High harmonic generation in hydrogen and helium atoms subject to one– and two–color laser pulses

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Abstract. We present simulations of harmonic generation in hydrogen and helium solving, for hydrogen, the time–dependent Schrödinger equation and for helium the time–dependent Hartree–Fock and the time–dependent Kohn–Sham equations, respectively. The calculations are performed for a variety of laser wavelengths and intensities as well as for one–color and two–color pulses. Optimum conditions for the generation of intense high–order harmonics are discussed.

1. Introduction

Since the discovery of harmonic generation in rare gas atoms exposed to strong laser pulses [1, 2, 3] the production of high–order harmonics has become an important subject of experimental [4, 5] and theoretical [6, 7, 8] work in the field of intense laser–atom physics. One of the main reasons for this activity lies in the potential applications of this process as a route towards the generation of new coherent VUV or soft X–ray pulsed sources. Recently the first experiments using high harmonic radiation like time–resolved spectroscopy of helium [9] and pump–probe–experiments on surfaces [10] have been reported. The determination of optimum conditions for high–order harmonic generation has thus become an important experimental and theoretical challenge. The aim of our work is to study the influence of the laser frequency and the field strength on the intensities of the resulting harmonics in the plateau region by solving the time–dependent Schrödinger equation for the hydrogen atom in a strong laser field. Varying the two laser parameters either independently or simultaneously in a controlled way will provide us with first indications for the optimum choice of the laser parameters. As a second method to increase the efficiency of harmonic generation we present simulations for two–color laser pulses. These pulses consist of two different laser frequencies, usually a fundamental frequency and a low–order harmonic. The two-color calculations are performed for the helium atom.

2. Numerical procedure

To study the process of high harmonic generation in hydrogen we solve the time–dependent Schrödinger equation (atomic units are used throughout)

$$i\frac{\partial}{\partial t}\psi(\mathbf{r},t) = \left(-\frac{\nabla^2}{2} - \frac{1}{r} + E_0 f(t) z \sin(\omega_0 t)\right)\psi(\mathbf{r},t) \tag{1}$$

for an electron interacting with the nucleus and with a laser field, linearly polarized in z–direction, with a peak strength E_0 and frequency ω_0. As usual we make use of the dipole approximation written in the length form. The envelope function $f(t)$ has the form of a linear ramp over the first three laser cycles and is then held constant for the following 15 cycles.

We solve this equation in cylindrical coordinates (ρ, z, φ) with a finite–difference scheme very similar to Kulander [11] using a finite non–uniform grid as introduced by Pindzola *et al.* [12]. The spatial extent of the ρ–z grid is about 20 a.u. $\times$ 60 a.u. Due to the linear polarization of the laser field the angular part of the wave function is conserved. As initial state for our calculation we use the numerical ground state of hydrogen obtained by direct diagonalization of the Hamiltonian on the grid. This procedure yields a ground-state energy of -0.5 Hartrees in perfect agreement with the exact value.

We simulate ionization by an absorbing grid boundary [11] so that the norm of the wave function

$$N(t) = \int_{\text{grid}} |\psi(\mathbf{r},t)|^2\, d^3r \tag{2}$$

taken over the finite volume of the grid decreases with time. The resulting decay rates can then be interpreted as ionization rates.

To obtain the harmonic spectrum, we calculate the induced dipole moment

$$d(t) = \int z\, n(\mathbf{r},t)\, d^3r \tag{3}$$

from the electron density $n(\mathbf{r},t) = |\psi(\mathbf{r},t)|^2$ which is then Fourier transformed over the last 5 cycles of the constant–intensity interval. It can be shown [13] that this method gives results which are insensitive to the linear ramping of the laser intensity during the first three cycles of the simulation. The absolute square of the resulting Fourier transform, $|d(\omega)|^2$, has been shown [14] to be proportional to the experimentally observed harmonic distribution to within a very good approximation.

Harmonic generation in helium will be treated within the time–dependent Hartree–Fock approximation

$$i\frac{\partial}{\partial t}\psi(\mathbf{r},t) = \left(-\frac{\nabla^2}{2} - \frac{2}{r} + \int \frac{|\psi(\mathbf{r}',t)|^2}{|\mathbf{r}-\mathbf{r}'|} d^3r + E_0 f(t) z \sin(\omega_0 t)\right)\psi(\mathbf{r},t). \tag{4}$$

Here $\psi(\mathbf{r},t)$ denotes a (doubly occupied) spin orbital.

To solve Eq. (4) we use the same numerical methods as described for the hydrogen atom. However, the calculation of the initial helium ground state yields an energy eigenvalue of -0.955 Hartrees, which is 3.9 % off the exact Hartree-Fock value of -0.918 Hartrees. This error is due to the relatively coarse grid spacings in the vicinity of the nucleus which is inevitable to keep the numerical effort tractable during the time propagation.

To investigate the influence of electronic correlations in helium beyond the time–dependent Hartree–Fock approximation we also solve the time–dependent Kohn–Sham equation:

$$i\frac{\partial}{\partial t}\psi(\mathbf{r},t) = \left(-\frac{\nabla^2}{2} - \frac{2}{r} + \int \frac{|\psi(\mathbf{r}',t)|^2}{|\mathbf{r}-\mathbf{r}'|}d^3r + V_c(\mathbf{r},t) + E_0 f(t) z \sin(\omega_0 t)\right)\psi(\mathbf{r},t). \quad (5)$$

This equation arises from the optimized–effective–potential version [15] of time-dependent density functional theory [16, 17]. Heavier atoms have also been treated with this method [18, 19, 20]. $V_c(\mathbf{r},t)$ in Eq. (5) is a correlation potential for which we employ the local–density approximation in its parametrization by Vosko, Wilk and Nusair [21].

An approximation of the expected cutoff in the harmonic spectrum is given by the well known $I_0 + 3.2U_p$ rule [22, 23] where I_0 denotes the atomic ionization potential and $U_p = E_0^2/4\omega_0^2$ the ponderomotive shift. Despite the simplicity of the semiclassical model on which this rule of thumb is based its predictions are usually well confirmed by fully quantum mechanical calculations. Therefore this rule will be used as a guideline in the choice of laser parameters for an efficient generation of high harmonics.

3. Results

The first set of laser parameters considered are a wavelength of $\lambda = 1064$ nm and an intensity of $I = 2.0 \cdot 10^{13}$ W/cm^2. The resulting harmonic spectrum is given in Fig. 1. It agrees very well the result of Krause, Schafer and Kulander [24] for the above laser parameters. After a rapid decrease over the first few harmonics the calculated intensities form a plateau which extends approximately until the 17th harmonic. Using the $I_0+3.2U_p$ rule we find a value of 0.75 a.u. for the cutoff frequency in good agreement with the full quantum mechanical calculation.

In the following we investigate the variation of the harmonic spectrum if the laser parameters E_0 and ω_0 are changed. First we vary only the wavelength λ keeping the intensity $I = 2.0 \cdot 10^{13}$ W/cm^2 fixed. The semiclassical model then predicts a shift of the cutoff energy towards the ionization potential of 0.5 a.u. in the hydrogen atom for decreasing wavelengths. We calculate the harmonic spectra for laser frequencies ω_0 which are 1.5, 2.0, 2.5 and 3.0 times the frequency 0.04284 a.u. corresponding to the wavelength $\lambda = 1064$ nm. The results are presented in Fig. 2. Comparing the different spectra one finds the following general behavior: The extent of the plateau decreases with increasing wavelength as expected from the semiclassical model. At the same time the intensities of the harmonics in the plateau regions increase by several orders of magnitude.

If the objective is the generation of intense *and* high harmonics one has to change both laser parameters simultaneously. This is done by multiplying both the field strength $E_0 = 0.02384$ a.u. and the laser frequency $\omega_0 = 0.04284$ a.u. of our first example by the *same* factor. In this way, the ponderomotive potential $U_p = E_0^2/4\omega_0^2$ remains unchanged so that we expect the same cutoff frequency of 0.75 a.u. for all multiplicative factors. In our simulations, factors between 1.0 and 3.0 have been used. The resulting spectra are displayed in Fig. 3. For all parameter sets, we find that the plateau extends roughly until

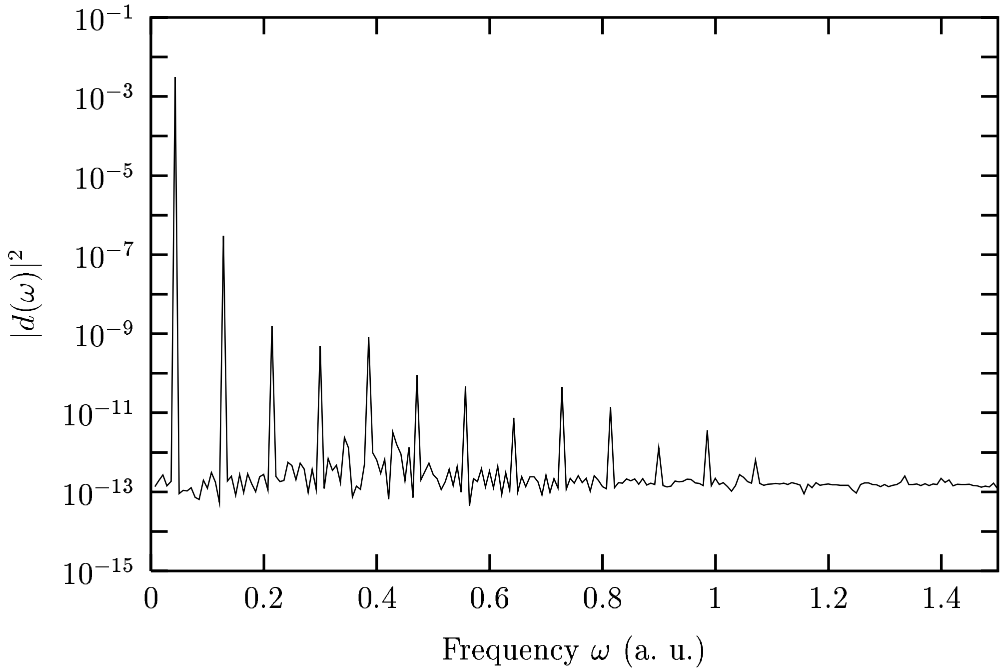

Figure 1. Harmonic spectrum of hydrogen for $\lambda = 1064$ nm and $I = 2.0 \cdot 10^{13}$ W/cm^2. The corresponding laser frequency has a value of 0.04284 in atomic units.

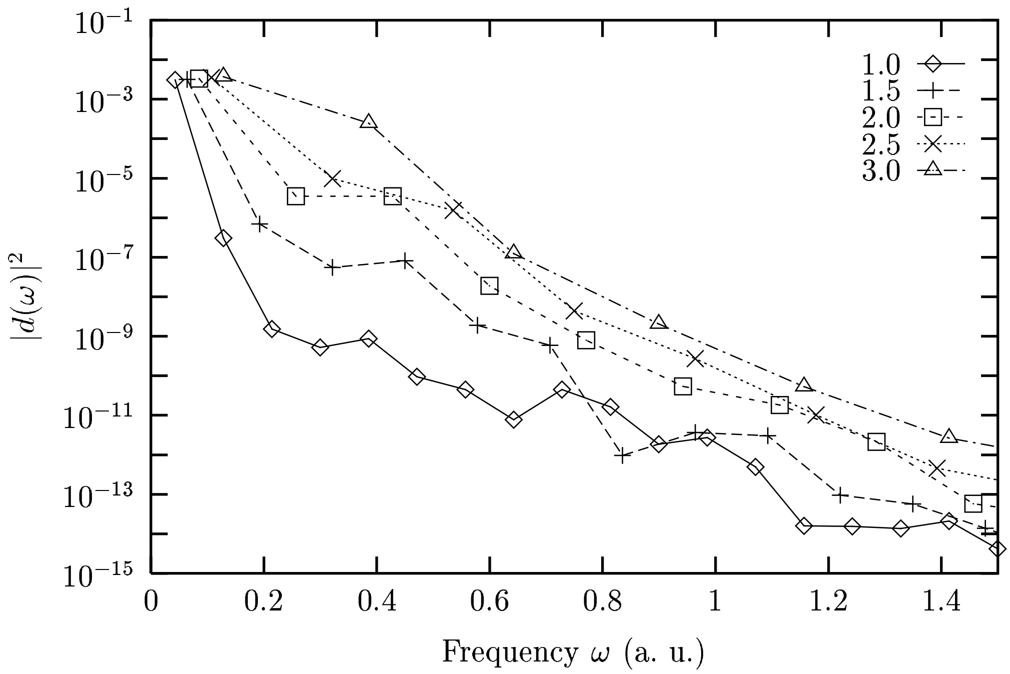

Figure 2. Harmonic spectra of hydrogen for various laser frequencies but fixed intensity of $I = 2.0 \cdot 10^{13}$ W/cm^2 (see text for details).

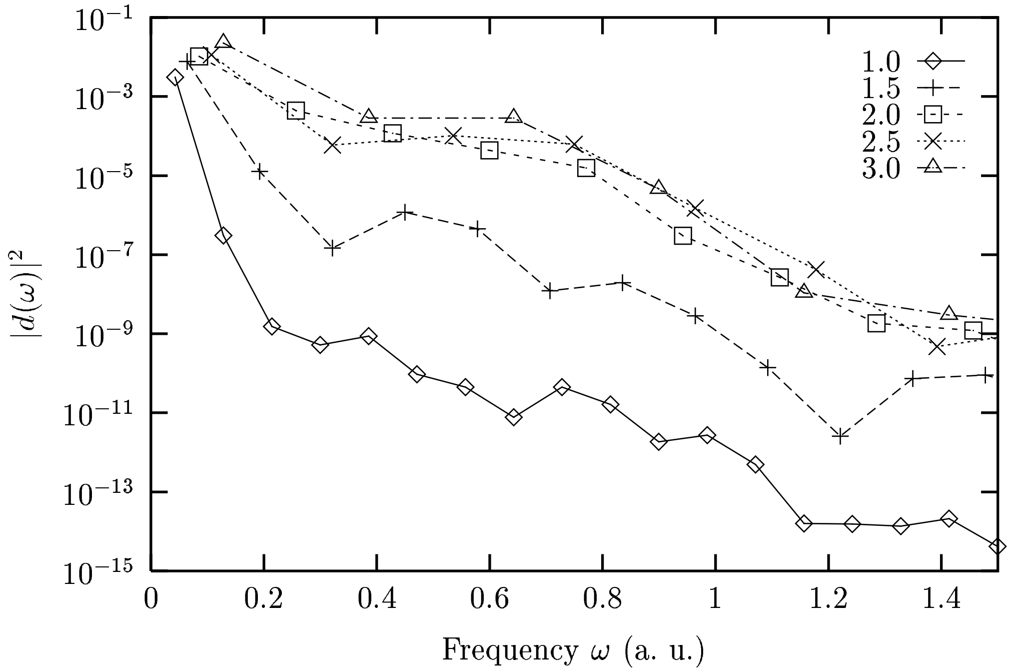

Figure 3. Harmonic spectra of hydrogen for various laser frequencies and intensities yielding the same ponderomotive potential (see text for details).

the semiclassical cutoff. More interesting is the observation that the intensities of the harmonics generated in the plateau regions increase when the field strength and the laser frequency ω_0 are raised by the same factor. However, above a factor of 2.0, a saturation in the intensities of the produced harmonics occurs. One possible cause for this effect is that the process of ionization is becoming more and more important: While, for a factor of 1.0, the norm of the wave function taken over the finite grid is 99% at the end of the simulation, only 25% of the norm is left within the finite grid for the factor 3.0.

Finally we investigate how the harmonic spectrum changes if only the intensity I is raised. The wavelength is held constant at a value of $\lambda = 1064$ nm. From the semiclassical cutoff rule we expect that the extent of the plateau will expand with increasing intensity. Fig. 4 shows harmonic spectra for intensities between $I = 2.0 \cdot 10^{13}$ W/cm^2 and $I = 3.2 \cdot 10^{14}$ W/cm^2 . In the intensity range below $I = 8.0 \cdot 10^{13}$ W/cm^2 the efficiency of harmonic generation is dramatically increased since the plateau regions are raised and expanded simultaneously with growing intensity. Above an intensity of $I = 1.0 \cdot 10^{14}$ W/cm^2 on the other hand the semiclassical cutoff frequencies are no longer reproduced by the full quantum mechanical calculation. Moreover, the generation of high harmonics gets more and more suppressed for intensities higher than $I = 2.5 \cdot 10^{14}$ W/cm^2. Both effects are most probably due to the competing process of ionization which becomes relevant at intensities above $I = 2.5 \cdot 10^{14}$ W/cm^2.

Another approach to increase the efficiency of high harmonic generation is the use of two–color laser fields. Evidence for this has been found in experiments [25, 26] as well as in numerical simulations for the hydrogen atom [27, 28, 29]. Here the laser fields employed are of the general form

$$E(t) = f(t)[E_0 \sin(\omega_0 t) + E_1 \sin(\omega_1 t + \delta)]. \tag{6}$$

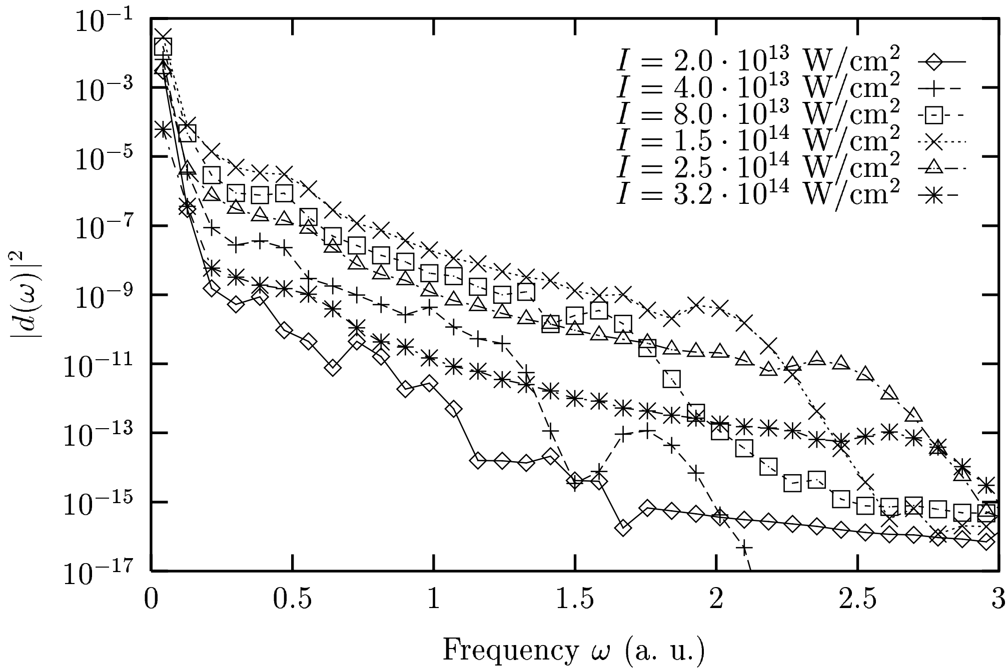

Figure 4. Harmonic spectra of hydrogen at a laser wavelength of $\lambda = 1064$ nm for various laser intensities.

In most cases E_0 and E_1 are of the same strength and ω_1 is an integer multiple of ω_0. A possible phase shift between the two fields is taken into account by the constant δ.

In order to investigate how the harmonic spectrum is influenced by the choice of the second laser frequency we have studied the harmonic generation in helium for different values of ω_1. The calculations were performed for $E_0 = E_1 = 0.010$ a.u. resulting in a total intensity of $I = 7.0 \cdot 10^{14}$ W/cm^2. The laser frequency ω_1 has been chosen to be the second and third harmonic of $\omega_0 = 0.0740$ a.u. which corresponds to a wavelength of $\lambda = 616$ nm. The phase difference δ has been set equal to zero in both cases. Fig. 5 shows the resulting harmonic spectra together with the results of a one–color calculation with the same total intensity $I = 7.0 \cdot 10^{14}$ W/cm^2 and the fundamental frequency $\omega_0 = 0.0740$ a.u. alone. The generated harmonics found at even multiples of ω_0 in the case $\omega_1 = 2\omega_0$ are due to nonlinear mixing processes of the two fields [26]. Most of the harmonics in the plateau region produced by the two–color field for $\omega_1 = 2\omega_0$ are one to two orders of magnitude more intense than those resulting from the one–color calculation.

In the case $\omega_1 = 3\omega_0$ an even stronger enhancement in the generation of high harmonics is found. This enhancement however approximately lasts only until the 29th harmonic. The results of this simulation therefore resemble the effects found in hydrogen (see Fig. 2) where the plateau region was raised and shortened with increasing laser frequency.

To investigate the contributions of the two laser frequencies to the process of harmonic generation we now vary the intensities I_0 and I_1 of the fundamental frequency ω_0 and the third harmonic $\omega_1 = 3\omega_0$ respectively. The total intensity $I = I_0 + I_1$ is held fixed at a value of $7.0 \cdot 10^{14}$ W/cm^2 to allow for a direct comparison of the resulting

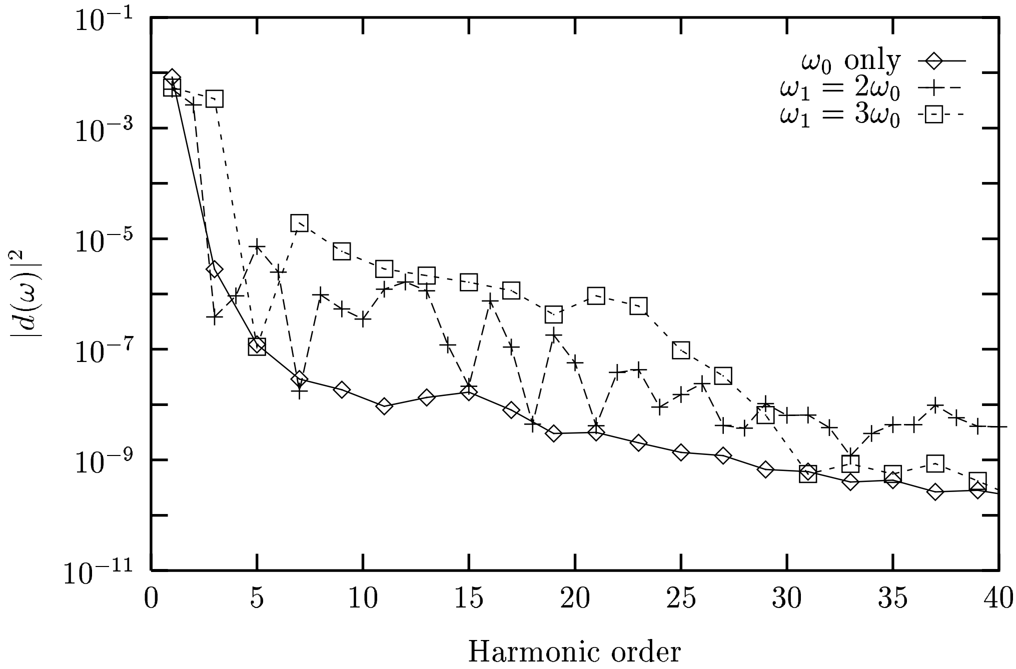

Figure 5. Harmonic spectra of helium resulting from a one–color calculation and from two two–color calculations. One of the latter includes the second and the other includes the third harmonic in addition to the fundamental frequency. The laser parameters used are $\lambda = 616$ nm and $I = 7.0 \cdot 10^{14}$ W/cm^2.

spectra. The intensities I_0 and I_1 are therefore chosen as

$$I_0 = \frac{1}{q+1} I, \tag{7}$$

$$I_1 = \frac{q}{q+1} I \tag{8}$$

where q denotes the ratio of the intensities. The harmonic spectra for different values of q and a phase difference of $\delta = \pi/2$ are given in Fig. 6. One finds that adding only a small amount (q=0.01) of the third harmonic yields a considerable increase of the intensities of the generated harmonics compared to the one–color simulation (q=0). The best efficiency of harmonic generation is already reached for a ratio of q=1. q–values greater than unity do not lead to a significant enhancement.

All of the above calculations for helium were done within the time–dependent Hartree–Fock approach. To investigate the influence of electronic correlations on the generation of high harmonics we finally compare the time–dependent Hartree–Fock approach with the results of a time–dependent density functional calculation. The latter is based on Eq. (5) where electronic correlations are taken into account through a local correlation potential. Fig. 7 compares the two approaches for the harmonic spectrum of helium, calculated with the laser parameters $\lambda = 616$ nm and $I = 7.0 \cdot 10^{14}$ W/cm^2. The intensities of the produced harmonics are found to be generally reduced in the calculation with correlation, typically by a factor of 2 – 3. Although this is not exactly a small deviation, the overall structure of the harmonic spectrum is changed very little by electronic correlations.

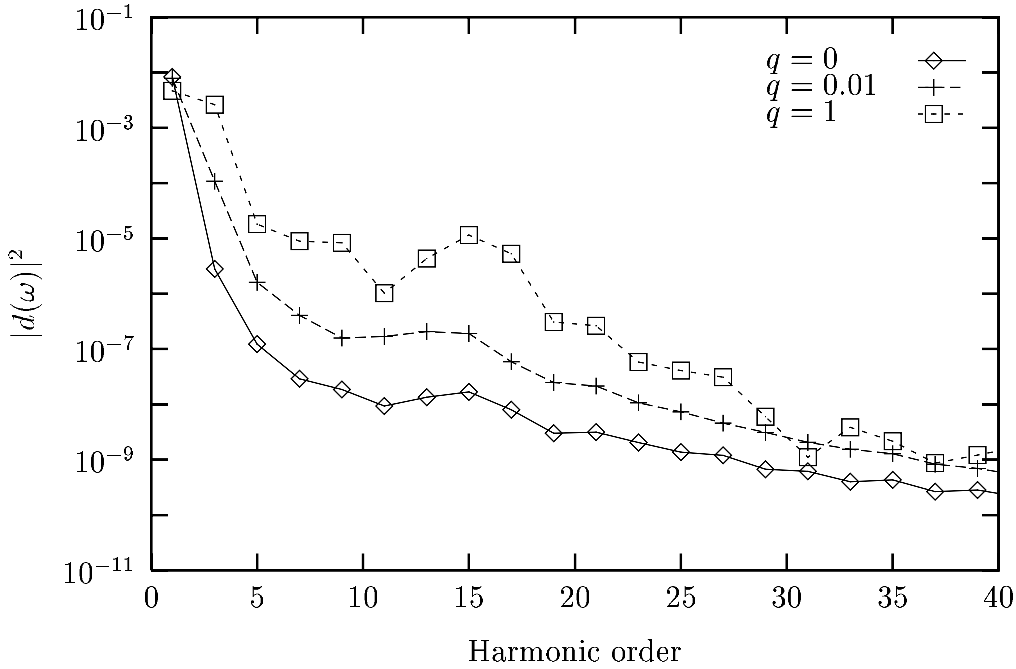

Figure 6. Harmonic spectra of helium resulting from two-color calculations with different intensity ratios q of the third harmonic to the fundamental frequency.

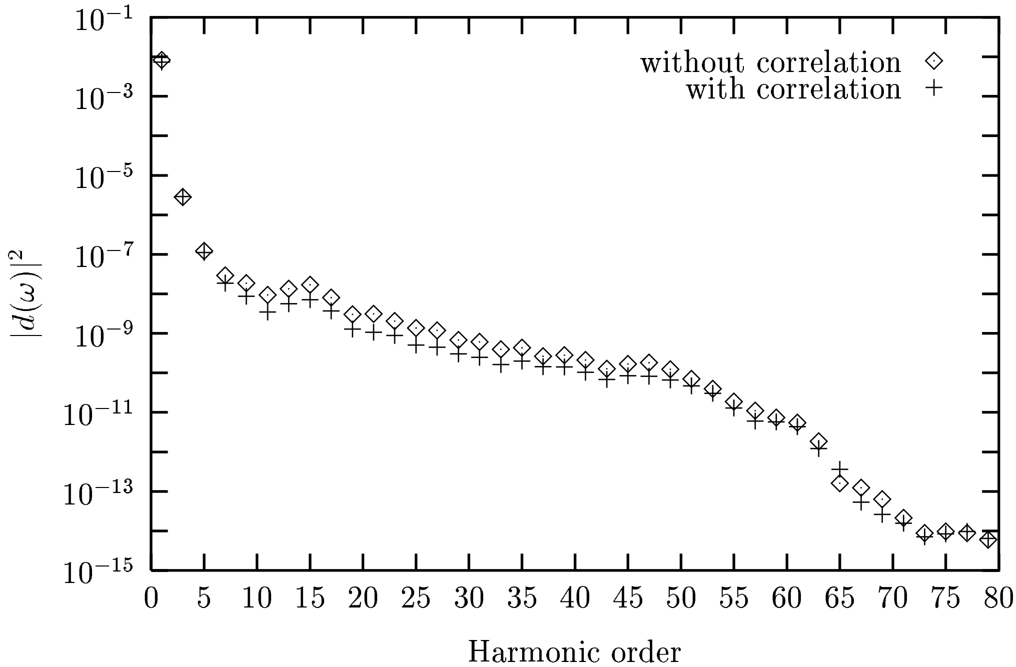

Figure 7. Harmonic spectra of helium resulting from the Hartree–Fock equation (without correlation) and the Kohn–Sham equation (with correlation). The laser parameters are $\lambda = 616$ nm and $I = 7.0 \cdot 10^{14}$ W/cm^2.

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