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Role of Gas Pressure in Quasi-Phase Matching in High Harmonics Driven by Two-Color Laser Field

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Abstract: The results of a study on the effect of pressure in a medium consisting of a set of gas jets separated by vacuum gaps, interacting with two-color laser fields formed by the fundamental and the second harmonics of a laser, are presented herein. It has been demonstrated that a decrease in pressure leads to a shift in the region of harmonics where quasi-phase matching (QPM) occurs towards shorter wavelength radiation, accompanied by an increase in the efficiency of amplification of these harmonics. A feature of this process is the identical power-law character of the shift in the region and the increase in the efficiency of harmonic QPM amplification. Additionally, the study presents the results of the effect of inaccurately setting the width of the gas jets on the shape of the spectrum of harmonic QPM amplification.

Keywords: high harmonics generation; quasi-phase matching; interference model; seeding FEL

1. Introduction

High-order harmonic generation (HHG) is one of the methods used to produce pulsed radiation in the ultraviolet (UV) and X-ray ranges [1]. The HHG effect was first observed in dense laser plasma generated by focusing powerful picosecond and nanosecond laser radiation on a target [2]. Later, this technique was studied on gases [3,4]. Despite its over 45-year history, the HHG effect continues to attract significant interest [5]. This interest is mainly due to the development of methods to increase the efficiency of conversion to the short-wavelength part of the spectrum, i.e., UV and X-ray radiation [6]; the development of methods to control the polarization state of the generated radiation [7]; the increase of its spectral width [1]; and the methods for generating attosecond pulses [8].

One of the classical and still popular mediums for HHG is atomic gas [9–11]. The low efficiency of conversion due to the low concentration of atoms in the gas can be considered a disadvantage of generating in gas jets, while an advantage is the relatively simple experimental setup. Despite the generally low efficiency of harmonics generated by gas, they can be successfully used as a source of extreme-ultraviolet field [12,13] of attosecond time-scale [14]. A review of modern methods for increasing the efficiency of HHG in continuous gas media is presented in [15], and plasma (which is the closest to the gas in terms of HHG mechanism media) is presented in [16]. In [15,16], methods for overcoming phase mismatch during the propagation of harmonic and laser radiation in the medium (Δk), which leads to atoms in the medium being spaced at a distance of $L_{coh} = \pi/\Delta k$, generating radiation that interferes destructively, are discussed. A phase shift in the wave was demonstrated when crossing the boundary between media [17–19], resulting in the frequency conversion process being effectively continued in another part of the medium. This has led to the development of quasi-phase matching (QPM) methods to increase the efficiency of the HHG.

A review of the features of phase matching and QPM is presented in [20]. The QPM effects have been actively studied in gases [17,21] and plasma media [20,22]. The QPM

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phenomenon is observed not only in HHG but also in the generation of the THz radiation in two-color laser fields interacting with a sequence of gas jets [23].

The HHG phenomenon that occurs when a two-color laser field propagates through a gas medium consisting of a set of gas jets separated by vacuum gaps was investigated in [24–26]. It was shown that both the interaction medium parameters (jet width, their number) and the two-color field parameters (primarily wavelength of the driver laser) allow for significant control over both the efficiency of the QPM amplification and the position of the QPM region of amplified harmonics, as well as their angular spectrum. It was also demonstrated that as the pressure in the medium increases, the region of the QPM harmonics shifts towards lower values, but the study of the influence of pressure on the QPM amplification was beyond the scope of the conducted research. The present work is dedicated to filling the aforementioned gap.

Moreover, we have shown that in the spectrum of the HHG by a set of gas jets, several groups of harmonics are amplified [24,25]. In this work, it is shown that the efficiency of the QPM amplification depends significantly on the accuracy of jet width determination.

2. Methods

For this study, we have used the interference model presented in [27] and non-perturbative theory of the single-atom response calculation discussed in [28]. The non-perturbative theory is based on the use of exact solutions of the boundary value problem of "an atom in the external field" when expanding the wave function of the Schrödinger equation, which describes the interaction of a single atom with an electromagnetic field in the non-relativistic approximation:

$$i\hbar\frac{\partial\psi(\vec{r},t)}{\partial t} = \left[\frac{1}{2m}\left(\vec{p} - \frac{q}{c}\vec{A}(\vec{r},t)\right)^2 + U(\vec{r})\right]\psi(\vec{r},t),$$

where $U(\vec{r})$ is the intra-atomic potential, $\vec{A}(\vec{r},t)$ is the vector potential of the external electromagnetic wave, $\psi(\vec{r},t)$ is the wave-function of the valence electron, $\vec{p}$ is the momentum operator, $\hbar$ is the Planck constant, c is the speed of light in vacuum, and q and m are the charge and mass of the electron, respectively. The use of exact solutions of the boundary value problem of "an atom in the external field" allows us to take into account the symmetry transformation of the an electron interaction problem in a spherically symmetric Coulomb potential with a laser field, the intensity of the electric component of which becomes comparable to the intra-atomic one when calculating the current of the atomic response, the spectrum of which in the far-field zone coincides with the spectrum of the generated electromagnetic radiation [28].

The non-perturbative approach was applied to calculate the parameters of harmonics (their amplitudes and phases) generated by single atoms located in a gaseous medium and interacting with a laser field, the parameters of which depend on the coordinates of the atoms due to propagation. The obtained information was used to calculate the response of an extended medium, which is a chain of atoms distributed along with the laser field propagation direction using an interference model [27]. It possesses a unique combination of advantages: it allows calculation of the response of the medium, taking into account the responses of single atoms calculated quantum-mechanically using the developed non-perturbative theoretical approach [28]. This makes it possible to trace the modifications of photoemission spectra when laser radiation propagates in the medium, preserving the spectral features of the response of single atoms, which becomes critically important when studying high-order harmonics, as it is assumed that the nature of the HHG is based on the response of a single atom.

The method (calculation of amplitudes and the phases of harmonics generated by single atoms which are used to calculate the overall harmonics field at the detector in the far-field zone within the interference model) was verified in a study of the QPM concept in plasma plumes media for silver plasma [26,29], indium plasma [30], and clusters [31] by direct comparison between the calculated and measured peculiarities of harmonic generation. A good coincidence was shown between numerically calculated and experimentally measured phenomena.

In numerical simulations, we assumed that the perforated gas media consisted of 50 Ar gas jets with spatial sizes $d + d_1 \cdot f$ divided by the same-size free spaces forming the multi-jet gas medium (MJGM) (please see inset in Figure 1). In the current research, d = 50 µm, and by changing d_1, the influence of the non-ideal determination of the jet width on the shape of the amplification spectrum was investigated. MJGM interacts with a two-color laser field of moderate intensities (~10^{14} W/cm^2 for the fundamental radiation and ~10^{13} W/cm^2 for the second harmonic corresponding to the ~10% of conversion efficiency of laser energy in the second harmonic) consisting of two 30 fs linearly polarized components with a Gaussian envelope and a zero degree angle between the polarization directions of the Ti:Sa driver laser [24]. As for relative phase (delay time) between the two-color components, it was chosen to be equal to zero at the first layer of the gas, and it changed along with laser field propagation.

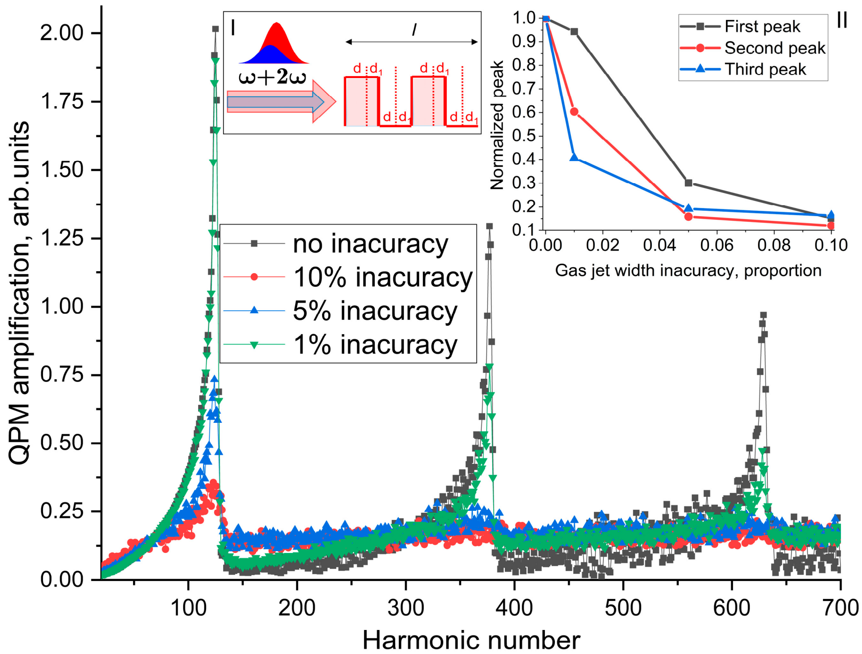

Figure 1. The QPM amplification in the MJGM with d = 50 mkm and d_1 = 0 (black curve with squares), d_1 = 0.01 d (green curve with inverse triangles), d_1 = 0.05 d (blue curve with triangles), and d_1 = 0.1 d (red curve with circles) interacting with the two-color laser field formed by the fundamental and the second harmonics of the Ti:Sa laser (λ = 800 nm). The calculations were performed for the pressure of 230 mbar and the total MJGM (50 jets) length of 0.5 cm. Inset: I. A schematic setup. II. Normalized peak values as a function of gas jet inaccuracy value.

It was assumed that the parameters of laser field change during the propagation in Ar gas due to dispersion. The influence of free electrons was taken into account in accordance with the Lorentz classical theory [32]. Re-absorption of the generated radiation was not taken into account. It was also assumed that the laser field is a plain wave. As a result, no influence of the Gouy phase was taken into account in the calculation. The latter can be achieved in loose focusing configuration.

To present the results of the calculation, it was decided to demonstrate the ratio of the calculated total extended gas harmonics spectrum and the typical single-atom spectrum instead of the total extended gas harmonics spectrum. As a result, we defined the QPM amplification as the ratio of the MJGM and the single-atom response (the QPM amplification of the given harmonics was calculated as the ratio of harmonics efficiency calculated within the interference model [27] and harmonics efficiency calculated within the non-perturbative theory [33]). It was close to the enhancement factor (the ratio of harmonics spectra generated by the multi-jet plasma and the one generated by the extended plasma) which is used to study the QPM effects in plasma plumes [34]. Such a definition of the QPM amplification is satisfactory for studying generated spectrum modification due to collective effects, the influence of intra-atomic non-linearities (i.e., single-atom response) is almost neglected, and as a result, the QPM amplification can be defined despite of the small efficiency in the single-atom response.

3. Results

To investigate the influence of the accuracy of determining the geometric dimensions of the MJGM, calculations of the spectra of the QPM amplification in the MJGM were carried out with variations of d_1, which was set within the range (0, 0.1 d). For this study, f was chosen as a set for a random variable defined within (-0.5, 0.5) and changed its value when each MJGM jet was formed. Figure 1 shows the spectra of the QPM amplification in the MJGM calculated for $d_1 = 0$ (black curve with squares), $d_1 = 0.01$ d (green curve with inverse triangles), $d_1 = 0.05$ d (blue curve with triangles), and $d_1 = 0.1$ d (red curve with circles). It can be seen that with accurate jet width determination, a sequence of amplification peaks can be observed in the spectrum [24,25]. However, inaccurate jet width determination primarily affects the efficiency of higher-frequency QPM amplification peaks. Thus, at $d_1 = 0.01$ d, the efficiency of the first QPM amplification peak decreases by 5%, while the amplitude of the second and third QPM amplification peaks decreases to 60% and 40%, respectively. Further increasing the value of d_1 leads to a more significant decrease in all QPM amplification peaks (see inset of Figure 1). Apparently, such a high sensitivity of the QPM amplification efficiency to the accuracy of determining and regulating the widths of gas jets is an obstacle to experimental confirmation of the presence of the high-frequency QPM amplification peaks.

The conducted research, in which a linear, quadratic, exponential, and sinusoidal function of the order number of the gas jet was used as f, allowed us to calculate the response of the MJGM with changing geometric dimensions of the gas jets according to the specified law when transitioning from jet to jet. However, this did not reveal any new possibilities for increasing the efficiency of the QPM amplification compared to the regular determination of jet sizes. Nevertheless, it should be noted that when sinusoidally changing the widths of the gas jets with an amplitude of $d_1 = 0.1$ d, an increase in the width of the first QPM amplification peak is observed with a simultaneous four-fold reduction in the total amplitude (see the red curve with circles in Figure 2). Amplification of a relatively large group of harmonics can be useful in the creation of attosecond pulses. In the calculations carried out, it was assumed that $f = sin((2\pi/50)\cdot n)$, where n is the order number (index) of the gas jet in the medium (variates from 1 to 50).

Figure 3 shows the spectra of the QPM amplification calculated at different values of pressure in the gas jets (for these studies, d_1 was chosen to be equal to zero). It can be seen that in full accordance with the formula presented in [24], the position of the QPM amplified harmonic peak changes inversely proportional to the pressure in the medium; by reducing the pressure in the medium, it was possible to shift the peak of the QPM amplification to the short-wavelength region of the spectrum (see Figure 3a and black curves on Figure 3b). This can be explained by the fact that the coherence length is inversely proportional to the Δk, and at the same time, Δk for the plane wave is proportional to $p \cdot h$ (p is a pressure in the gas; h is a harmonic order) [35]. As a result, the equality of coherence length to the gas jets' spatial size (d) was achieved for higher harmonics with lower values of gas pressure. At the same time, calculations show that, together with the shift of the peak to the short-wavelength region of QPM amplification, its amplitude increases. This counterintuitive result (when the pressure decreases, the number of atoms generating harmonics also decreases) can be explained by the dependence of the refractive index on pressure—with a decrease in pressure, the period of oscillations of the efficiency of harmonic generation by atoms in the medium [27] increases, which leads to an increase in the number of atoms generating radiation that interferes constructively.

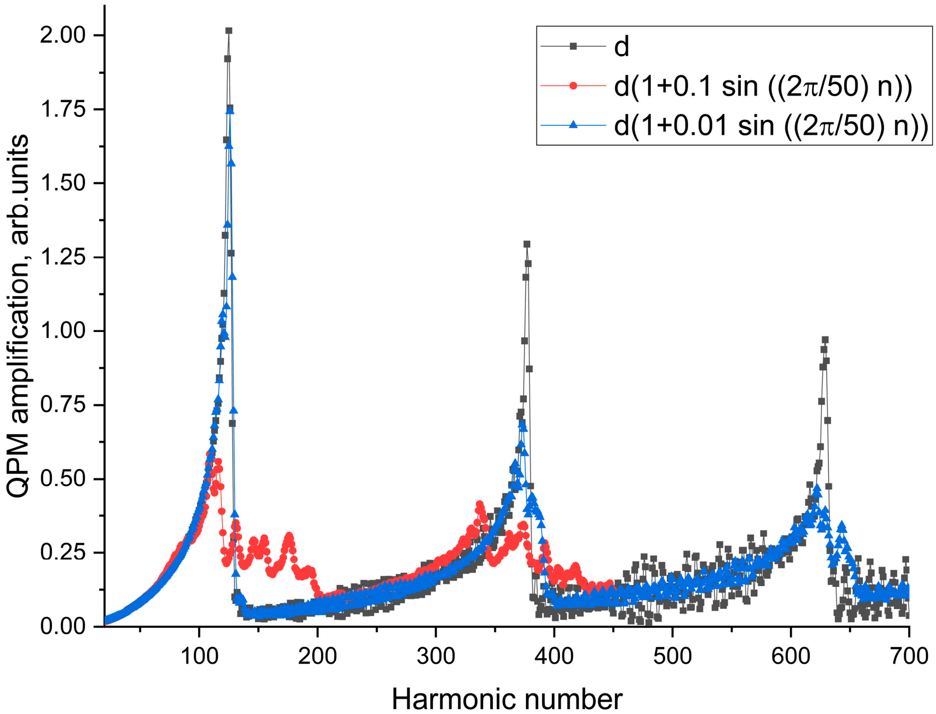

Figure 2. The QPM amplification in the MJGM with d = 50 mkm, $f = sin((2\pi/50){\cdot}n)$ and $d_1 = 0$ (black curve with squares), $d_1 = 0.01$ d (blue curve with triangles), and $d_1 = 0.1$ d (red curve with circles) interacting with the two-color laser field formed by the fundamental and the second harmonics of the Ti:Sa laser (λ = 800 nm). The calculations were performed for the pressure of 230 mbar and the total MJGM (50 jets) length of 0.5 cm.

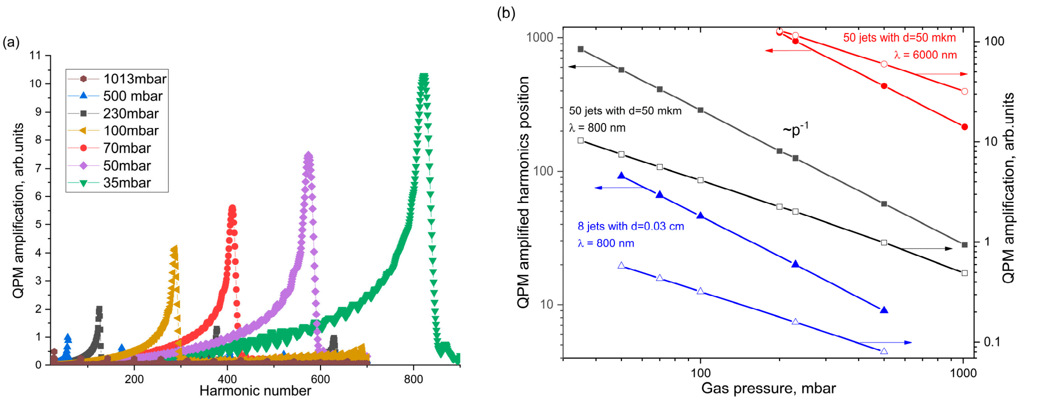

Figure 3. (**a**) The QPM amplification in the MJGM with d = 50 mkm interacting with the two-color laser field formed by the fundamental and the second harmonics of the Ti:Sa laser (λ = 800 nm). The calculations were performed for the different values of gas pressures. The total MJGM (50 jets) length of 0.5 cm. (**b**) Position of the QPM amplified harmonics (solid points) and the value of QPM amplification (open points) as a function of pressure calculated for the MJGM with d = 50 mkm interacting with the two-color laser field formed by the fundamental and the second harmonics of the Ti:Sa laser (black curves), for the MJGM with d = 50 mkm interacting with the two-color laser field formed by the fundamental and the second harmonics of the mid-IR laser (λ = 6000 nm) (red curves), and for the MJGM with d = 0.03 cm interacting with the two-color laser field formed by the fundamental and the second harmonics of the Ti:Sa laser (blue curves).

In spite of the fact that for the laser field parameters used in the numerical experiment (for the MJGM with d = 50 mkm interacting with the two-color laser field formed by the fundamental and the second harmonics of the Ti:Sa laser) the QPM-amplified harmonics lie beyond the cut-off frequency [36], the proposed optimization of harmonics efficiency can be useful, since the position of the QPM-amplified harmonics (in photon energies) does not depend on the carrier laser wavelength [24] and the given QPM-amplified harmonic can lie ahead of the cut-off frequency if, for example, the carrier laser wavelength increases [1].

To demonstrate this, calculations for the MJGM with d = 50 mkm interacting with the two-color laser field formed by the fundamental and the second harmonics of the mid-IR laser (λ = 6000 nm; other parameters of the laser field, such as the values of two-color laser field components and the number of the field's oscillations inside the temporal envelope coincide with the one for the Ti:Sa laser) were carried out (see the red curves in Figure 3b). It was demonstrated that both the position of the QPM-amplified harmonics and the value of QPM amplification are inversely proportional to the value of the gas pressure.

Another method to shift the position of the QPM-amplified harmonic ahead of the cut-off frequency (to low-order region) is to increase the value of the gas jets' length (d) [26]. The following calculations for the MJGM with d = 0.03 cm interacting with the two-color laser field formed by the fundamental and the second harmonics of the Ti:Sa laser were also carried out (see the blue curves in Figure 3b). The detected trends in the gas pressure dependencies were also demonstrated.

As a result, the similar dependence of the position of the QPM-amplified harmonics and the value of QPM amplification under variation the pressure in MJGM with different values of d and laser wavelength has been demonstrated. For all the results presented in Figure 3b, the total length of the MJGM (l) was kept the same.

4. Conclusions

The interference model of the HHG by the extended gas media was applied to the investigation of the QPM amplification under variation of gas pressure. It was shown that the position of the QPM-amplified harmonics and the QPM amplification are inversely proportional to the value of the gas pressure; lower gas pressure value corresponds to a higher QPM amplification value and the QPM-amplified harmonics position (shorter wavelength). The results of the theoretical simulations open wide prospects of efficient harmonics generation with high photon energies.

It was also demonstrated that QPM amplification is very sensitive to the quality of the geometrical characteristics of the MJGM. Even a 1% error in determining the sizes of gas jets can significantly change the spectrum of QPM amplification by substantially reducing the efficiency of generating short-wavelength peaks. This can be an additional opportunity to isolate a given harmonic from the spectrum for possible use as a seeding radiation in free electron lasers [37].

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