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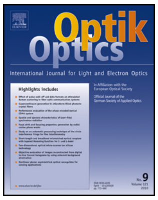

# Influence of absorption on phase-matched generation of coherent extreme ultraviolet radiation in a long interaction geometry

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## ABSTRACT

The pressure dependent high-order harmonic generation in a semi-infinite gas cell with two different absorbing gaseous media (argon and helium) is analyzed to reveal the influence of absorption on the radiation process in the extreme ultraviolet region. The absorption cross section of a particular wavelength in this region is measured. Moreover, in consideration of macroscopic response, the geometrical phase mismatch and the dipole phase mismatch which are independent to the pressure are clearly studied. A Young's double slit experiment is also performed to indicate a high spatial coherence of the harmonic radiation. This measurement shows that a narrow bandwidth, bright, and highly coherent high harmonic source can be generated in gas cell filled with absorbing gases which could be useful for coherent diffractive imaging.

## 1. Introduction

High harmonic generation (HHG) which is an extreme nonlinear optical process occurs when there is the interaction of an intense laser field with an atomic or molecular gas [1,2]. The high order harmonics which are emitted in a series of attosecond bursts with high spatial and temporal coherence are very interesting for time-resolved studies of atom and molecular [3] and coherent diffractive imaging [4]. The high harmonic generation process can be described by using a classical or quantum treatment through the time-dependent Schrödinger equation in a non-pertubative approximation [1,2,5,6]. A classical picture of HHG is usually referred to as the three-step model in which the interaction of a strong laser field leads to the ionization of the active electrons and then the acceleration before recombination with their parent ions to emit high-energy photons [5,6]. 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. Recently, the observation of nonlinear optical wave-mixing in HHG [7,8] also suggests the possibility to treat the physics in the XUV range with a perturbative nonlinear optics theory although perturbative and nonperturbative nonlinear optics seem conceptually very different. In pertubative theory, the response of the medium with the interaction of the ultrashort laser pulse can be expanded in many orders, expressed as linear and nonlinear terms and very high orders of response need to be taken into account [9].

In order to obtain a high intensity of high-order harmonic radiation the effects of propagation and phase mismatch between the harmonic field and the fundamental field in the macroscopic medium need to be considered. The phase mismatch consists of four contributions: the geometrical wave vector mismatch caused by focusing, the dispersion of the neutral atom and of the free electrons, and the atomic dipole phase [10,11]. Even if phase matching is achieved, the conversion efficiency is still limited by re-absorption of the harmonic emission in the generating gas medium [12–14]. Under the good phase matching condition, the coherent harmonic intensity increases with increasing gas pressure and/or interaction length. On the other hand, when a harmonic is passing through an absorptive medium the signal also decreases in proportion to the negative exponential of these factors. Thus, when the harmonic yield approaches the maximum value for a particular interaction length and gas pressure the harmonic signal has

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reached the re-absorption limit. Constant et al. [12] have performed an analysis of the time dependent factors that control the output photon flux including the atomic response, phase matching conditions and absorption of the medium and they found that the overall optimizing conditions in the absorbing medium are $L_{\rm med} > 3L_{\rm abs}$ and $L_{\rm c} > 5L_{\rm abs}$, where $L_{\rm med}$ is the interaction length, $L_{\rm c}$ is the coherence length, and $L_{\rm abs}$ is the absorption length.

In this paper, we experimentally study the influence of the absorption on the high-order harmonic intensity in a semi-infinite gas cell filled with two gases: argon and helium. Both the pressure dependent harmonic intensity and harmonic spectrum are clearly investigated. In addition, based on the theoretical fittings, the absorption cross-section of the harmonics is measured. We also show that the geometrical phase mismatch plays a more important role than the atomic dipole phase mismatch in this configuration. The high spatial coherence measured by employing a Young's double slit experiments allows us to confirm that intense and narrow bandwidth radiation in the extreme ultraviolet radiation which is indeed useful for coherent diffractive imaging can be generated as phase-matched and absorption-limited in the long interaction geometry.

## 2. Theoretical background

For the case of a slow intensity variation, which is important for the case of a long driving laser pulse, the driving field $E(z, t)$ and the field of the generated $q$th harmonic $E_q(z, t)$ propagating in an isotropic medium along the $z$ axis is given by [14].

$$E(z,t) = \frac{1}{2}A(z,t)\ \exp\left[i(kz-\omega t)\right] + c.c.,$$

$$E_q(z,t) = \frac{1}{2}\sum_q A_q(z,t)\ \exp\left[i(k_q z-\omega_q t)\right] + c.c., \tag{1}$$

where $k$, $k_q$ are the wave vectors of the driving laser pulse and the $q$th harmonic, respectively, $\omega$ is the angular frequency of the driving laser and $\omega_q$ is the angular frequency of the $q$th harmonic.

The propagation of the envelopes of the $q$th harmonic $A_q(z, t)$ in an absorbing medium along the optical axis of the laser pulse can be described by

$$\frac{dA_q}{dz} + \frac{\alpha_q}{2}A_q = i\frac{2\pi\omega_q}{cn_q}Nd_q\ \exp\left[-i\Delta k_q z\right] \tag{2}$$

where $\alpha_q$ is the absorption coefficient, $n_q$ is the refractive index for the $q$th harmonic, $N$ is the gas density, $d_q$ is the amplitude of the atomic dipole moment resulting from the fundamental laser and $\Delta k_q = k_q - qk$ is the phase mismatch between the driving field and the field of the $q$th harmonic. The phase mismatch $\Delta k_q$ along the propagation axis is the sum of four terms which are the neutral dispersion phase mismatch, the plasma dispersion phase mismatch, the geometric phase mismatch and the atomic dipole phase mismatch [10,11].

$$\Delta k(z) = q\left[N(1-\eta(t))\frac{2\pi}{\lambda}\delta n - \eta(t)Nr_e\lambda\right] + (\text{geometrical term}) + (\text{dipole phase term}) \tag{3}$$

where $\lambda$ is the centre wavelength of the fundamental pulse, $\eta(t)$ is the ionization fraction, $r_e$ is the classical electron radius, and $\delta n$ is the difference between the refractive indices, including the nonlinear components, at the fundamental and harmonic wavelengths.

In Eq. (3), the first two phase mismatches (neutral dispersion and plasma dispersion) are gas pressure dependent phase terms and the others are pressure independent terms. The major sources of phase mismatch are pressure-dependent, which allows the phase matching condition to be satisfied in a plasma with arbitrary electron density. Full phase matching can be achieved within a few optical cycles around the peak intensity of the laser pulse. The phase mismatch $\Delta k$ is a complex function of the atomic density which can be expanded in the form

$$\Delta k = a_0 + a_1 p + a_2 p^2 \tag{4}$$

where $a_0$, $a_1$, $a_2$ are coefficient and represent independence, linear dependence and nonlinear dependence of the phase mismatch on the gas pressure, respectively.

By integrating Eq. (2) over the medium length $L$, we obtain an expression for the intensity of the $q$th harmonic in the presence of the absorption [14]:

$$\begin{aligned} I_q &= \frac{\pi\omega_q^2}{2cn_q}\left|d_q^{NL}\right|^2 N^2 L^2\ \exp\left(-\frac{\alpha_q L}{2}\right)\frac{\sin^2\left(\frac{\Delta k_q L}{2}\right) + \sin h^2\left(\frac{\Delta k_q L}{4}\right)}{\left(\frac{\Delta k_q L}{2}\right)^2 + \left(\frac{\Delta k_q L}{4}\right)^2} \\ &= \frac{\pi\omega_q^2}{2cn_q}\frac{\left|d_q^{NL}\right|^2}{\sigma_q^2}\cdot\frac{1}{1+\delta^2}\left[1+\exp\left(-\tau_q\right) - 2\cos\left(\frac{\delta\tau_q}{2}\right)\exp\left(-\frac{\tau_q}{2}\right)\right] \end{aligned} \tag{5}$$

where $\sigma_q = \alpha_q/N$ is the absorption cross section, $\tau_q = \alpha_q L$ and $\delta = 2\Delta k_q/\alpha_q$.

The variation of the gas density affects not only the harmonic intensity as shown in Eq. (5), but also the spectrum of the HHG. Indeed, the change of the gas density induces a change in the refractive index leading to the self-phase modulation (SPM) of the propagating laser pulse in the medium during ionization [15–17]. The frequency shift of the driving laser spectrum is given as [17]

$$\Delta\omega = \frac{\omega}{2n_c c}\frac{\partial\bar{n}_e}{\partial t}L \tag{6}$$

where $c$ is the speed of the light, $n_c$ is the critical ionization, and $\bar{n}_e$ is the averaged electron density.

This spectral shift of the driving pulse in turn results in the fraction spectral shift of the $q$th harmonic which is independent on the harmonic orderand is given simply by

$$\frac{\Delta\omega_q}{\omega_q} = \frac{\Delta\omega}{\omega} \tag{7}$$

The harmonic spectrum can also be shifted because of the propagation of the harmonic signal in the ionizing medium. However, this shift is much smaller than the effect of the spectral shift of the driving field for the harmonics and can be neglected.

When the phase matching condition is achieve, i.e., $\Delta k \approx 0$, it is expected that the spectrally sharp, bright harmonics are generated which can be then employed in a lensless imaging technique known as coherent diffractive imaging (CDI). One of the crucial requirements of the CDI technique is spatial coherence which is about as large as the sample size times the linear oversampling factor. In order to determine the degree of spatial coherence, a Young's double slit which has slit spacing $d$, slit width $w$, and slit height $h$, is used. Choice of an appropriate value of h can ensure that the interferogram covers a reasonable number of pixels on the CCD in a direction perpendicular to the interference pattern. By illuminating a double slit, the fringe visibility, i.e., $\vartheta \approx (I_{\rm max} - I_{\rm min})/(I_{\rm max} + I_{\rm min})$, is measured. Here, $I_{\rm max}$ is the intensity of the centre and $I_{\rm min}$ is the intensity of the first minimum of the interference pattern. The source would be perfectly spatially coherent when $\vartheta = 1$.

## 3. Experimental results and discussion

The harmonic source in our experiments is driven by a 1 kHz, 800 nm multi-pass multi-stage chirped-pulse amplifier laser system (Quantronix Odin-II HE). This laser produces 30 fs pulses with