetic energy on the time of release, presented on Fig. 1, is very similar to the curve for the pure cosine wave. We can expect, therefore, results of the wavelet transform in this case to be qualitatively similar to those shown on Fig. 7. That this is indeed the case can be observed from Fig. 8.

For the field waveform containing both odd and even harmonics in Eq. (1), the classical analysis reveals a somewhat different picture. As one can see from Fig. 1, there are two pairs of the classical trajectories per cycle for which kinetic energy of the returning electron is such, that less than approximately 60 harmonics can be formed. When the harmonic order increases and reaches the value of approximately 75 (cutoff region for the smaller maximum of the corresponding curve in Fig. 1), there are three returning trajectories per cycle. For higher energies, there remain only two classical trajectories, which can participate in the forma-
terms with both odd and even order of 20%. This value represents a maximum increase in the value of the maximum classical kinetic energy of the subharmonic terms in expansion of the harmonics. For higher yet energy, a single such trajectory exists.

As can be observed from Fig. 9, the quantum calculation apparently confirms these classical considerations. Wavelet spectra do demonstrate that number of maxima per cycle progressively decreases with the increase in the harmonics order.

**IV. CONCLUSION**

We demonstrated an increase in the cutoff value for the HHG process when a superposition of several harmonics of a given frequency is used to build a waveform of the driving EM field. We analyzed the classical returning electron trajectories for the fields thus constructed. Such an analysis shows, that a field spectral composition can be found, for which a 20% increase in the value of the maximum classical kinetic energy of the recombining electron is achieved as compared to the case of a cosine wave of the same intensity.

TDSE calculation of the HHG spectrum for such a driving field, performed for the Li atom, confirms the classical result. It does demonstrate the increase in the cutoff value of the order of 20%. This value represents a maximum increase which can be achieved if we restrict the trial waveform to that given by Eq. (1) under condition of a fixed intensity. Indeed, the classical calculation shows that no further noticeable increase in the maximum classical kinetic energy of the recombining electron can be achieved by adding higher order harmonic terms in expansion (1).

Our result thus presents an upper limit in the increase in the HHG cutoff achieved for the class of the waveforms given by Eq. (1), i.e., for the waveforms which are periodic with a given period $T$ and do not contain the static components. This suggests, that to achieve more substantial increase in the HHG cutoff condition, one should use the waveforms which cannot be described by Eq. (1). Such are the ideal waveform proposed in [10], for which we should allow the term with $k=0$ to the sum in Eq. (1), or the field configurations containing subharmonic fields with frequencies $\Omega/2$, as those used in [11,27]. As results of these works indicate, a considerably more important gain in the cutoff energy can be achieved for such waveforms. These results, and the result obtained in the present work, allow us to draw the following conclusion. The strategy based on the low-frequency (subharmonic) modifications of the waveform may be more efficient than the strategy relying on introducing multiple-frequency components in the trial waveform as in Eq. (1). This may provide a useful guide to the problems related to modification of the high frequency part of the HHG spectrum.

The time-frequency analysis of the results of the TDSE calculation illustrates the role, which the classical trajectories play in the formation of the harmonics. The usual picture of HHG rendered by this technique exhibits traces of four (for the plateau harmonics) or two (harmonics near cutoff) trajectories per optical cycle, which participate in forming a particular harmonic. In the case of the waveform constructed from the terms of odd and even order in Eq. (1), the picture revealed by the wavelet analysis is different. Number of contributing trajectories in this case varies with energy in agreement with the classical picture of Fig. 1. Depending on the harmonics order, there may be four, three, two, or just a single such trajectory.

For a single atom, each such a trajectory leads to the formation of a short burst of EM radiation, producing a pulse train. In the case of the HHG driven by the single color $T$-periodic EM field, such a train is a $T/2$ periodic sequence of bursts, with two bursts on each interval of the length $T/2$, corresponding to the short and long trajectories within a half cycle. For each harmonic order, the contributions of these two trajectories interfere strongly, leading to the random distribution of the phases of different harmonics in the plateau region.

This situation is changed [28] if propagation effects are taken into account. Depending on the particular propagation geometry, one of the contributions (of either short or long trajectories) is suppressed, the propagated harmonic components become locked in phase, and the macroscopic signal is a train with one pulse per every half cycle.

For the case of the waveform with only odd harmonics in Eq. (1), propagation should have exactly the same effect, as for the single color field. For this waveform, the classic curve in Fig. 1 has exactly the same form as in the single color case, giving rise to the same set of long and short trajectories per every half cycle of the laser field. Analysis given in the work [28] shows, that propagation effects reduce contribution of one of the trajectories since their phases change differently with laser intensity, and hence behave differently in the nonlinear medium. Depending on the particular geometry, contribution of one of the trajectories can thus be reduced. The curve in Fig. 1 suggests, that in the case of the waveform with only odd harmonics in Eq. (1) we should have analogous situation.

For the case of the waveform with odd and even harmonics in Eq. (1), the pulse train produced by the single-atom is no longer $T/2$ periodic, but $T$ periodic. This is clearly seen from the terms of odd and even order in Eq. (1).
from Fig. 1. It is, of course, also obvious from the fact, that even harmonics are present in the HHG spectrum in this case, separation of the harmonics is not 2Ω but Ω, consequently the signal is a T-periodic function. On each of the intervals of length T we have, depending on the number of the classical trajectories four, three, two, or a single pulse of different intensities. For the harmonics with orders N > 80, when, as seen from Fig. 1, there are only two trajectories to consider, propagation should produce essentially the same effect as in the case of the single color field. These two trajectories interfere, their phases depending differently on the laser intensity. Thus, as in the single color case, propagation may reduce contribution of one of these trajectories, making harmonics phase locked. The macroscopic signal will be in this case a train with one pulse per every cycle.

For the lower order harmonics, when all four trajectories contribute with different amplitudes and phases, situation is more complicated. It can hardly be expected, that propagation effects may suppress contributions of all but one trajectory, and thus eliminate the interference of the contributions due to different trajectories completely. The harmonics, therefore, may not be locked in phase in this case.

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