Effects of pressure and gas-jet thickness on the generation of attosecond pulse*

Li Xiao-Yong(李小勇)[a)b)], Wang Guo-Li(王国利)[a)], and Zhou Xiao-Xin(周效信)[a)†]

[a)] College of Physics and Electronic Engineering, Northwest Normal University, Lanzhou 730070, China
[b)] Experimental Center, Northwest University for Nationalities, Lanzhou 730030, China

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We investigate how the intensity and duration of an attosecond pulse generated from high-order harmonic generation are affected by the pressure and thickness of the gas jet by taking into account the macroscopic propagation of both fundamental and harmonic fields. Our simulations show that, limited by the propagation effects, especially the absorption of harmonics, the intensity of an attosecond pulse cannot be improved by just independently increasing the gas pressure or the medium length. On the other hand, due to good phase-matching conditions, the duration of a generated attosecond pulse can be improved by changing the gas pressure.

Keywords: high-order harmonic generation, attosecond pulse, macroscopic propagation

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1. Introduction

For attosecond probing and other potential applications, it is crucial to increase the attosecond pulse energy. In the last decade, it has been proven that, under the right conditions, the high-order harmonic generation (HHG) can produce extreme ultraviolet (XUV) pulses with sub-femtosecond duration, i.e., attosecond duration.[1] These pulses provide scientists with the shortest controllable probes currently available for probing ultrafast dynamics; their application has resulted in an exciting and vibrant research field known as attosecond science.[2,3] Their main drawback, however, is the low conversion efficiency,[4,5] which limits many applications. Even today, very few research groups have a working attosecond spectroscopic setup in their laboratory.[6] Therefore, improving the intensities of attosecond pulses (AP) is an important issue for attosecond science.

As the experimentally observed HHG spectra are generated from the interaction of an intense laser field with a macroscopic medium, the full description of the HHG should involve a quantum treatment of the harmonic emission from a single atom or molecule, together with the propagation of the laser light and harmonics in the medium.[7–9] While a single-atom HHG process can be understood easily using a semi-classical three-step model,[10,11] the macroscopic harmonic generation is complicated. The laser field and the harmonics can be dispersed and absorbed by the medium.[12] The intense laser light also ionizes the atoms and generates free electrons, thus changing the optical properties of the gas medium.[13] As a result, the HHG yield will not be increased monotonously by increasing the laser intensity, gas pressure and gas-jet length. Clearly, a proper description for these effects is important to understand the generation of harmonics and AP. We note that although there have been some studies of the propagation effects on the HHG,[14–16] the studies of AP are still scarce.

Recently, we successfully simulated the macroscopic HHG spectra of Ar, Ne, Xe and Kr.[17,18] By solving the proper Maxwell wave equations, the simulations showed a good agreement with the experimental measurements. In this paper, our goal is to investigate how the pressure and the thickness of the gas jet affect the generation of AP.

2. Theory

A detailed description of the theoretical method used in these simulations can be found in Ref. [19]. Briefly, we consider the propagation of both the fundamental and the harmonic fields in an ionizing medium. For the fundamental field, we include the dispersion, absorption, Kerr and plasma effects. For the harmonics, the dispersion and absorption due to the neutral atoms are included. We only describe the main equations below.

The evolution of a fundamental driving laser $E_1(r,z,t)$ in an ionizing medium is described by a three-dimensional (3D) Maxwell equation

$$\nabla^2 E_1(r,z,t) - \frac{1}{c^2}\frac{\partial^2 E_1(r,z,t)}{\partial t^2} = \mu_0 \frac{\partial J_{\rm abs}(r,z,t)}{\partial t} + \frac{\omega_0^2}{c^2}(1-\eta_{\rm eff}^2)E_1(r,z,t), \quad (1)$$

where

$$\eta_{\rm eff}(r,z,t) = \eta_0(r,z,t) + \eta_2 I(r,z,t) - \frac{\omega_{\rm p}^2(r,z,t)}{2\omega_0^2} \quad (2)$$

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†Corresponding author. E-mail: zhouxx@nwnu.edu.cn
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is the effective refractive index of the gas medium. The first term $\eta_0 = 1 + \delta_1 - i\beta_1$ takes into account the refraction (δ_1) and absorption (β_1) effects of the neutral atoms; the second term accounts for the optical Kerr nonlinearity; and the third term is from free electrons. The absorption term J_{abs} due to the ionization of the medium is expressed as

$$J_{abs}(t) = \frac{\gamma(t) n_e(t) I_p E_1(t)}{|E_1(t)|^2}, \tag{3}$$

where $\gamma(t)$ is the ionization rate, which can be calculated from the Ammosov–Delone–Krainov (ADK) theory,[20,21] and I_p is the ionization potential.

The 3D propagation equation for the harmonic field $E_h(r,z,t)$ is

$$\nabla^2 E_h(r,z,t) - \frac{1}{c^2}\frac{\partial^2 E_h(r,z,t)}{\partial t^2} = \mu_0 \frac{\partial^2 P(r,z,t)}{\partial t^2}, \tag{4}$$

where $P(r,z,t)$ is the polarization and can be separated into linear and nonlinear components $P(r,z,t) = \chi^{(1)}(t)E_h(r,z,t) + P_{nl}(r,z,t)$. Here, the linear susceptibility is related to the refractive index $n(\omega)$ and scattering factors (f_1, f_2) by

$$\begin{aligned} n(\omega) &= \sqrt{1+\chi^{(1)}(\omega)/\varepsilon_0} \\ &= 1 - \delta_h(\omega) - i\beta_h(\omega) \\ &= 1 - \frac{1}{2\pi} n_0 r_0 \lambda^2 (f_1 + i f_2), \end{aligned} \tag{5}$$

where $\delta_h(\omega)$ and $\beta_h(\omega)$ represent the dispersion and the absorption of the harmonics by the medium, respectively. The nonlinear polarization term $P_{nl}(r,z,t)$ can be expressed as

$$P_{nl}(r,z,t) = [n_0 - n_e(r,z,t)]D(r,z,t), \tag{6}$$

where n_0 is the neutral atom density, $n_e(r,z,t)$ is the free-electron density, which can be calculated from the ADK theory. $D(r,z,t)$ is the single-atom induced dipole moment caused by the fundamental driving laser field, which can be obtained by using a strong-field approximation (SFA).

Macroscopically, when harmonics are emitted at the exit plane, a spectral filter is usually used to select a range of harmonics (ω_2–ω_1) to synthesize AP. In theory, we can obtain the total signal of APs as[22]

$$I(t) = \int_0^\infty 2\pi r\,dr \left| \int_{\omega_1}^{\omega_2} E_h(r,\omega) e^{i\omega t} d\omega \right|^2. \tag{7}$$

3. Results and discussion

We first simulate the macroscopic HHG spectra of Ne generated by a 800-nm, 8-fs full width at half maximum (FWHM) laser pulse. We assume that the laser beam at the entrance of the gas jet has a Gaussian shape in both time and space, the laser beam waist in the center of the gas jet is 25 μm and all the harmonic signals (i.e., the total signal) from the exit of the medium are collected. From the HHG spectra, we then obtain the APs according to Eq. (7) by using the harmonic orders from 45 to 65. Below we aim at the gas pressure and jet length effects on the intensity and FWHM of the generated AP. For the energy range considered here, four pulses can be produced; we only focus on the strongest one.

Figure 1 shows the dependence of the AP intensity on the gas pressure for several different lengths of gas jet (0.5 mm, 1.0 mm, 1.5 mm, 2.0 mm, 2.5 mm and 3.0 mm), the laser peak intensity is 3.0×10^{14} W/cm^2 at focus (for each length, the center of the gas jet is always placed 2 mm after the laser focus). From the figure, we can see that for each fixed thickness, the intensity curve of the AP goes up with increasing gas pressure if the pressure is relatively low; once the intensity reaches its maximum (optimal) value at some pressures, it starts to decrease with increasing gas pressure. On the other hand, if the gas pressure is fixed, the intensity of the AP also cannot be improved by just increasing the gas-jet thickness. For example, at the pressure of 80 Torr, 2.0 mm is the optimal length of the gas jet, rather than 2.5 mm or 3.0 mm.

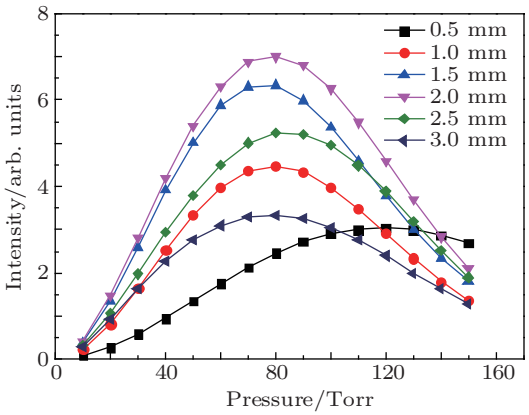


Fig. 1. (color online) The AP intensity of Ne as a function of gas pressure for different gas-jet thicknesses. The laser peak intensity in the center of the gas jet is 3.0×10^{14} W/cm^2.

The above variation must come mainly from the pressure dependence of the harmonic generation yield. In the HHG simulation, we include the dispersion δ_1 and Kerr nonlinearity η_2 of the fundamental field, and the dispersion δ_h and absorption β_h of the harmonic fields. All of these effects can limit the harmonic generation and they are all pressure dependent. To see how each term contributes to the HHG yield, we remove each term successively from our model. We find that the absorption of harmonics is the most important effect under the conditions in our simulation. In Figs. 2(a) and 2(b), we show the harmonic spectra without and with β_h in the model, respectively, for three pressures, 10 Torr, 80 Torr and 150 Torr, at the laser peak intensity of 3.0×10^{14} W/cm^2 for a 3-mm-long gas jet. By comparing Fig. 2(a) with Fig. 2(b), we can see that without absorption, the harmonics will grow monotonously (quadratically) with the number of gas atoms. In contrast, if

the absorption is considered, the yield increases first to reach a maximum at 80 Torr and finally drops with even higher pressure for harmonics higher than H30. (Note that the scattering factor f_2 is large near 50 eV, see the inset of Fig. 2(b).) In fact, the intensity of the HHG (or the AP intensity) is determined by both the radiation and the absorption of atoms in the medium. Both are proportional to the neutral atom density n_0 (or the number of atoms in the gas), but they are of different scales with pressure increasing. When the gas pressure is lower, n_0 is also lower and the HHG radiation of atoms is greater than the absorption effect. As the pressure goes up, the intensity of the HHG increases accordingly. However, as the pressure reaches saturation, the absorption effect becomes more important than the radiation, so the intensity of the HHG decreases with increasing pressure. The dependence upon the gas-jet length of the AP intensity in Fig. 1 can also be understood in the same way.

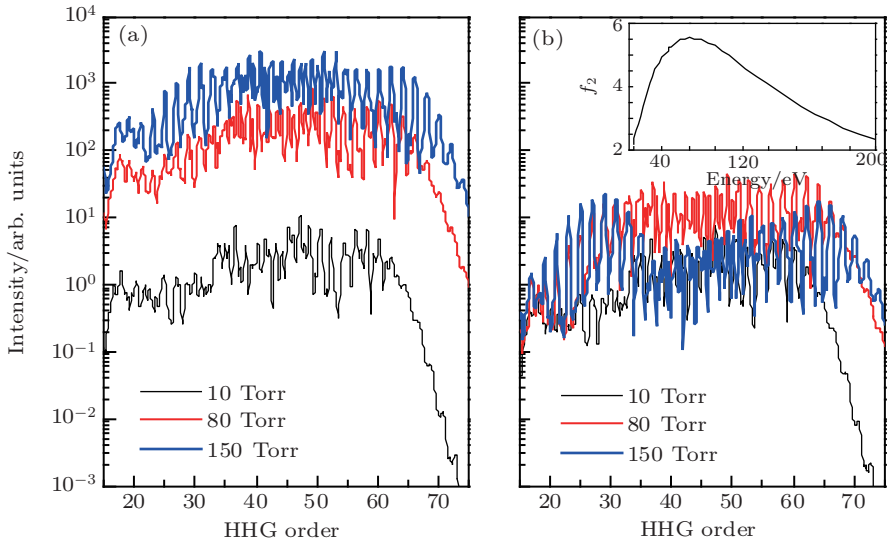


Fig. 2. (color online) The influence of absorption of harmonic field. (a) HHG spectra of Ne for pressures of 10 Torr (black), 80 Torr (red) and 150 Torr (blue) without considering β_h in the model. The laser peak intensity in the center of the gas jet is 3.0×10^{14} W/cm^2. The gas-jet length is 3 mm. (b) The same HHG spectra for three pressures under the same laser conditions but including all effects. The atomic scattering factor f_2 for Ne,[23] which is related to β_h, is shown in the inset.

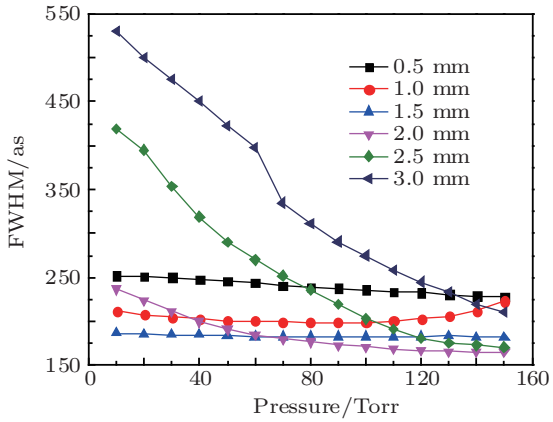


Fig. 3. (color online) FWHM of the AP as a function of gas pressure for several different gas-jet thicknesses. The laser peak intensity in the center of the gas jet is 3.0×10^{14} W/cm^2.

The duration (i.e., FWHM) is another important parameter for AP. In Fig. 3, we show the AP FWHM with changing gas pressure for several lengths of the gas, using the peak laser intensity of 3.0×10^{14} W/cm^2 in the center of the gas jet. For short gas jets (0.5 mm, 1.0 mm and 1.5 mm), we can see that the pressure has little influence on the AP FWHM. However, with increasing the gas-jet length (>2 mm), the pressure begins to play an important role. The AP FWHM can be improved from 530 as to 220 as with a 3-mm-long gas jet, for example, when the pressure increases from 10 Torr to 150 Torr. It should be noted that for other experimental conditions, i.e., for a higher laser intensity, an optimal pressure may also be found that will generate the shortest AP. Figure 3 also shows that for a given pressure, there is an optimal jet length to generate the shortest AP.

Because the phase differences between consecutive harmonics are significant for the generation of attosecond pulses,[24,25] to understand the results shown in Fig. 3, we extract the harmonic phase differences at radial distance $r=0$ mm for two gas-jet lengths (1 mm and 2.5 mm) at the three pressures of 10 Torr, 80 Torr and 150 Torr; the results are shown in Fig. 4. From Fig. 4(a), we can see that for a short gas jet, the phase differences versus the harmonic order for the three pressures are all regular, and the differences between them are tiny, especially in the range of H50–H64, so the FWHMs of the APs generated by these harmonics are nearly independent of pressure. On the other hand, if the gas jet is long (e.g., 2.5 mm), the phase difference versus the harmonic order for low pressure (10 Torr) is irregular (see Fig. 4(b)); correspondingly, the FWHMs of the APs generated by these harmonics are wide. With increasing gas pressure, the phase difference becomes more and more regular, which leads to a narrower FWHM of the AP.

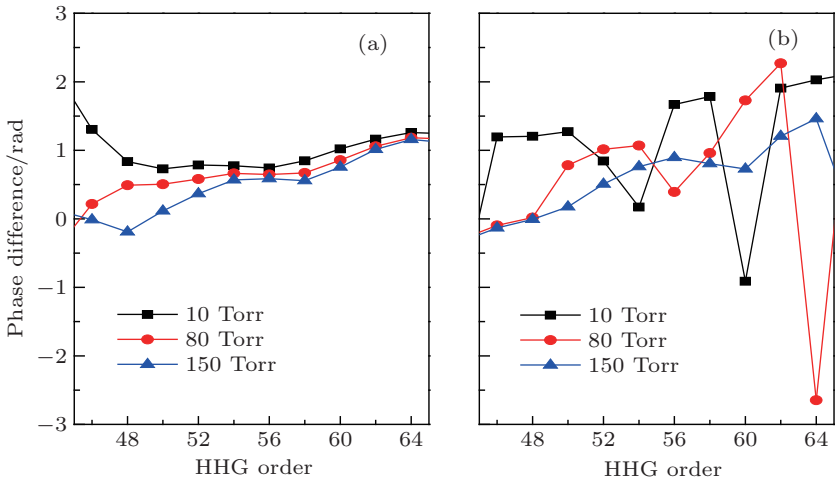


Fig. 4. (color online) The phase difference between neighboring harmonics at $r = 0$ mm generated in (a) 1-mm gas jet and (b) 2.5-mm gas jet for pressures of 10 Torr (black squares), 80 Torr (red dots) and 150 Torr (blue triangles).

4. Conclusion

Our research indicates that the intensity of APs does not increase continuously with increasing pressure and increasing thickness of the gas jet. Actually, because of the harmonic absorption, it shows a maximum at a certain pressure (jet thickness) for a fixed gas jet thickness (pressure). In other words, there is an optimal pressure for a fixed gas jet thickness and an optimal thickness for a fixed gas pressure to generate APs. On the other hand, for most gas jet thicknesses, the FWHM curve of the APs goes down continuously when the pressure goes up at this low laser intensity. However, for a fixed pressure, the FWHM varies with the gas-jet thickness and has a minimum. Therefore, in the future, to improve the intensity and bandwidth of APs, the gas pressure and jet thickness can be optimized simultaneously with the well-established macroscopic propagation model.

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