Intense dependent spectral features in high harmonic generation

Khuong Ba Dinh, a Peter Hannaford, and Lap Van Dao
ARC Centre of Excellence for Coherent X-Ray Science and Centre for Atom Optics and Ultrafast Spectroscopy, Swinburne University of Technology, Melbourne, Victoria 3122, Australia

(Received 12 December 2012; accepted 23 January 2013; published online 8 February 2013)

We study the influence of the driving laser intensity on the spectral features of high harmonic generation in a semi-infinite gas cell. The effects of the harmonic dipole phase and the dispersion phase mismatches induced by the ionized medium during the harmonic generation process are revealed and the interplay between the macroscopic response and the single-atom response is discussed. We consider the conditions for generation of a narrow bandwidth, bright, and highly coherent high harmonic source. © 2013 American Institute of Physics. [http://dx.doi.org/10.1063/1.4790708]

I. INTRODUCTION

High order harmonic generation (HHG) produced by the interaction of an intense laser pulse and a gas medium can provide a table-top coherent radiation source in the short wavelength range from the ultraviolet to the soft x-ray region.1,2 The generation of highly spatial and temporal coherent sources is an important requirement in certain time-resolved spectroscopy applications3 and especially in coherent diffusive imaging.4

A physical understanding of the HHG process is provided by the three-step model in which the interaction between the laser field and the atoms is separated into ionization, acceleration, and recombination stages.1,5 A full quantum mechanical theory which recovers the semi-classical model and considers quantum effects such as tunneling, diffusion, and interference has been developed within the strong field approximation (SFA) model to describe aspects of the HHG process more precisely.6 In the SFA theory, the HHG from a single atom can be obtained by calculating the dipole acceleration of a returning electron which has gained momentum in the presence of the oscillating laser electric field. However, in general, an understanding of the HHG process based only on single-atom dynamics is not sufficient to explain the experimental data completely. In order to achieve good agreement between theory and experiment, one must also take into account the effects of propagation and phase mismatch between the harmonic field and the fundamental field in the macroscopic medium.7,8 A high laser field produces a large harmonic dipole dq(t) that may be obtained from a quantum mechanical theory.6,8,9 The propagation of the fundamental and harmonic fields in the medium depends strongly on the laser intensity because of induced non-linear refraction or variation of the atomic phase. When the laser intensity is varied, the total output harmonic intensity can be increased or decreased because the phase mismatch between the fundamental laser field and the harmonic field is changed.7,8

The propagation of the fundamental and harmonic fields in a gas medium leads to a phase modulation which causes a modulation of the harmonic spectrum.10 Through modification of the laser chirp condition, a coherent control process of high harmonic generation for production of sharp and strong harmonics has been demonstrated11,12 A theoretical study of the spectral structure of the high harmonic emission from atomic gases was conducted by Kan et al.13 who showed that the splitting and blue shift of the harmonic spectrum is due to the intensity dependent or time dependent phase shift of the dipole acceleration responsible for the harmonic generation. However, it is difficult to study the splitting attributed to this mechanism in the experimental measurements especially when a long laser pulse is applied. Recently, the contribution of interference between the quantum paths, e.g., between the long path and short path, to the harmonic emission has been theoretically and experimentally investigated.14,15 An electron traversing the long path following ionization returns to the parent ion in a time close to one period of an optical cycle while an electron traversing the short path has a return time less than half an optical cycle.16 The fast regular oscillations of the intensity dependent harmonic signal and the interference fringes in the harmonic spectrum due to quantum path interferences have been studied theoretically by Zait et al.14 In order to observe the interference between these two quantum paths, spatial filtering has been used to minimize the spatial averaging that would otherwise blur out the interference in the macroscopic response.14,15

In this paper, we investigate the influence of the driving laser intensity on the spectral features of the harmonics. In our studies, the macroscopic phase matching which plays a significant role in the harmonic generation using a long gas cell and the interplay between the single-atom response and the macroscopic response is analyzed. With spatial selection, the contribution of the different electron trajectories to the HHG spectrum is considered. In addition, based on these studies, we can determine the conditions for the production of narrow bandwidth and strong harmonics with high coherence.

II. THEORETICAL BACKGROUND

When atoms or molecules interact with an intense laser field, a high order nonlinear polarization is induced which

8aAuthor to whom correspondence should be addressed. Electronic mail: kdlqh@swin.edu.au. Tel.: +61 3 9214 5680. Fax: +61 3 9214 5160.
acts as a source for the generation of a high order harmonic field. The qth harmonic field at the exit of the medium with an interaction length $L_{\text{med}}$ is the integral of all the atomic dipoles along the medium and is given by:

$$E_q(L_{\text{med}}) = \frac{\imath q \omega}{\omega_0 c} \int \frac{d_q(I, z) \exp \left( z - \frac{L_{\text{med}}}{2L_{\text{abs}}} \right)}{d_I} \times \exp[i \Delta k_q(I, z) z] \, dz \, dI,$$

where $\Delta k_q(I, z)$ is the phase mismatch between the harmonic radiation and the laser induced polarization, $d_q(I, z)$ is the amplitude of the high harmonic dipole, $L_{\text{abs}}$ is the absorption length, $c$ and $\omega_0$ are the speed of light and vacuum permittivity, $\omega$ is the angular frequency of the laser pulse, and $I$ is the laser intensity. The integration is over the length of the interaction medium and the intensity envelope of the driving laser field. The amplitude of the high harmonic dipole $d_q$ is dependent on the intensity of the driving laser field and in the single-atom response model $d_q$ scales as $d_q \propto I^3$ in the intensity window of the driving pulse $I$ to $I + dI$.

The phase mismatch $\Delta k_q$ is the sum of four terms and can be expressed as:

$$\Delta k_q = \frac{2 \pi q}{\lambda} \rho \hat{n}(1 - \eta) - \eta N_{\text{atm}} r_e \lambda \left( q - \frac{1}{q} \right) + \text{(geometric term)} + \text{(atomic dipole phase)},$$

where $\lambda$ is the laser wavelength; $\rho$ and $\eta$ are the gas pressure and ionization fraction, respectively; $\hat{n} = n_{\text{laser}} - n_i$; $n_{\text{laser}}$ and $n_i$ are the refractive indices for the fundamental and the qth harmonic field; and $N_{\text{atm}}$ is the atomic number density at 1 atm. In this expression for the phase mismatch, the first (positive) term is related to the medium dispersion and the second (negative) term is due to the plasma dispersion. The geometric term is negative in a waveguide or self guide. For a focused Gaussian beam, the geometrical phase shift around the focal point is due to the Gouy phase shift. The sign of the atomic dipole phase term is not fixed since this phase varies with the intensity of the laser field. To a first approximation, this phase scales linearly with the laser intensity, $\varphi_q = -z_q I$, where $z_q$ is a coefficient related to the electron trajectories ($z_q \approx 1 - 5 \times 10^{-14}$ cm$^2$/W for the short trajectory and $20 - 25 \times 10^{-14}$ cm$^2$/W for the long trajectories). During the laser pulse, the intensity $I$ and therefore the atomic dipole phase vary with time. Moreover, the variation of the laser intensity with time modifies the neutral gas density and the plasma density leading to a change of the neutral gas dispersion and plasma dispersion. Consequently, according to Eq. (2), a variation of the fundamental laser intensity influences the harmonic intensity through a change of $\Delta k_q$ and the amplitude of the harmonic dipole, $d_q$.

The modulation of the intensity dependent dipole phase affects not only the total phase mismatch leading to a change of the harmonic intensity, which has been mentioned above, but also the spectrum of the HHG. Indeed, the variation in time of the fundamental laser intensity, $I(t)$, induces a change in the instantaneous frequency of the harmonic emission through a harmonic phase modulation process. The instantaneous harmonic frequency during the pulse is given by:

$$\omega_q(t) = q \omega + \frac{q_0}{\sigma} \frac{\partial I}{\partial t}.$$  

Here, $q \omega$ is the central harmonic frequency and the term $\frac{q_0}{\sigma} \frac{\partial I}{\partial t}$ gives the variation of the instantaneous frequency with time. From Eq. (3), the spectral broadening is given by:

$$\Delta \omega_q(t) = \frac{q_0}{\sigma} \frac{\partial I}{\partial t}.$$  

For a Gaussian intensity profile, during the laser pulse, the phase matching is transiently optimized at certain intensities $I_n$ at given times $t_n$ depending on the peak intensity, $I_0$, and the instantaneous harmonic frequency at the laser intensity $I_n$ can be calculated as a function of the scanning intensity, i.e., the peak intensity, $I_0$.

Based on Eq. (5), one can obtain an equation which describes the exponential shape of the maximum signal in the harmonic spectrum as a function of the laser intensity:

$$I_0(\omega_q) = I_0 \exp \left[ \frac{\sigma^2 (q \omega - \omega_q)^2}{4 \omega_q^2 I_0^2} \right].$$

III. EXPERIMENTAL RESULTS AND DISCUSSION

A 1 kHz multi-stage, multi-pass chirped pulse amplifier system which produces up to 10 mJ pulses (only <5 mJ are needed for this experiment) with a duration of 30 fs and centred at 805 nm is used for our experiments. The laser is focused into a semi-infinite argon filled gas cell by a 300 mm focal length lens. Using an aperture with diameter ranging from 5–10 mm placed in front of a focusing lens, the radius of the focused intensity distribution of the laser beam varies from approximately 50 to 100 μm. The aperture is also used to limit ionization of the gas by decreasing the applied intensity. When the laser pulse energy is set to 5 mJ or higher, the ionization rate is very high (>20%), the harmonic intensity is very low, and the spectrum and profile of the harmonic beam are very complicated. With consideration of the defocusing effect due to plasma dispersion, the ionization is ~10%–15% at a laser pulse energy of ~3.5 mJ and ~5% at a laser pulse energy of ~2 mJ. The gas pressure in the cell is kept at 70 Torr to minimize absorption effects. The laser focus is set close to the exit of the cell. Further details of the HHG setup and detection system have been described.
A. Spectral features of HHG at high laser intensity

When a large aperture diameter (10 mm) is used, the beam profile of the fundamental is selected from the central part for better homogeneity and the maximum laser pulse energy is measured to be 3.5 mJ. Using this pulse energy, a high ionization fraction (~10%) is produced which is much higher than the critical value (~5%). The harmonics can be generated in the observed spectral range at laser pulse energies from 0.2 mJ. Figure 1 shows the H21 intensity, which is the strongest harmonic, as a function of the laser pulse energy. A similar dependence is observed for other harmonic orders. The inset shows the harmonic spectrum at a laser pulse energy of 0.4 mJ at which a small bandwidth of harmonics can be obtained. It is worth noting that small bandwidth harmonics can be generated for laser pulse energies up to 1 mJ but the flux in rather low. At low laser pulse energy (<0.6 mJ), the harmonic intensity increases with increasing laser pulse energy because the phase-mismatch is small.

When the laser pulse energy is high, a large phase mismatch due to the ionization reduces the total harmonic intensity. When the laser pulse energy is larger than 2.3 mJ, the harmonic intensity saturates due to the defocusing effect resulting from the high ionization fraction.

We find that the maximum photon flux of H21, which is the strongest harmonic in the HHG spectrum, is \(~1.5 \times 10^{10}\) photon/(cm²s) (corresponding to \(~3.5 \times 10^{9}\) photon/s and a conversion efficiency \(~8.5 \times 10^{-9}\) when the laser pulse energy is \(~2.3\) mJ. This intensity is low because of a large phase mismatch due to the large plasma dispersion resulting from the very high free electron density in the medium. Moreover, the defocusing effect increases the diameter of the focus and therefore the laser intensity at the focus is not increased.

Figure 2 shows the measured harmonic spectrum of the H21 harmonic as a function of the laser pulse energy. A similar figure is found for the other harmonic orders. Figure 2(b) shows the harmonic spectrum when the on-axis region (around the propagation direction of the fundamental field) is selected. In this case, the contribution of the short trajectories is large and phase matching can be better controlled for harmonics in this region. Figure 2(a) is the harmonic spectrum for off-axis selection.

The change of the harmonic spectrum with the laser pulse energy for the off-axis region, which has a dominant contribution from the long trajectory with large coefficient, \(\alpha_q\), is stronger than that for the on-axis region. In Figure 2(a), spectral interference fringes are clearly visible. A first fringe, a second fringe, and a third fringe are observed at laser pulse energies of \(~0.6\) mJ, \(~0.9\) mJ, and \(~1.6\) mJ, respectively. The blue-shifting and splitting observed in the experiment may be associated with the harmonic phase modulation which is indicated by Eq. (5). To compare the experiment with theory, we perform a calculation of the H21 spectrum as a function of the laser pulse energy using Eq. (6). For simplification, we neglect the defocusing effect in this laser pulse energy range and then the laser intensity axis can be converted linearly from the laser pulse energy axis within an error of 10%. For different intensities \(I_{in}\), i.e., different fringe patterns, the coefficient, \(\alpha\), is adjusted to obtain the best agreement with the experimental harmonic spectral lines. The simulated results are shown by the black lines. In Figure 2(a), we find that the maximum coefficient is around \(\alpha_{q,\text{max}} \approx 11.5 \times 10^{-14}\) cm²/W, which is
consistent with a larger contribution from the long trajectory in the off-axis region.

Figure 2(b) shows the variation of the spectral line in the on-axis region where the short trajectory predominantly contributes to the harmonic radiation leading to a weaker blue-shifting compared to that in the off-axis region. For laser pulse energies greater than 0.6 mJ, the spectrum exhibits a blue-shifting resulting in the first fringe. We should observe a second fringe when the laser energy is \( \sim 0.9 \) mJ. However, because this shifting may be very small, a second fringe cannot be seen, and in this case, we just see the central component. In a similar way to the case of the off-axis selection, by performing a simulation and fitting to the experimental results, we find a maximum coefficient \( \chi_{q,\text{max}} \approx 7 \times 10^{-14} \text{cm}^2/\text{W} \), which is consistent with a larger contribution from the short trajectory. In Figure 2, for both off-axis and on-axis selections, as shown by the simulations, an exponential shape of the harmonic spectral distribution (Eq. (6)) should be visible. However, in the experiment, we cannot see the red wings clearly (dashed lines) and this is due to the significant plasma phase mismatch induced by the strong ionization fraction on the trailing edge of the pulse which distorts the harmonic emission on this edge. In addition, unlike the simulations, the experimental spectral lines do not shift anymore for laser pulse energies higher than 2.3 mJ, and this is a direct consequence of the near saturation of the laser intensity induced by the defocusing effect.

When the ionization fraction is high at high laser pulse energy, we are not able to produce a sharp and strong harmonic spectrum. In this case, with an increase of the laser pulse energy, the harmonic conversion efficiency is low because of significant plasma phase mismatch and a large contribution of different quantum pathways must be considered in the generation process leading to the fact that the harmonic spectrum exhibits strong shifting and splitting which can be seen in Figure 2.

**B. Spectral features of HHG with limitation of ionization fraction**

We apply an aperture for optimizing the harmonic generation process. All available harmonics with intense on-axis emission and narrow spectral bandwidth, which is typical for a dominant contribution from the short trajectory emission, are considered. The laser pulse energy for the optimal case is \( \sim 2.1 \) mJ corresponding to a reduced diameter (6 mm) of the aperture of the full beam. Under these conditions, the ionization rate should be lower than the critical value (\( \sim 5 \)% over the full range of available intensity when the laser pulse energy is varied continuously from a minimum value of 0.3 mJ, at which the harmonics start to be observed, up to a maximum value of 2.1 mJ by rotating the half-wave plate.

Figure 3 shows the selected H25 intensity as a function of the laser pulse energy. A similar dependence is observed for the other harmonic orders. The maximum photon flux of H25 is \( \sim 10^{12} \text{photon}/(\text{cm}^2\text{s}) \) (corresponding to \( \sim 2.5 \times 10^{11} \text{photon/s} \) and a conversion efficiency \( \sim 7.5 \times 10^{-7} \)).

For low laser pulse energies (below 1.2 mJ), the increase of the H25 intensity can be fitted well with the function

\[
I_q = a_0 \left(1 - a_1 \frac{\sin^2 \left(\frac{\Delta k_{q,\text{eff}}L_{\text{med}}}{2}\right)}{\left(\frac{\Delta k_{q,\text{eff}}L_{\text{med}}}{2}\right)^2}\right),
\]  

where

\[
\frac{\Delta k_{q,\text{eff}}L_{\text{med}}}{2} = a_2 + a_3 I_{\text{pulse}}.
\]  

\( I_{\text{pulse}} \) where \( I_{\text{pulse}} \) is the laser pulse energy. Indeed, for low laser pulse energies, the ionization fraction is very small. In this energy regime, there is only a minor influence of the ionization on the phase mismatch and the variation of the atomic dipole phase is also negligible. This means that macroscopic effects can be neglected (\( \Delta k \approx 0 \)), and a single-atom response is exhibited in this regime. Therefore, from Eq. (1), the harmonic intensity scales linearly with \( |E_q|^2 \) which is proportional to \( d_q^2 \), where \( d_q \approx 1 \) is the amplitude of the harmonic dipole.\(^1\) In addition, for low laser energies, the defocusing effect can be ignored. As a result, the harmonic intensity in this region can be fitted by the function \( I_{6\,\text{pulse}} \).

When the laser pulse energy is above 1.2 mJ, the ionization fraction becomes higher and, especially for the gas cell geometry, the increase of ionization causes strong plasma dispersion. The laser intensity dependence of the harmonic yield in this region becomes more complicated and the development of the harmonic intensity with laser pulse energy, i.e., the harmonic conversion efficiency, is slower because of the stronger influence of the phase mismatch. The considerable phase mismatch and the defocusing leads to near saturation of the harmonic yield at a laser pulse energy of \( \sim 1.9 \) mJ.

In the high pulse energy regime, the development of the harmonic intensity as a function of pulse energy can be fitted by the equation

\[
I_q = a_0 \left(1 - a_1 \frac{\sin^2 \left(\frac{\Delta k_{\text{q,eff}}L_{\text{med}}}{2}\right)}{\left(\frac{\Delta k_{\text{q,eff}}L_{\text{med}}}{2}\right)^2}\right),
\]  

\[
\frac{\Delta k_{\text{q,eff}}L_{\text{med}}}{2} = a_2 + a_3 I_{\text{pulse}}.
\]  

\( \Delta k_{\text{q,eff}} \) is an effective phase mismatch which is proposed in order to simplify our calculation. The dependence shown in Eq. (7) suggests that when the laser pulse energy is higher than 1.2 mJ, the phase mismatch plays a significant role in the variation of the harmonic yield and results in a slow rise of the harmonic conversion efficiency as indicated in Figure 3. Moreover, the amplitude of the harmonic dipole moment...
remains nearly unchanged in this region. Based on Eq. (8), the linear dependence of the phase mismatch on laser pulse energy (for $L_{\text{med}} \sim 4 \text{ mm}$, $\Delta k_{\text{eff}}$ varies linearly from 11 cm$^{-1}$ to 15.3 cm$^{-1}$ as the laser pulse energy increases from 1.2 mJ to 2.1 mJ) reflects the important contribution of the harmonic dipole phase term to the phase mismatch.

Figure 4 shows the spectrum of the four strongest harmonics (Figure 4(a)) and the selected harmonic H25 (Figure 4(b)) as a function of the laser pulse energy. The spectrum remains narrow at high intensity on-axis during scanning of the laser pulse energy from the minimum to the maximum value. This behavior indicates that the short trajectory contributes predominantly to the harmonic emission during the scanning of the laser pulse energy.

In order to determine the degree of spatial coherence, a Young’s double slit (YDS) experiment was performed. The YDS consists of two parallel slits with a slit spacing of 20 μm, a slit width of 4 μm, and a slit height of 100 μm. The YDS is illuminated by all available harmonics. The interference fringes are detected by a CCD. For more detailed measurements, a spectrometer is included and the YDS is mounted perpendicular to the slit of the spectrometer (Y direction). The interference fringes are directly detected in the Y-direction of the CCD, while different harmonic orders are observed along the X-direction. Figure 5 shows the high contrast of the interference fringes for H25 (blue line) and for the case when all available harmonics are used (red dotted line). The inset shows the CCD image from the spectrometer. The degree of coherence of the source is 0.97 which indicates the very high spatial coherence of the HHG source.

IV. CONCLUSIONS

We have experimentally studied the influence of the driving laser intensity on the spectral features of HHG. Through these studies, the influence of the harmonic dipole phase and the dispersion phase mismatches induced by a strongly ionized gas medium on the HHG spectral features was clearly revealed. Furthermore, the use of an aperture to confine the ionization fraction to less than a critical level at a high laser pulse energy for strong coherent buildup of the on-axis harmonic radiation is critical for generating a sharp and intense harmonic spectrum with high spatial coherence.