Spectral splitting in high-order harmonic generation

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Using superintense 46-fs Ti:sapphire laser pulses, high-order harmonic generation in argon gaseous medium is experimentally studied. When the driving laser intensity is pushed well above the ionization threshold (saturation intensity) of the medium applied, the double-peak spectral splitting of high harmonics has been clearly observed. This splitting is mainly due to the propagation effects in the rapidly ionizing medium, which is verified by means of one-dimensional calculations. The numerical simulations agree well with the experimental results.

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I. INTRODUCTION

Many nonperturbative phenomena appear when an atom is irradiated by an ultrashort superintense laser field, among which high-order harmonic generation (HHG) is one of the most important subjects [1]. In the past decade, HHG has been extensively studied both theoretically and experimentally, and much progress has been made. On one hand, HHG is a promising tunable source for coherent XUV and soft x rays [2]. On the other hand, HHG paves the way to the production of subfemtosecond pulses [3]. X rays in the ''water window'' has been observed by means of HHG in a helium gaseous medium using 5-fs laser pulses [4].

In general, the laser pulses with the intensity of 10^{14}–10^{15} W/cm^2 can efficiently excite HHG in a noble gaseous medium, provided the gas density is high enough. During the process of HHG, a well-known effect is the inherent ionization of the used medium, which reduces the conversion efficiency as a consequence of the neutral atom population depletion. When the intensity of the driving laser pulses is much higher than the saturation intensity of the medium applied, the ionization causes a rapid increase in electron (plasma) density on the optical axis and hence a decrease of the refractive index in time and space, resulting in the spectral blueshifting and broadening of the fundamental field and its harmonics, as well as the self-defocusing of the driving laser pulses. The propagation of the driving laser field and its harmonics in ionizing gaseous medium plays a fundamental role in HHG and laser-plasma interaction.

The propagation dynamics of the fundamental laser field in ionizing a helium gaseous medium were studied and the spectral splitting was observed by Chessa *et al.* [5]. The similar splitting of intense femtosecond fundamental pulses in a dispersive bulk medium was observed [6]. An experimental observation of the spectral splitting in high harmonics was reported by the Michigan group [7]. By allowing harmonic emission twice during the one laser pulse with the help of a birefringent quartz plate, the splitting in the harmonic spectra which was mainly attributed to the atomic physics process was also observed by Altucci and coworkers [8].

In the present paper we report on an experiment in which we clearly observed the double-peak spectral structures in high harmonics generated in an argon gaseous medium when pushing the driving intensity well above the saturation intensity of argon atoms. This spectral splitting is mainly due to the propagation effects of high harmonics in the ionizing medium, which is verified by means of one-dimensional calculations. This paper is organized as follows. Section II gives a brief description of the experimental setup and procedure. We present the experimental results and discussion in Sec. III. A brief summary is given in Sec. IV.

II. EXPERIMENTAL SETUP

The laser facility used in our experiments, which is based on the chirped pulse amplification technique and operated at 10 Hz, can produce pulses of 250-mJ maximum energy, 46-fs minimum full width at half maximum (FWHM), with a 57-nm FWHM spectrum centered at 790 nm. The argon gas is supplied by a solenoid-valve gas jet with a 0.5-mm-diam nozzle located inside a vacuum chamber. The gas density under the nozzle end can be varied by adjusting the backing pressure. The linearly polarized 50-mm-diam laser beam is focused by a 50-cm-focal-length lens into the vacuum chamber. The gas target is placed 3 mm after the focus, where the beam diameter is approximately 200 μm, measured with a charge-coupled-device (CCD) camera. The vertical distance between the focus and the nozzle exit is about 2 mm. The gas pressure in the target chamber remains lower than 10^{-4} Torr when experiments are being conducted.

The harmonic signals are dispersed by a grazing incidence monochromator (λ-Minutemen 310G), with a 1-m-radius Au-coated grating of 300 grooves/mm. The blazing wavelength of the grating is 6.09 nm at the grazing incidence angle of 2°. The entrance and exit slit of the spectrometer are 100 and 130 μm, respectively. The distance along the optical axis between the gas target and the entrance slit is about 45 cm so as to avoid possible damage of the grating

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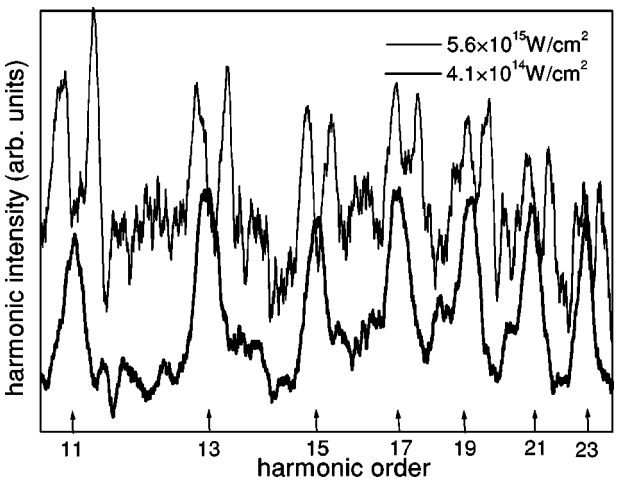

FIG. 1. Measured harmonic spectra in a 50 Torr argon gaseous medium with different driving laser intensities: (a) 5.6×10^{15} W/cm^2 (thin line) and (b) 4.1×10^{14} W/cm^2 (thick line).

due to the superintense driving laser. The spectral profile of the driving laser can be real-time monitored with a CCD camera when the harmonic spectra are being acquired. The details of the experimental diagram are referred to in Ref. [9].

III. EXPERIMENTAL RESULTS AND DISCUSSION

The measured high harmonic spectra in an argon gaseous medium irradiated by different driving intensities under the gas pressure of 50 Torr are shown in Fig. 1. Both of the spectra are obtained by averaging the signals over 300 shots, and not corrected for the response of the detection system. When the energy of the driving pulse is $82(\pm2)$ mJ (corresponding to the laser intensity of 5.6×10^{15} W/cm^2 in the interaction volume under our focusing geometry), the splitting of the harmonics is clearly shown in the spectrum. The appearance of the double-peak structures is impossible due to the second-order diffraction of the corresponding harmonics because the strength of the harmonics beyond order 23 is too weak, which could be seen clearly in Fig. 1. Comparatively, there is no observable splitting when the driving intensity is about 4.1×10^{14} W/cm^2.

The splitting of high harmonics observed would be easily interpreted, if there existed double-peak structures or the like in the spectral profile of the driving laser. We carefully examined and dismissed this possibility. Firstly, the spectral profile of the laser monitored with a CCD camera does not exhibit any sort of double peaks. Secondly, the spectral profile of the second harmonic in a helium gaseous medium (due to the presence of density gradients in the plasma) measured by another group in our laboratory does not show such structures either [10]. Finally, although the ionization of the medium may significantly affect the spectral properties of the fundamental laser, this is unlikely to cause the double-peak structures but rather only an overall spectral blueshifting. Therefore, the double-peak splitting of high harmonics observed is unlikely derived from the structures of the driving laser field itself. There must be another mechanism(s) answering for the spectral splitting.

It is now well known that HHG results from two aspects, i.e., the single-atom response and the propagation effects. The propagation effects may significantly modify the result of the single-atom response. Kan and his co-workers [11] studied HHG in ionizing atomic gases, and found that atomic process (single-atom response) could also lead to spectral splitting and blueshifting as a consequence of an intensity- or time-dependent phase shift of the dipole acceleration. However, in the view of experimental measurements, the splitting aroused by atomic response may be very difficult to observe according to the results of Ref. [11]. The spectral splitting we observed is so striking that it is unlikely to be due only to the single-atom response.

When the splitting emerges in the high harmonic spectra, the laser intensity of driving pulses applied is far higher than the saturation intensity of the neutral argon atoms, which is about 4×10^{14} W/cm^2, taking into account the effect of the 46-fs duration [12]. There must then be a volume of electron plasma with a considerable time-dependent density in the interaction regime, which makes it complicated to explore the physical mechanism governing the HHG in ionizing medium.

It is instructive to see how the propagation effects modify the harmonic fields, especially when there is a rapid ionization in the medium involved. To do so, we solve the one-dimensional wave equation by using the slowly varying envelope (SVE) and paraxial approximations [13]. In the case of the plane wave, the wave equations of the fundamental field and its harmonics in the isotropic ionizing medium along the **Z** axis (propagation direction) are given by

$$\frac{\partial^2 E(z,t)}{\partial z^2}=\frac{\mu n_e(t)e^2}{m_e}E(z,t)+\mu\varepsilon\frac{\partial^2 E(z,t)}{\partial t^2}+\mu\frac{\partial^2 P_{NL}(z,t)}{\partial t^2}, \tag{1a}$$

$$E(z,t)=E_1(z,t)+E_q(z,t)\quad (q>1), \tag{1b}$$

where $E_1(z,t)$ and $E_q(z,t)$ are the fundamental and the qth harmonic field, $n_e(t)$ is the time-dependent electron density, m_e the electron mass, ε the electric permittivity, and μ the magnetic permeability. We neglect the absorption of the medium. The nonlinear polarization $P_{NL}(z,t)$ is given by [14]

$$P_{NL}(z,t)=N_0[1-W(t)]D_q(t)\exp[-iq(\omega_1 t-k_1 z)], \tag{2}$$

where $N_0=1.6\times10^{18}$ cm^{-3}(50 Torr) is the initial density of the neutral gas, $D_q(t)$ is the atomic dipole moment, and ω_1 and k_1 are the angular frequency and wave number of the fundamental field. $W(t)$ is the tunneling ionization rate obtained under the Ammosov, Delone, and Krainov (ADK) model [15]. We neglect the harmonics generated from ions and electrons as well as the influence of the wave mixing process involving harmonic fields. Both assumptions are justified owing to the relatively low conversion efficiency for these high-order processes. For simplifying our calculations, we introduce a uniform neutral medium and scale $D_q(t)\propto I^3(t)$ [16], which will not affect the main results. Here $I(t)$

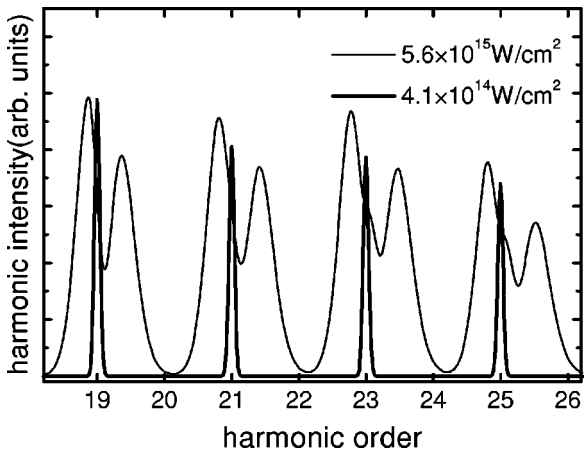

FIG. 2. Calculated on-axis harmonic spectra after propagating through 2.0 mm in ionizing a 50 Torr argon gaseous medium with different driving laser intensities in the one-dimensional limit: (a) 5.6×10^{15} W/cm^2 (thin line) and (b) 4.1×10^{14} W/cm^2 (thick line).

is the time-dependent intensity of the driving field. The laser pulse with a 46-fs FWHM duration is used in all of the following numerical calculations.

The calculated on-axis harmonic spectra in an argon gaseous medium with different peak driving intensities after propagating through 2 mm in 50 Torr argon gas are shown in Fig. 2. In the one-dimensional propagation limit, the spectral splitting is well reproduced when the driving laser intensity is much higher than the saturation intensity of the neutral argon atoms, as well as the spectral broadening and the blueshifting. The propagation of the harmonics from just outside of the target to the detector is not taken into account in our one-dimensional model. The diffraction of the harmonic signals by the slits is also neglected. These assumptions are justified since the refractive indexes of the harmonics in the soft-x-ray regime under 10^{-4} Torr pressure are almost the unit in the ideal vacuum, and the widths of the slits in the spectrometer are three orders larger than the wavelengths of the harmonics in our interests.

The calculated electric fields of the 19th harmonic with the driving intensity of 5.6×10^{15} W/cm^2 are shown in Fig. 3. Due to the rapid ionization of the medium, harmonic emission terminates long before the advent of the peak of laser pulse. The temporal evolution of the 19th harmonic field after propagating through 2 mm in the ionizing medium [Fig. 3(b)], obtained by Fourier transforming the 19th harmonic spectrum shown in Fig. 2, is very different from that of the single-atom response [Fig. 3(a)]. The one pulse of the harmonic field is split into two main pulses after propagating through 2 mm. It seems that there was a *time gate* between the two splitting pulses. It is very similar to the case in Ref. [8] where Altucci *et al.* allowed harmonic emission twice in one driving pulse by controlling the ellipticity of the driving pulses with the help of a birefringent quartz plate. But there is a remarkable difference between these two cases. While the time gate is artificially generated by mechanical means in the case of Ref. [8], the appearance of the time gate in the present case is completely owing to the propagation effects of the harmonic fields in the ionizing medium.

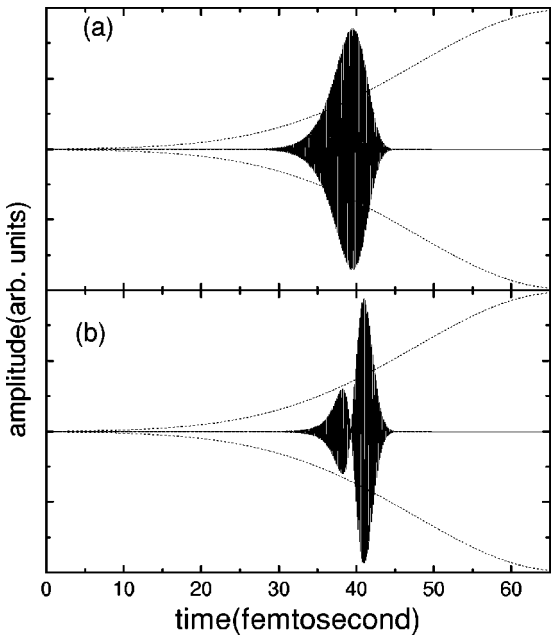

FIG. 3. Electric-field profiles of the 19th harmonic with the driving intensity of 5.6×10^{15} W/cm^2: (a) single-atom response (no propagation taken into account); (b) assembly response (after propagating through 2 mm in the 50 Torr ionizing argon gaseous medium), obtained by Fourier transforming the 19th harmonic spectrum shown in Fig. 2. The temporal envelope of the driving pulse is denoted with the dotted lines. The electric fields are normalized for the sake of clarity.

The rapid ionization of the medium induces that the refractive indexes of the harmonics in the medium are strongly time dependent. Therefore, the generated high harmonics are chirped during their propagation through the ionizing medium owing to the strongly time-dependent refractive indexes, resulting in the spectral splitting and the time gate. The multiple peak structures in high harmonics reported in Ref. [7] were also predicted in our one-dimensional model. According to our one-dimensional calculations, the multiple-peak splitting emerges when the driving intensity is pushed to a higher level and/or when the harmonics propagate a longer distance.

Generally, the phase matching between the fundamental field and its harmonics plays a very important role in the overall emission. Surrounding the focus is a relatively large-volume region where the laser intensity is sufficient to excite HHG in our case. It is self-evident that this is not taken into account in our one-dimensional calculations. In spite of this incompleteness, our one-dimensional simulation is still acceptable owing to the following considerations. On one hand, the gas jet is located 3 mm after the laser focal point, where the laser has begun to propagate with the 0.1-rad divergence angle. The entrance slit of the spectrometer is 100 μm, and the corresponding reception angle is as small as 2×10^{-4} rad. Therefore the harmonic spectra observed in our case are mainly derived from the focus regime. On the other hand, all of the harmonics we can clearly observe exhibit the double-peak structures at the same time. There is little possibility that these structures are aroused by the

phase-matching effects, for it is very difficult in fact to synchronistically implement the phase matching of so many different harmonics, especially when no additional operation is undertaken.

IV. CONCLUSION

In a rapidly ionizing medium, the fast temporal change in the electron density and the resulted variation in refractive indexes are well known to induce spectral blueshifting and broadening. However, the spectral splitting of high harmonics due to the propagation effects has rarely been reported. Using the 46-fs Ti:sapphire laser, we clearly observed the double-peak spectral splitting in high harmonics generated in the argon gaseous medium when pushing the driving field well above the ionization threshold. This splitting is mainly attributed to the propagation effects of the high harmonics in the ionizing medium. In other words, this splitting originates from the fact that the rapid ionization of the medium induces the strongly-time-dependent refractive indexes and electron density, chirping the harmonic pulses in the process of their propagation through the ionizing medium. It is shown that the propagation effects could significantly modify the spectral properties of the harmonics when the intensity of the driving laser pulses is much higher than the saturation intensity of the medium. All calculations in the one-dimensional limit are shown to be in good agreement with experimental measurements.

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