Transition from perturbative to nonperturbative interaction in low-order-harmonic generation

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We present results of *ab initio* numerical calculations for low-order-harmonic generation as well as calculations of the higher-order terms in the respective perturbative power-series expansions of the susceptibilities for third- and fifth-order-harmonic generation. We find that the transition from perturbative to nonperturbative interaction in these low-order nonlinear processes occurs at about 10^{13} W/cm^2. Our findings confirm previous results that any deviation from the predictions of lowest-order perturbation theory indicates that the perturbative series expansion is not applicable and, if required, needs to be replaced by a nonperturbative treatment of the interaction between the atom and the field. In particular, the results also show that the observation of low-order-harmonic yields cannot be considered as a test of higher-order Kerr effects.

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I. INTRODUCTION

High-order-harmonic generation in intense short laser pulses is a highly nonlinear process, which has been extensively studied as a route to generate coherent bright x rays and attosecond pulses (for a review, see, e.g., Ref. [1]). On the other hand, low-order-harmonic generation (also, termed below-threshold harmonics) in the strong-field regime has received less attention. However, recent experiments [2–4] have demonstrated the potential to generate bright coherent low-order harmonics in the vacuum ultraviolet with photon energies below the threshold of the ionization potential of the target atom. These ultrafast sources have gained interest as tools for ultrafast spectroscopy of electron wave-packet dynamics in atoms and molecules [3] as well as for precision measurements [2,4].

Beyond this renewed interest in low-order-harmonic generation for spectroscopic purposes, it has been proposed [5] that the ratio of fifth- to third-order-harmonic generation offers a stringent test to the controversially discussed role of higher-order Kerr effects (HOKE) (for a review, see, e.g., Ref. [6]). The HOKE debate was initiated by the consideration that higher-order terms in the perturbative power-series expansion of the electric susceptibility χ_ω of a gas in an external electric field $\mathbf{E}$ [7],

$$\chi_\omega = \chi_\omega^{(1)} + \chi_\omega^{(3)}|\mathbf{E}|^2 + \chi_\omega^{(5)}|\mathbf{E}|^4 + \chi_\omega^{(7)}|\mathbf{E}|^6 + \cdots \quad (1)$$

are required in order to explain the experimentally observed [8,9] (and, previously, theoretically predicted [10,11]) negative slope in the electric susceptibility as a function of the peak laser intensity in the middle of 10^{13} W/cm^2.

Our recent theoretical studies of the electrical susceptibility of atomic hydrogen have indicated [12] that the power-series expansion Eq. (1) does not converge at intensities above about 2×10^{13} W/cm^2 since the magnitudes of the higher-order terms do exceed a significant fraction of the lowest-order nonlinear term. The calculations of the higher-order coefficients were performed using a numerical basis-state method [13], which also enabled us to determine the nonlinear electrical susceptibility from the *ab initio* solution of the corresponding Schrödinger equation. Comparison of the results of both kinds of calculations let us conclude that a change in the intensity dependence of the susceptibility has to be interpreted as a signature of the nonperturbative interaction between the intense laser light and the gas, whereas perturbative concepts, such as HOKE, are not applicable.

In view of the renewed interest in strong-field below-threshold harmonic generation and the proposed studies regarding third- and fifth-order-harmonic generation in the HOKE debate, it is interesting to ask whether or not similar conclusions regarding the transition from perturbative to nonperturbative interaction hold for low-order-harmonic generation as well. To this end, we extend our previous studies on the electrical susceptibility [12] and use the numerical basis-state method to determine the first few coefficients of the power-series expansion of the elements of the susceptibility tensor for third- and fifth-order-harmonic generation. We also calculate the harmonic spectrum generated by the interaction of a short laser pulse with atomic hydrogen using the direct numerical solution of the corresponding time-dependent Schrödinger equation. This allows us to perform an *ab initio* study of the generated power for each harmonic as a function of the peak laser intensity at the single-atom level. The rapidly increasing contribution of the higher-order terms as well as the deviation of the intensity dependence of the harmonic power from the power law, expected from perturbation theory, enable us to estimate that the corresponding breakdown of the perturbative power-series expansion occurs in the same intensity regime as for the electrical susceptibility.

II. THEORETICAL APPROACHES

Despite providing complimentary insight, our perturbative and *ab initio* methods are based on the same theoretical framework, namely, a set of numerically obtained field-free energy eigenstates, here for atomic hydrogen written as (Hartree atomic units, $e = \hbar = m = 1$, are used throughout)

$$|\psi_{nlm}(\mathbf{r})\rangle = |R_{nl}(r)Y_{lm}(\Omega)\rangle, \quad (2)$$

using the radial wave functions $R_{nl}(r)$ and spherical harmonics $Y_{lm}(\Omega)$. The radial wave functions are obtained as numerical solutions of the corresponding eigenvalue equation for the radial field-free time-independent Schrödinger equation using the Numerov method on a logarithmic one-dimensional finite-space grid of size R_0 with boundary conditions $rR_{nl}(r)|_{r=0} = rR_{nl}(r)|_{r=R_0} = 0$ [13]. Due to the finite size of the box the number of bound states is limited, and the continuum is discretized. Hence, the energy eigenstates in this numerical basis set can be indexed by a principal quantum number n. In our calculations we consider the ground state of atomic hydrogen and therefore can restrict the basis set to states with $m = 0$ since $\Delta m = 0$ in interactions with linearly polarized fields.

In order to perform *ab initio* calculations for the polarization response to a linearly polarized external field we use the field-free representation of the dipole operator, given by

$$\hat{\mu} = \sum_{n,l,n',l'} |\psi_{nl0}\rangle\langle\psi_{nl0}|\hat{z}|\psi_{n'l'0}\rangle\langle\psi_{n'l'0}|, \tag{3}$$

and propagate the time-dependent Schödinger equation using the Crank-Nicholson method [13],

$$[\hat{H}_0 + E(t)\hat{\mu}]|\Psi(\mathbf{r},t)\rangle = i\frac{\partial}{\partial t}|\Psi(\mathbf{r},t)\rangle, \tag{4}$$

where $\hat{H}_0$ is the diagonal field-free Hamiltonian and $E(t)$ is of the form

$$E(t) = \sqrt{I}\sin^2\left(\frac{\pi t}{T_0}\right)\sin(\omega t), \tag{5}$$

with I as the intensity and T_0 and ω as the pulse duration and central frequency of the field, respectively. We then determine the low-order-harmonic spectra by calculating the Fourier transform of the dipole moment,

$$P(\omega) = \mathcal{FT}[\mu(t)](\omega), \tag{6}$$

where $\mu(t)$ is the time-dependent expectation value of $\hat{\mu}$ from Eq. (3).

In Fig. 1 we present an example for a low-order-harmonic spectrum generated at a driver wavelength of 1600 nm, a peak intensity of 5×10^{13} W/cm^2, and a pulse duration of 10 cycles. The results have been determined for a box size of $R_0 = 1000$ a.u., a time step of $\Delta t = 0.05$ a.u., and a maximum principle quantum number $n_{\max} = 2000$ as well as $l_{\max} = 70$. The convergence of the results with respect to the size of the radial box is shown by the relative error between the results for box sizes of $R_0 = 500$ and $R_0 = 1000$. Please note that the minima in the error correspond to the peaks in the harmonics.

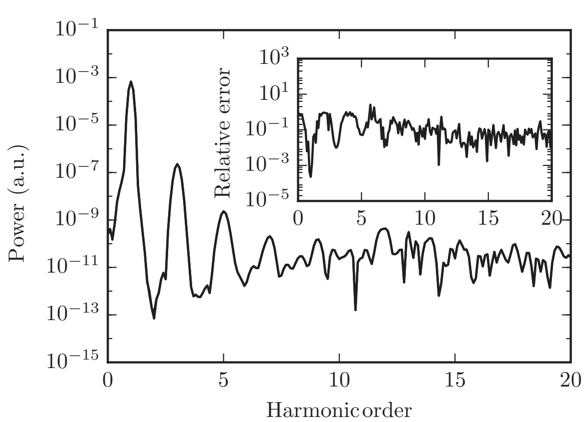

FIG. 1. Results of *ab initio* numerical calculations for a low-order-harmonic spectrum generated by a driver laser pulse at a central wavelength of 1600 nm, a peak intensity of 5×10^{13} W/cm^2, and a pulse length of 10 cycles. The inset shows the relative error between calculations using radial box sizes of $R_{\max} = 500$ and $R_{\max} = 1000$.

On the other hand, we use the eigenstates of the field-free energy basis to calculate the Nth coefficient of the perturbative power-series expansion of the ground-state wave function in the external field as given in Ref. [7],

$$|\psi^{(N)}(\omega_1,\ldots,\omega_N)\rangle = \sum_{j_N\neq j_0}\cdots\sum_{j_1\neq j_0}\left[\prod_{i=1}^{N}\frac{\langle\psi_{j_i}|\hat{\mu}E(\omega_i)e^{-i\omega_i t}|\psi_{j_{i-1}}\rangle}{\omega_{j_i}-\omega_{j_0}-\sum_{k=1}^{i}\omega_k}\right]|\psi_{j_N}\rangle, \tag{7}$$

where ω_{j_0} is the ground-state energy, ω_k and ω_i are the participating frequencies of the electric field, j_i denotes the state in the numerical basis set, and $\hat{\mu}$ is given by Eq. (3). The lifetimes of the excited states are neglected since all calculations performed in this study are far from resonance. The Nth-order term in the expansion of the single atom polarization in an overall $n\omega$ process can then be written

$$\langle\mathbf{P}^{(N)}(n\omega)\rangle = \mathcal{P}\sum_{j'=0}^{N}\langle\psi^{(j')}|\hat{\mu}|\psi^{(N-j')}\rangle, \tag{8}$$

where $n = 1,3,5,\ldots,$ $\sum_j \omega_j = n\omega$, and $\omega_j = \pm\omega$. $\mathcal{P}$ refers to the average of all permutations of the frequencies. The symmetry of the electric field with respect to positive and negative frequency components allows us to rewrite Eq. (8) as [7]

$$\langle\mathbf{P}^{(N)}(n\omega)\rangle = \epsilon_0\chi_{n\omega}^{(N)}\prod_{i=1}^{N}\mathbf{E}(\omega_i), \tag{9}$$

with $\chi_{n\omega}^{(N)}$ as the Nth-order term of the susceptibility at frequency $n\omega$ due to contributing electric fields at frequencies ω_i.

As for the *ab initio* calculations, we performed test calculations to ensure that the results of our calculations for the terms in the perturbation expansion of the susceptibility for low-order-harmonic generation are converged with respect to the size of the box R_0 and the size of the basis set $n_{\max}$. We note that the maximum angular momentum is determined by

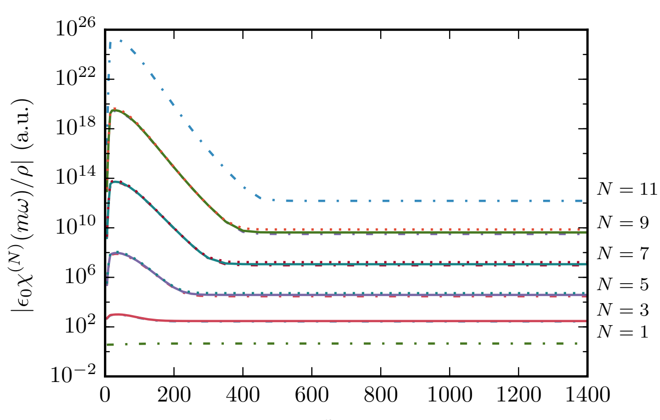

FIG. 2. (Color online) Results for perturbative power-series coefficients $\chi_{n\omega}^{(N)}$ for $n = 1$ (dashed-dotted lines), $n = 3$ (solid lines), and $n = 5$ (dotted lines) as a function of $n_{\max}$.

$l_{max} = (N+1)/2$, where N is the order of the coefficient calculated. In general, we have found that a box size of $R_0 = 500$ is sufficient for the present purpose. In Fig. 2 we show results for $\chi_{n\omega}^{(N)}$ for $n = 1$ (dashed-dotted lines), $n = 3$ (solid lines), and $n = 5$ (dotted lines) as a function of n_{max} for $R_0 = 500$ at a laser wavelength of 1600 nm. In general, we observe that the contributions from the bound states are positive, reflected in the increase in the susceptibilities for low n_{max}, and those from the continuum states are negative, corresponding to the decrease in the susceptibilities for higher n_{max}.

III. INTENSITY DEPENDENCE OF LOW-ORDER-HARMONIC GENERATION

We have applied both approaches to investigate the intensity dependence of low-order harmonics and the transition from perturbative to nonperturbative interaction. In Fig. 3 we present the integrated power of the (a) first, (b) third, and (c) fifth harmonics as a function of peak laser intensity at a central wavelength of 1600 nm and a pulse length of 10 cycles as obtained from our *ab initio* calculations. For these results we have calculated the harmonic spectrum and integrated the signal for the power of the nth harmonic over the energy range $[(n-1)\omega,(n+1)\omega]$. We compare the results of our numerical calculations with the power law I^n, which is expected for a perturbative n-photon process. The predictions from the power law were matched to the numerical results at low intensities. The inset in each of the panels shows the relative error between the *ab initio* results and the power-law predictions with respect to the *ab initio* results.

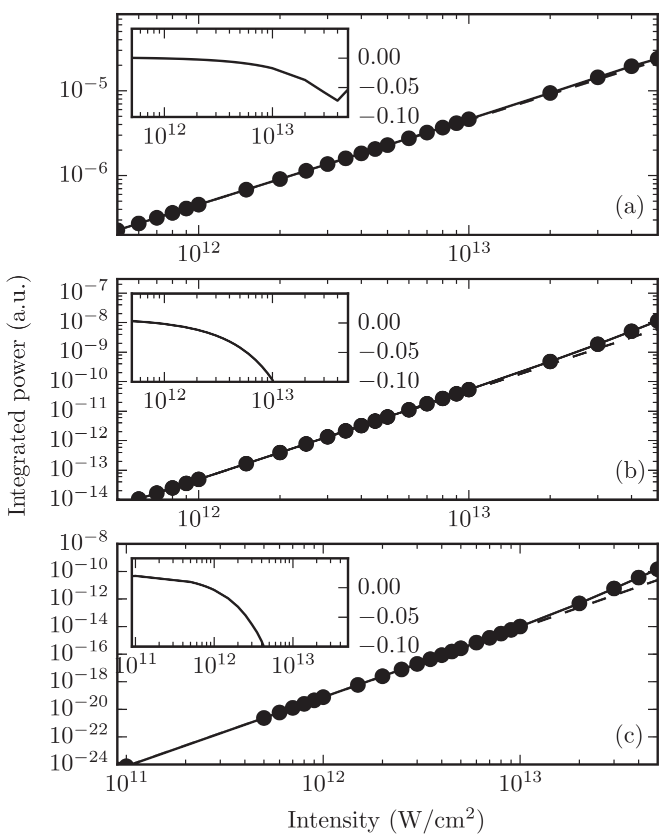

FIG. 3. Results of *ab initio* calculations for the integrated harmonic power (solid circles with solid lines) for the (a) first, (b) third, and (c) fifth harmonics as a function of the peak laser intensity of a laser pulse of 10 cycles at a wavelength of 1600 nm. The numerical results are compared to a perturbative I^n power-law fit, which is matched to the *ab initio* results at the lowest intensity. The insets show the relative error between *ab initio* results and power-law predictions with respect to the *ab initio* results.

The results show that in the intensity regime between 10^{12} and a few times 10^{13} W/cm^2 the *ab initio* results start to deviate from the respective power law. This is an indication of the transition from a perturbative to a nonperturbative electron-field interaction. These results are in agreement with the onset of other nonperturbative phenomena, e.g., above threshold ionization [14] and high-order-harmonic generation [15,16], in the same intensity regime.

Based on the *ab initio* results, we expect that the perturbative power-series expansion of the susceptibility corresponding to the process of low-order-harmonic generation should break down in this intensity range as well. In order to test this

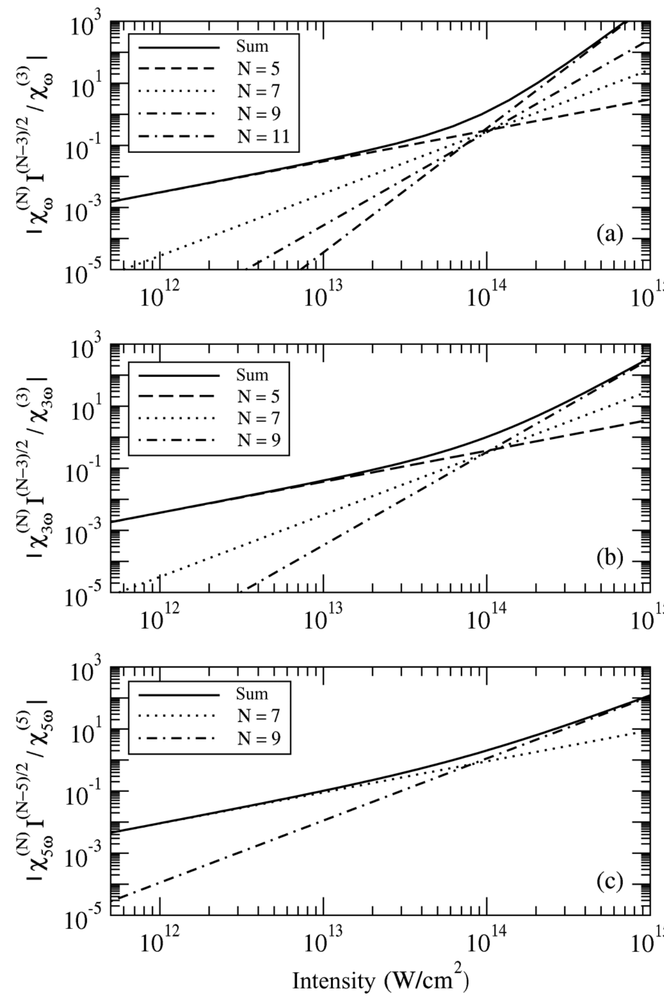

FIG. 4. Results for the ratio of higher-order terms to the lowest-order nonlinear term in the perturbative series expansion for (a) χ_ω, (b) $\chi_{3\omega}$, and (c) $\chi_{5\omega}$. Also shown is the ratio of the sum of all higher-order terms calculated with respect to the lowest-order term (solid lines).

expectation, we have calculated the first few terms in the expansion for χ_{ω}, $\chi_{3\omega}$, and $\chi_{5\omega}$ at 1600 nm. We study the relative contribution of higher-order terms in the expansion by presenting their ratios with respect to the lowest-order nonlinear term in Fig. 4. Also shown is the ratio of the sum of all higher-order terms calculated to the lowest-order term. From the results we observe the same behavior as previously reported for the electrical susceptibility χ_{ω} at a shorter wavelength [12], namely, each higher-order term is much smaller than the lowest-order term, indicating the convergence of the corresponding power-series expansion, at the lowest intensities studied. On the other hand, the breakdown of the series at the highest intensities is obvious as well since the contributions of the higher-order terms exceed that of the lowest-order term. We further note that in each case the sum of the calculated higher-order terms reaches about 10% of the lowest-order term for intensities in the range of 1×10^{13} to 2×10^{13} W/cm^2. Therefore, this limit can be considered as an indication for the breakdown of a perturbative series expansion in strong-field processes [17].

To summarize, our results for low-order-harmonic generation from both *ab initio* as well as perturbative calculations show the same onset of a transition from perturbative to nonperturbative interaction between the atom and the field as the previously reported results for the electrical susceptibility χ_{ω} at a shorter wavelength [12]. It is therefore not surprising that previous studies on low-order-harmonic yields [18–22] did not help in resolving the question about the significance of higher-order Kerr effects in the filamentation of short higher-power laser pulses in gaseous media. In contrast, we conclude that any deviations from the predictions of the lowest-order perturbation theory for the polarization (and other observables) should be interpreted as a signature for the nonperturbative character of the electron-field interaction. In particular, our results also show that a quantitative analysis of strong-field below-threshold harmonic generation requires a nonperturbative theoretical approach as, e.g., introduced in Ref. [2].

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