Phase-matched high-order harmonic generation in a gas cell with midinfrared femtosecond pulses

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We experimentally investigate the generation of high-order harmonics in a 4-mm-long gas cell using midinfrared femtosecond pulses at various wavelengths of 1240 nm, 1500 nm, and 1800 nm. It is observed that the yield and cutoff energy of the generated high-order harmonics critically depend on focal position, gas pressure, and size of the input beam which can be controlled by an aperture placed in front of the focal lens. By optimizing the experimental parameters, we achieve a cutoff energy at ~190 eV with the 1500 nm driving pulses, which is the highest for the three wavelengths chosen in our experiment.

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I. INTRODUCTION

As a potential table-top coherent extreme ultraviolet (euv) light source, high-order harmonic generation (HHG) has been attracting significant attention since it was discovered nearly two decades ago [1]. In most cases, high-order harmonics are generated with regeneratively amplified Ti:sapphire laser systems based on chirped pulse amplification at ~800 nm wavelength, which can offer multimillijoule femtosecond laser pulses at multi-kHz repetition rates. However, it has been realized that the use of longer wavelengths (e.g., midinfrared wavelengths, 1 $\mu m < \lambda < 5$ μm) will bring several important benefits for HHG such as extended cutoff energy and reduced chirp on harmonics, which can both result in shorter attosecond pulses [2–4]. Recently, the rapid development of ultrafast laser technology has enabled construction and commercialization of high-power femtosecond optical parametric amplifiers (OPAs), which are able to offer millijoule level pulses at midinfrared wavelengths [3–7]. Previously, by use of midinfrared pulses at wavelengths of 1.5 and 2 μm, harmonic cutoff energies had been extended to ~160 eV [3] and ~220 eV [4], respectively. Even more recently, fine interference fringes formed in high-order harmonic spectra were also observed with midinfrared driving pulses at 1240 nm wavelength [5], indicating the existence of new physical mechanisms for HHG driven by midinfrared pulses.

In this work, we carry out the HHG experiment in a 4-mm-long gas cell using midinfrared femtosecond pulses at various wavelengths of 1240 nm, 1500 nm, and 1800 nm. The use of the gas cell offers a longer interaction length in comparison with a gas nozzle, which could facilitate an enhanced phase matching. On the other hand, as compared with a hollow waveguide [8,9], the use of the gas cell makes it possible to employ a tight focusing geometry in order to achieve sufficiently high peak intensity at a limited pulse energy (e.g., 0.5 mJ) of the midinfrared beam from an optical parametric amplifier (OPA). We found that the harmonic yield and cutoff energy are sensitive to a series of experimental parameters, such as the focal position in the gas cell, the gas pressure, and the beam size of the input pulses (controllable by adjusting the size of an annular aperture placed in front of the focal lens). By optimizing these parameters, we demonstrate that a HHG cutoff energy of ~190 eV in argon can be achieved with the 1500 nm driving pulses.

II. EXPERIMENT

The experimental setup is shown in Fig. 1. Wavelength-tunable midinfrared laser pulses are generated by an optical parametric amplifier (OPA, TOPAS-C, Light Conversion,

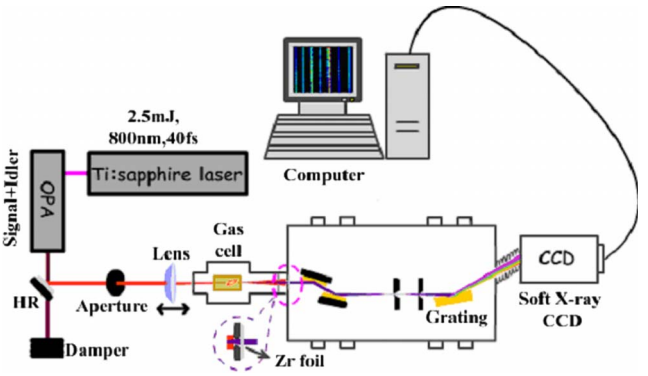

FIG. 1. (Color online) Schematic of experimental setup for generation of high-order harmonics with midinfrared driving pulses.

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Inc.) pumped by a commercial Ti:sapphire laser system (Legend, Coherent, Inc.). The Ti:sapphire laser, operated at a repetition rate of 1 kHz, provides ~40 fs [full width at half maximum (FWHM)] laser pulses with a central wavelength at ~795 nm and a single pulse energy of ~2.5 mJ. At the 1240 and 1500 nm wavelengths, the maximum pulse energy that the OPA can offer is ~0.5 mJ, whereas at the 1800 nm wavelength, the maximum pulse energy slightly decreases to ~0.4 mJ. The measured M^2 values of the beams at 1240, 1500, and 1800 nm wavelengths are ~1.3, ~1.2, and ~2.5, respectively. For achieving optimized HHG spectra, we carefully adjust the input pulse energy as well as the beam size using a variable aperture placed in front of the focal lens; therefore, there are no other attenuators used in this experiment. To select the midinfrared pulses with the wavelengths we need, dielectric coated broadband mirrors with high reflection coatings are used. A fused silica lens with a focal length of ~15 cm at 1500 nm wavelength (in this case, the focal length is determined after accounting for the 3-mm-thick window of the vacuum chamber) is used to focus the infrared beams. The lens is fixed on a high precision translation stage by which the lens can be freely shifted along the beam propagation direction. The back pressure inside the vacuum chamber is ~0.1 Pa. A 4-mm-long gas cell filled with argon gas is mounted inside a vacuum chamber for generating high-order harmonics, and variable gas pressures in a range from 10 to 40 mbar are used in this experiment. A flat-field grating spectrometer equipped with a soft x-ray charge coupled device (CCD, Princeton Instruments, 1340×400 imaging array PI:SX 400) is used to measure the HHG spectra. In order to block the low-order harmonics and the residual infrared driving pulses, we place a 500-nm-thick zirconium foil at the entrance of the spectrometer. In most of our experiments, an integration time (3×10^5 shots) of five minutes is sufficient for recording a single HHG spectrum. For some experiments performed with unoptimized parameters, HHG signals are much weaker; therefore, longer integrated times are employed.

III. HHG WITH MIDINFRARED DRIVING PULSES AT DIFFERENT WAVELENGTHS

Typical high-order harmonic spectra obtained by use of midinfrared driving pulses with wavelengths at 1240, 1500, and 1800 nm are presented in Figs. 2(a)–2(c). The pulse durations, all measured by a homemade single-shot autocorrelator (SSA), are ~30 fs for both the 1240 and 1500 nm pulses and ~90 fs for the 1800 nm ones. Figure 2(a) shows the harmonic spectrum generated by the 1240 nm driving pulses with an integration time of 5 min. In this case, the highest cutoff energy, produced at a gas pressure of ~28.4 mbar and a pump power of ~320 mW, is observed to be ~150 eV.

As it has been described by the simple man's model [10–12], the maximum photon energy attainable in HHG can be calculated by the following equation:

$$E_{\text{cutoff}} = \hbar\omega_{\max} = I_p + 3.17U_p, \tag{1}$$

where

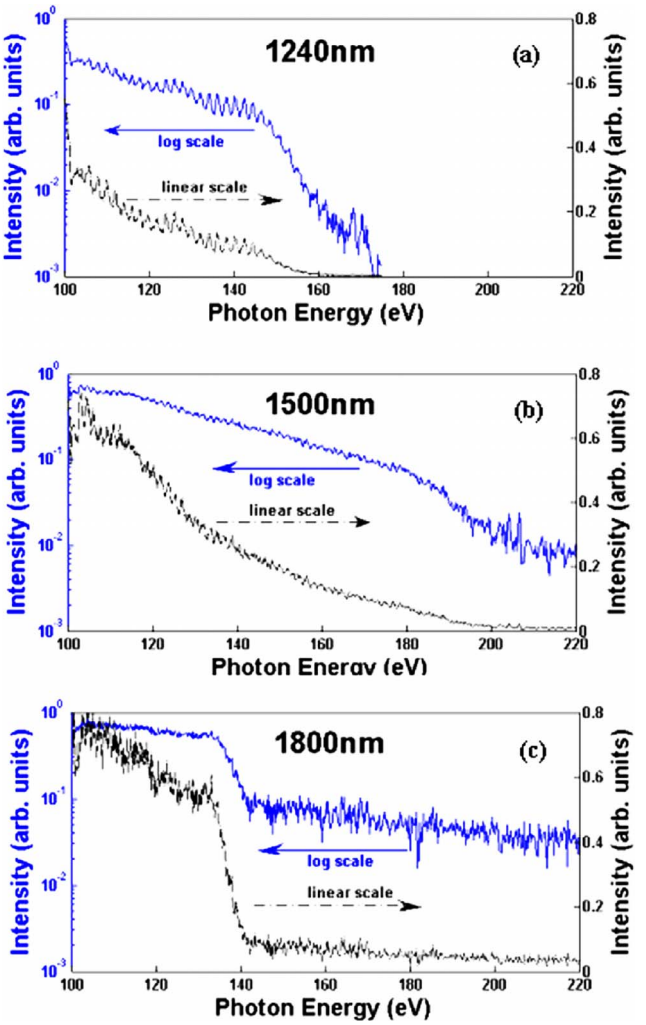

FIG. 2. (Color online) Normalized HHG spectra measured at different wavelengths of (a) ~1240 nm, (b) ~1500 nm, and (c) ~1800 nm.

$$U_p = \frac{e^2 E_L^2}{4 m_e \omega_L^2} \propto I\lambda^2. \tag{2}$$

Here e and m_e are the electron charge and mass, and E_L is the electric field of driving pulses with an angular frequency ω_L (or a wavelength λ). It is clearly indicated by Eqs. (1) and (2) that the use of long wavelength driving pulses will significantly extend the cutoff energy of HHG because the quiver energy of an electron in a light field is proportional to the square of the wavelength. Our result in Fig. 2(a) has proved this prediction since the HHG spectra produced in argon gas with traditional 800 nm driving pulses usually show a cutoff energy significantly less than 100 eV.

In order to further extend the cutoff energy, we tune the wavelength of driving pulses to ~1500 nm. It has to be noticed that in an ideal case, the radius of a focal spot will be proportional to the wavelength of the beam given that the input beam size remains the same; therefore, the laser intensity at the focus will be reduced for the 1500 nm driving pulses as compared to that of the 1240 nm driving pulses

because of the enlarged focal area. For this reason, we use a 2× telescope to expand the input beam size of the 1500 nm pulses in order to create a sufficiently high focal intensity, as shown in Fig. 1. The optimal HHG spectrum with the 1500 nm driving pulses is shown in Fig. 2(b). Clearly, the cutoff energy is indeed further extended to ~190 eV. In this case, a gas pressure of ~22.3 mbar is chosen and the mid-infrared beam is focused exactly at the entrance of the gas cell. A calculation using Eqs. (1) and (2) indicates that this cutoff energy corresponds to a focal intensity of ~2.5 $\times 10^{14}$ W/cm^2 at the 1500 nm driving wavelength.

With the intention of pushing the cutoff energy even higher, we then tune the wavelength from ~1500 nm to ~1800 nm. The maximum pump power is measured to be ~400 mW at this wavelength, which is already in the idler region. As shown in Fig. 2(c), a maximum cutoff energy at ~130 eV can be obtained by carefully optimizing the pump power to ~269 mW, the gas pressure to ~40 mbar, and the focal position to ~1 mm after the entrance of the gas cell. The decrease of the HHG cutoff energy at the increased pump wavelength could be attributed to several possible factors. First, it is observed in our experiment that the idler-beam quality is generally worse than the signal-beam quality, which will result in a degraded focus performance. Second, the longer wavelength of 1800 nm pulses will create a focal area that is ~1.5 times larger than that of the 1500 nm pulses even for a same input beam size and quality. Consequently, the peak intensity will be significantly reduced for the 1800 nm pulses. Third, the pulse duration of 1800 nm pulses is measured to be ~90 fs using the homemade SSA, which is approximately three times the duration of the 1500 nm wave. For this reason, the peak intensity at the focus will further decrease as compared to that of the 1240 or 1500 nm pulses whose pulse duration is only ~30 fs. All of these factors can contribute to the decreased cutoff energy as shown in Fig. 2(c). It should be mentioned that a longer integration time of ~30 min has to be used for recording the HHG spectrum in Fig. 2(c) because of the weak HHG signal, which could be caused not only by the lower focal intensity mentioned above but also by the wavelength scaling law of the HHG yield reported in Ref. [2]. Typically, the spatial beam profile of the high-order harmonics (integrated over the spectral range of 120–190 eV) generated with the pump pulses at 1500 nm wavelength shows a divergence half-angle of ~1.2 mrad.

Since we have observed the highest cutoff energy with the 1500 nm driving pulses, we then systematically investigate the influence of the aperture size, the gas pressure, and the focal position on the HHG at this wavelength. These parameters are critical to the harmonic yield as well as the cutoff energy as we found in the experiment.

IV. ROLES OF FOCAL POSITION, ANNULAR APERTURE, AND GAS PRESSURE

For systematically investigating the dependence of HHG on experimental parameters, we measure the HHG spectra by gradually changing one of the three parameters while the other two parameters are fixed at the optimal values. In this manner, three groups of HHG spectra are obtained and presented below in Figs. 3–5.

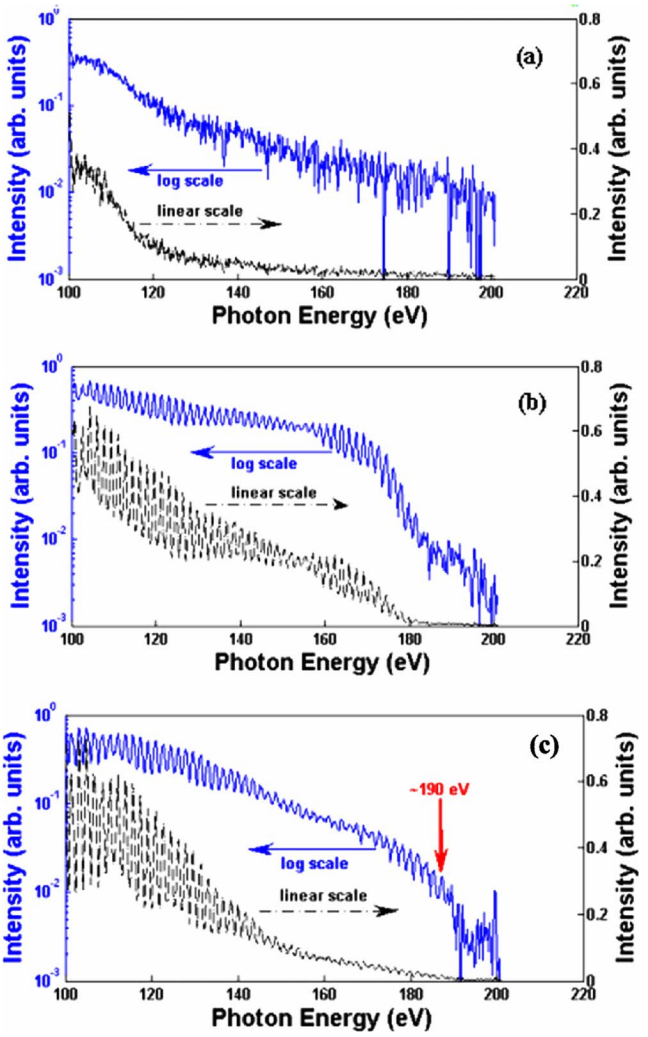

FIG. 3. (Color online) Normalized HHG spectra measured with the fixed pump power of ~310 mW and gas pressure of ~22.3 mbar when the driving pulses are focused (a) 3 mm and (b) 1 mm after the entrance of the gas cell, and (c) 1 mm before the entrance of the gas cell.

Figure 3 shows the high-order harmonic spectra observed by changing the focal position in the gas cell when the pump power is fixed at ~310 mW and the argon gas pressure is fixed at ~22.3 mbar. The three HHG spectra are recorded when the focal spot position is 3 mm after the gas cell entrance [Fig. 3(a)], 1 mm after the gas cell entrance [Fig. 3(b)], and 1 mm before the gas cell entrance [Fig. 3(c)]. It can be seen that the spectrum in Fig. 3(b) exhibits the highest photon energy up to ~190 eV, whereas the cutoff energy quickly drops to ~150 eV in Fig. 3(a). We speculate that the sharp decrease of cutoff energy could be attributed to the fact that the HHG spectrum shown in Fig. 3(a) is obtained when the focal position is located after the center of the gas cell. It has been pointed out in Ref. [13] that in order to generate intense harmonics, the laser focus should be placed before the nonlinear medium so that the intensity of the driving pulse is decreasing during the HHG process. In such a case, the phase mismatching induced by the Gouy phase shift [14] could be mitigated or even fully compensated by the phase

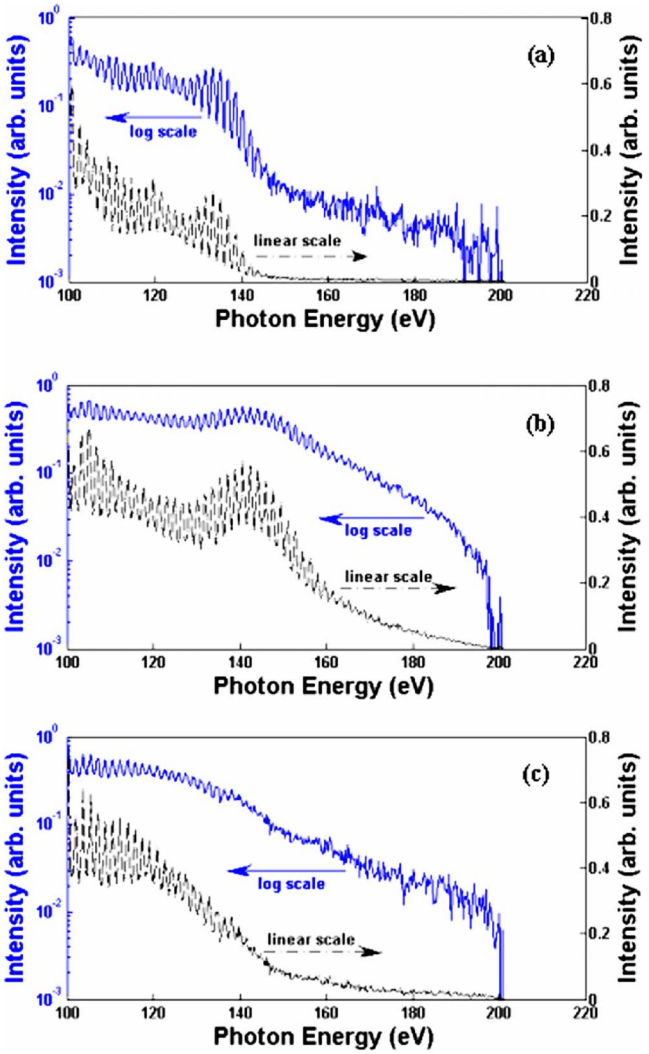

FIG. 4. (Color online) Normalized HHG spectra measured at various pump powers of (a) ~250 mW, (b) ~350 mW, and (c) ~400 mW when the driving pulses are focused at the entrance of gas cell and the pressure is set at ~22.3 mbar.

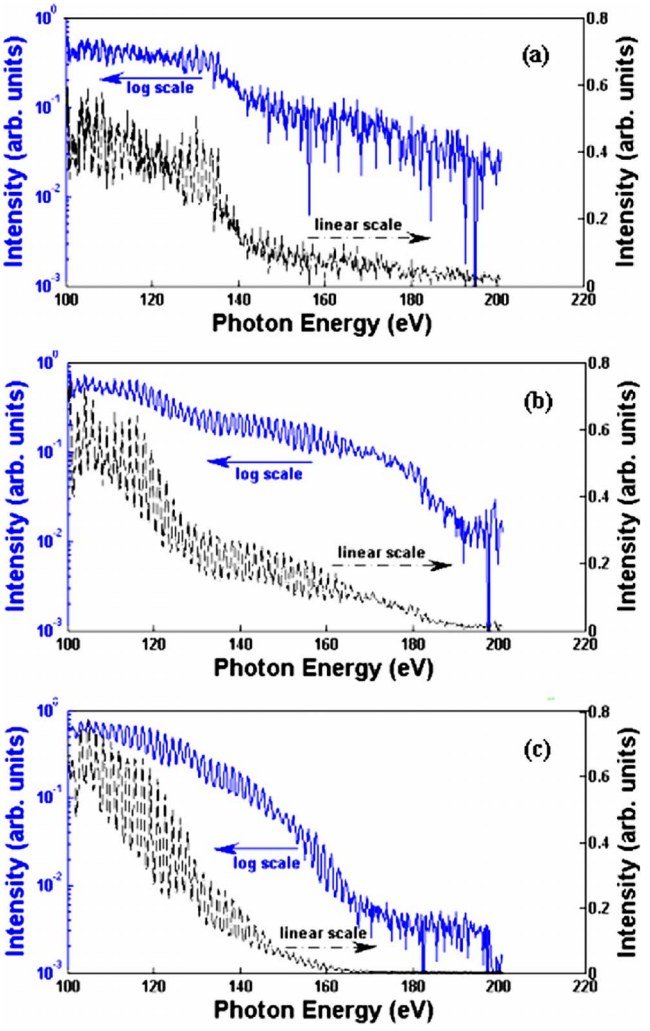

FIG. 5. (Color online) Normalized HHG spectra measured at different gas pressures of (a) ~10 mbar, (b) ~15.5 mbar, and (c) ~30 mbar. The pump power is fixed at ~310 mW and the driving pulses are focused at the entrance of the gas cell in (a) and (b), whereas the pump power is reduced to ~295 mW and the driving pulses are focused 1.5 mm before the entrance of the gas cell in (c).

mismatching induced by a rapidly varying dipole phase as a function of laser intensity. Since the dipole phase for a certain electron trajectory (e.g., short or long trajectory) can be written as $\phi_{\mathrm{dip}} \propto -U_p/\omega_1$ with U_p being the quiver energy of an electron in the light field and ω_1 the frequency of the driving pulse [15], it is clear that a driving pulse with a longer wavelength will lead to a larger dipole phase variation as compared to a driving pulse with 800 nm wavelength, given that the two pulses have the same pulse duration. This property indicates that an optimal phase-matching condition could be more easily fulfilled for infrared driving pulses as compared to traditional 800 nm driving pulses with a similar pulse duration [16]. For this reason, we observe that the cutoff energy can be significantly improved when the laser focus is positioned before the center of the gas cell, as shown in Figs. 3(b) and 3(c). In these cases, even for two focal positions separated by 2 mm, the HHG spectra appear to have a similar cutoff energy in Figs. 3(b) and 3(c). A cutoff energy of ~190 eV, which is defined by the photon energy of the highest-order harmonic, which can still be clearly identified on the HHG spectrum, is indicated in Fig. 3(c) by an arrow. When the laser focus is further shifted upstream away from the gas cell, the HHG conversion efficiency and cutoff energy both decrease rapidly, most likely caused by the rapid decrease of the laser peak intensity beyond the Rayleigh range.

Next, by fixing the gas pressure at ~22.3 mbar and the laser focus at the entrance of the gas cell, we record the HHG spectra as shown in Figs. 4(a)–4(c) at various pump powers of ~250, ~350, and ~400 mW merely by adjusting the aperture size. It is apparent that the cutoff energy first extends with the increasing pulse energy until reaching a maximum value of ~180 eV as shown in Fig. 4(b), and after that the cutoff energy starts to decrease. A cutoff energy is measured to be ~150 eV when the pump power is further raised to ~400 mW, as indicated by Fig. 4(c). It is noteworthy that the aperture plays a role more than the mere attenuation of

the pump energy since the cutoff energy does not increase monotonically with the increasing pump energy. When the aperture size is increased, it may induce a stronger plasma generation due to the enhanced laser intensity, as well as a faster variation of the Gouy phase near the focus due to the shortened Rayleigh length [17]. Both these effects could spoil the phase matching between the driving pulses and high harmonics, leading to the reduction of cutoff energy and conversion efficiency of the HHG when the aperture is fully open [18].

Last, the HHG spectra obtained at different gas pressures are compared in Fig. 5. The HHG spectra shown in Figs. 5(a) and 5(b) are measured when the pump power is set at ~310 mW and the focal position is fixed at the entrance of the gas cell. At a low gas pressure of ~10 mbar, the HHG signal is relatively weak and the cutoff energy is only ~130 eV, as indicated in Fig. 5(a). The low conversion efficiency should be a result of the low density of the nonlinear medium, and the reduced cutoff energy could be attributed to an unfavorable phase matching. When the gas pressure is slightly increased to ~15.5 mbar, the cutoff energy is dramatically extended to ~180 eV as shown Fig. 5(b). Further increase of the gas pressure causes the reduction of both the cutoff energy and HHG yield. For example, when the gas pressure is chosen as ~30 mbar, an optimized HHG spectrum showing a cutoff energy of ~160 eV is recorded with a pump power of ~295 mW and a focal position ~1.5 mm before the entrance of the gas cell, as shown in Fig. 5(c). Based on the estimated focal intensity of the 1500 nm wavelength beam, which is $\sim 2.5 \times 10^{14}$ W/cm^2 and the measured pulse duration of ~30 fs, the ionization ratio can be calculated to be ~15% using the Ammosov-Delone-Krainov tunneling model [19]. Therefore, it is likely that at the high pressure of argon gas, the plasma density could be too high to support a good phase matching between the driving pulse and the high-order harmonics in the cutoff region.

V. CONCLUSIONS

To conclude, we have shown that high-order harmonics up to ~190 eV can be achieved in a 4-mm-long gas cell filled with argon gas with infrared ultrashort driving pulses. It is observed that both the yield and the cutoff energy of the HHG are very sensitive to a series of experimental conditions, such as the gas pressure, focal position of the laser beam, and the aperture size, indicating that the phase-matching mechanism plays an important role. For a femtosecond OPA with a limited output energy of ~0.5 mJ/pulse, the best wavelength for efficiently extending the HHG cutoff energy seems to be around 1500 nm, which is almost the longest wavelength in the signal-beam region for an OPA pumped by an 800 nm femtosecond source. Longer wavelengths up to ~2400 nm are attainable for the idler pulses, which in principle should be able to further extend the cutoff energy according to the wavelength scaling law of HHG cutoff energy. However, practically speaking, the output energy, temporal duration, and beam quality of the idler pulses are all significantly degraded as compared to those of the signal pulses, leading to the unexpected reduction of cutoff energy at the 1800 nm driving wavelength as we observe in the experiment.

With the aim of extending HHG cutoff energy up to the water window (2.2–4.4 nm) or even the keV x-ray region using infrared driving pulses, higher pulse energy is desirable. Currently, due to the limited pulse energy from most femtosecond OPA sources, almost all of the HHG experiments with the infrared driving pulses are carried out with low ionization potential gases including xenon, krypton, and argon [3,20,21]. The combination of a higher pulse energy and a longer driving wavelength would allow for efficient HHG in a gas with a high ionization potential such as neon or helium at a low plasma density, thereby extending the cutoff energy [22,23]. In addition, with a high driving pulse energy, it is also possible to employ a loose focusing geometry to create a relatively flat phase front and extend the interaction length, which both could boost the yield of HHG [17]. For these reasons, the future of HHG driven by infrared driving pulses will critically rely on the development of high peak power infrared femtosecond laser sources [24,25].

Note added. Recently, we became aware that a coherent water window x ray was generated by phase-matched high-order harmonic generation using midinfrared driving pulses at 1.6 μm wavelength [23].

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