A tunable XUV monochromatic light source based on the time preserving grating selection of high-order harmonic generation*

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We accomplish a laboratory facility for producing a femtosecond XUV coherent monochromatic radiation with a broad tunable spectral range of 20 eV–75 eV. It is based on spectral selected single-order harmonics from intense laser driven high harmonic generation in gas phase. The time preserving for the selected harmonic radiation is achieved by a Czerny–Turner type monochromator designed with a conical diffraction grating mount for minimizing the time broadening caused by grating diffraction and keeping a relatively high diffraction efficiency. Our measurement shows that the photon flux of the 23-order harmonic (H23) centered at 35.7 eV is 1×10^9 photons/s approximately with a resolving power $E/\Delta E \approx 36$. This source provides an ultrashort tunable monochromatic XUV beam for ultrafast studies of electronic and structural dynamics in a large variety of matters.

Keywords: high harmonic generation, conical diffraction, time preserving monochromator, femtosecond extreme ultraviolet pulse

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1. Introduction

Femtosecond laser technology based on Ti:Sapphire crystal has enabled us to follow atomic and molecular motion in real time[1] and then to control chemical reactions[2] and molecular processes.[3] However, the experiments with femtosecond time resolution in extreme ultraviolet (XUV) or soft x-ray region have only recently been made possible, partly owing to femtosecond-slicing techniques at synchrotron radiation,[4] partly because of the realization of x-ray free electron lasers,[5] and primarily due to the development of high harmonic generation (HHG) in intense laser fields during the last decades.[6–9] HHG occurs when an electron, liberated from an atom or molecule by incident intense laser fields, gains energy from the field and recombines with the parent ion.[10] The HHG spectrum features a sequence of peaks corresponding to the odd harmonics of the driven laser wavelength with an intensity distribution characterized by a plateau spanning into XUV or soft x-ray region. Thus, HHG radiation can be used as a short-wavelength radiation source for studies of electronic structure and dynamics of atoms, molecules, and condensed matters,[11,12] with a table-top scale, opposed to large synchrotron and free-electron-laser facilities. For many spectroscopic applications, the selection of a single harmonic radiation from consecutive multiple harmonics is necessary with various optical methods,[3,13–16] such as multilayer mirrors, gratings with grazing incidence, and selected enhancement by laser pulse shaping. Particularly, by using the gratings, a good spectral resolution with a broad tunable spectral range has been achieved. However, it has been demonstrated that the harmonic selection using a diffraction grating could significantly stretch the radiation pulse in the time domain[17,18] even that the stretching can be limited by reducing the incident angle but with the cost of grating reflectivity.[19,20] To overcome this shortcoming, Poletto *et al.*[21] have demonstrated a time-preserving monochromator for XUV and soft x-ray regions in which a single diffraction grating is operated in a conical diffraction. Due to its super temporal properties and relatively high efficiency, this conical diffraction grating monochromator has been employed in the spectral selection of HHG radiation.[16,22–24] In this paper, we describe an accomplishment of a tunable ultrafast XUV coherent source with a resolving power ($E/\Delta E \approx 36$) and an ultrashort pulse duration ($\sim$ 100 fs) in a broad tunable range (20 eV–75 eV), based on a design of time-preserving monochromator for intense laser driven HHG radiation in atomic gas Ar or Ne. These properties make it attractive for ultrafast electronic and structural dynamics in a variety of matters.

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2. Time preserving selection of HHG radiation

We design a conical diffraction grating monochromator for selecting the HHG radiation produced in the gaseous atom (e.g., Ar or Ne) by a 35 fs, 800 nm pulsed laser. As depicted in Fig. 1(a), the monochromator is a Czerny–Turner type with a pair of toroidal gold coated mirrors with the focal length $f = 500$ mm and a plane grating operated in conical diffraction, in which the incident and diffracted wave vectors are almost parallel to the grating grooves (different from the classical diffraction mount, that is, perpendicular to the groove). During the operation, the toroidal mirrors can collimate the incident light from the source and focus the diffracted light after grating on the exit slit at a 3° incident angle. With the equal focal length of the mirrors, the magnification of the monochromator is unity, minimizing the aberrations. For selecting the wavelength, the grating is rotated around an axis that is tangent to the surface and parallel to the grooves. The instrument is operated without the entrance slit since the size of the XUV source is in the range of few tens of micrometers. We use the plane gratings (Newport) mounted on a vacuum motorized linear stage (Physik Instrumente VT-80) for selecting a different spectral region which is mounted on a vacuum motorized rotation stage (Physik Instrumente PRS-110) for grating rotation.

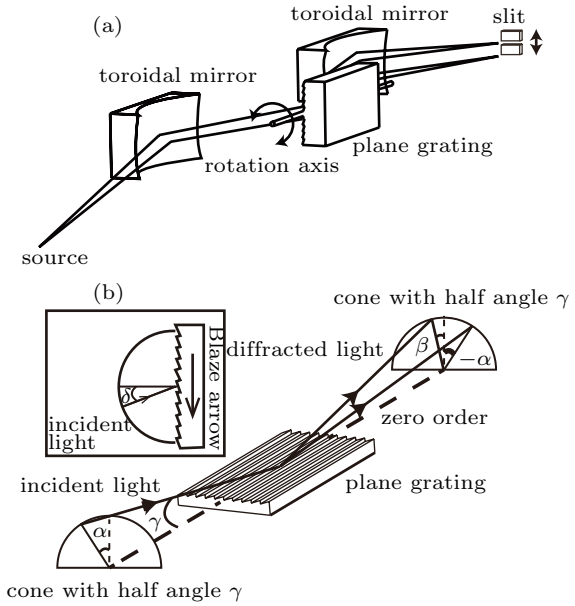

Fig. 1. (a) The design of the monochromator. Wavelength scanning is performed by rotating the grating around the axis that is tangent to the surface and is parallel to the grooves. (b) Geometry of the conical diffraction mount. The inset shows blaze condition of maximum efficiency where δ is the blaze angle.

As shown in Fig. 1(b), in this optical scheme, the direction of the incoming ray is defined by the altitude angle γ, the angle between the direction of the incoming ray and the direction of the grooves, and the azimuth angle α which is zero if they lie in the plane perpendicular to the grating surface and parallel to the rulings. Thus, under this definition, γ also gives the half-angle of the cone into which the light is diffracted, and $-\alpha$ is the azimuth of the zero order light. Let β define the azimuth of the diffracted light at wavelength λ and order m, the grating equation now is[25]

$$\sin\gamma(\sin\alpha+\sin\beta) = m\lambda\sigma, \tag{1}$$

where σ is the groove density. With the operation condition of $\alpha = \beta$,[26] the grating equation becomes

$$2\sin\gamma\sin\beta = m\lambda\sigma. \tag{2}$$

The inset of Fig. 1(a) shows the blaze condition ($\alpha = \beta = \delta$) of maximum diffraction efficiency where δ is the blaze angle. According to Eq. (2), the blaze wavelength can be expressed as

$$\lambda_{\mathrm{B}} = \frac{2\sin\gamma\sin\delta}{m\sigma}. \tag{3}$$

In the designed monochromator, we choose two gratings with different groove densities, 300 gr/mm (blaze angle δ =6.5°) and 600 gr/mm (blaze angle δ =5.2°). At the altitude angle γ of 3.5° and the diffraction order $m = 1$, the blaze wavelength can be calculated from Eq. (3), giving a value of 46 nm for 300 gr/mm grating and 18 nm for 600 gr/mm, respectively. The grating can be used in the 2/3∼3/2 range of the blaze wavelength, i.e., the 300 gr/mm grating covers the photon energy 18 ∼ 41 eV and the 600 gr/mm grating the photon energy 45 ∼ 103 eV, respectively, if we take the limitation for the spectral range from the diffraction efficiency larger than 50% of that at blaze wavelength.[16] The grating parameters in the designed monochromator are summarized in Table 1.

Table 1. Grating parameters of the time preserving monochromator.

Spectral region	20–75 eV (13–62 nm)	
Grating	Altitude angle γ	3.5°
G1	Energy region	20–45 eV
	Groove density	300 gr/mm
	Blaze angle δ	6.5°
	Blaze wavelength	27 eV (46 nm)
G2	Energy region	45–75 eV
	Groove density	600 gr/mm
	Blaze angle δ	5.16°
	Blaze wavelength	67.8 eV (18 nm)

The main advantages of conical diffraction mount of the grating are the time preserving for pulsed duration and the high diffraction efficiency. It has been shown in the previous studies[27] that the diffraction efficiency of the grating with the conical diffraction mount is close to the reflectivity of the grating's coating, which is two to three times higher than the classical grating mount. Normally, the grating will inevitably cause a time broadening of the ultrafast pulse because of the diffraction: the total pulse elongation by the grating diffracted by N grooves illuminated by the radiation at wavelength λ is

$$\Delta t = \frac{Nd}{c}(\sin\alpha - \sin\beta) = \frac{Nm\lambda}{c}. \tag{4}$$

Thus, we can calculate the pulse broadening of the grating with the classical diffraction and conical diffraction mounts for our 300 gr/mm and 600 gr/mm gratings. The results indicate that the time broadening of the pulse is inversely proportional to the photon energy, as shown in Fig. 2.

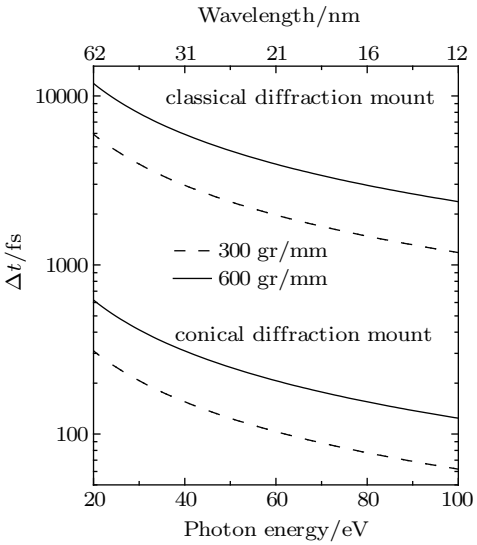

Fig. 2. Calculated time broadening of the classical diffraction mount and conical diffraction mount for two types of grating groove densities, dashed lines for 300 gr/mm and solid lines for 600 gr/mm. In the calculations, the divergence angle is 10 mrad, the focal length is 500 mm, and the incident angle is 87°.

It is clear that the classical diffraction mount brings a pulse spread to few picoseconds, while the conical diffraction mount reduces largely the spread since much less grating grooves are illuminated at the same incident angle which results in less time elongation of XUV pulse duration. For the same reason, the pulse broadening of 600 gr/mm grating (solid line) is higher than that of 300 gr/mm grating (dashed line). The divergence of the XUV beam is expected to be less than 10 mrad,[16] therefore, the duration of the XUV pulse is about 100 fs.

3. Spectral characterization of the selected HHG radiation

We used a Ti:Sapphire laser system to provide 800 nm, 35 fs laser pulses up to 6 mJ with a repetition rate of 1 kHz. The laser beam was focused by a 30 cm focal length lens, giving a focus spot about 50 μm. HHG was generated in a nickel (Ni) gas cell of 3 mm diameter at the laser focus. The holes in the tube wall were burned by the laser beam itself, minimizing the gas load to the surrounding vacuum. The driving laser was blocked after the cell by using a 500-nm-thick Al filter which allows the transmission of HHG radiation. Figure 3(a) shows the HHG intensity distribution of Ar gas (50 Torr) driven by the laser intensity of 5×10^{14} W/cm^2, which is measured at the exit slit (about 50 μm) by scanning the 300 gr/mm grating, with 0.01° rotation each step. It is measured that, at the pressure of 50 Torr, the phase matching of HHG for Ar can be realized for a high photon flux. This HHG photon flux is measured by the current of a calibrated XUV diode (AXUV100G). The responsivity of the diode for the conversion from the amper to flux is 0.26 A/W. The measurement gives a value of 1×10^9 photons/s of the photon flux for the selected 23 order HHG (H23, 35.7 eV) at the exit slit of the monochromator (corresponding to an XUV radiation energy of 5.7 pJ per pulse). From this measurement, we can also estimate the transmission of the designed monochromator that reaches a value of 8% approximately by giving a conversion efficiency of 5×10^{-7} for the Ar HHG irradiated under 1 mJ, 800 nm femtosecond laser beam.

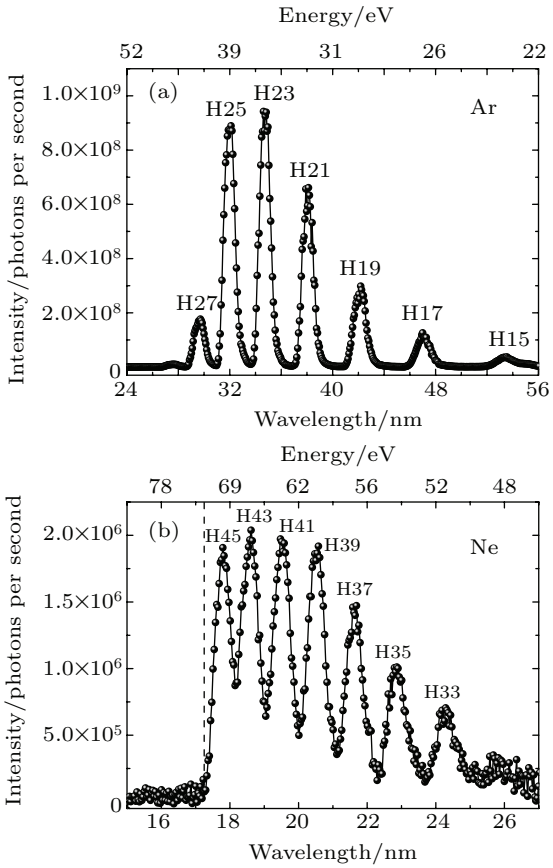

Fig. 3. (a) The harmonic spectrum generated by Ar is recorded by scanning the azimuth angle of the grating with 300 gr/mm. (b) The harmonic spectrum generated by Ne is recorded by scanning the angle of the grating with 600 gr/mm, and the intensity of the single-order HHG radiation is recorded by using a calibrated XUV diode behind the exit slit. The dashed line indicates the absorption edge of Al filter.

For the XUV radiation above 45 eV, we use atomic Ne gas as the generation medium, because of its high HHG cutoff due to the high ionization potential of Ne (21.6 eV), which is higher than that for Ar and at 3 mJ driving laser pulse energy. The high driving laser power is crucial for ionizing Ne to generate a high ponderomotive energy of the electrons. Figure 3(b) shows the HHG intensity distribution of Ne gas (300 Torr), and there is a plateau of harmonic spectrum up to

the aluminum absorption edge (72 eV). The measured photon flux for H43 (66.7 eV) is 2×10^6 photons/s. Moreover, a higher pressure for Ne is needed to achieve phase matching in optimizing the HHG radiation because the gas dispersion of Ne is lower than that of Ar.

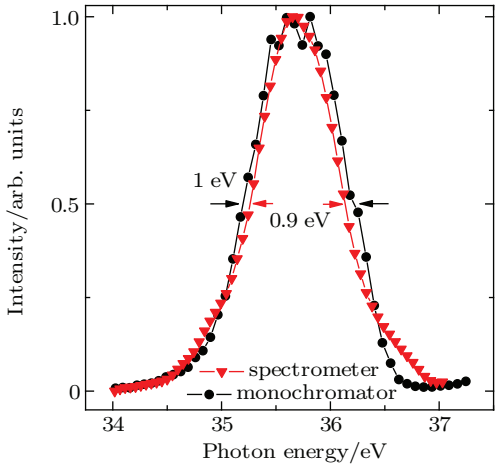

Fig. 4. (color online) The measured spectral profile for the H23 harmonic radiation. The central photon energy is 35.7 eV and the FWHM is 1 eV, as indicated by the black arrows. The red line shows the spectral profile of H23 from the home-built flat field spectrometer in the same experiment condition and the FWHM is 0.9 eV, as denoted by the red arrows.

For a XUV or soft-x ray radiation source, monochromatic feature is important in many applications. We calculated the spectral resolution of the designed monochromator and measured the spectral profiles of the selected single HHG radiation. Figure 4 presents the measured spectral profile of the H23 harmonic radiation (35.7 eV) from a 50 μm exit slit of the monochromator (the black line). For comparison, the H23 harmonic spectrum measured by a home-built flat-field XUV spectrometer in the same experimental condition is also given in the figure (the red line). The spectral profiles obtained by the monochromator and spectrometer are almost consistent with each other and show a measured spectral width (the full width at half maximum, FWHM) of 1.0 eV roughly, which gives an experimental resolving power of 36 for the monochromator. A theoretical resolution ($\Delta\lambda$) of the monochromator can be deduced from the angular dispersion $\mathrm{d}\beta/\mathrm{d}\lambda$ of conical diffraction in Eq. (2), which leads to the linear dispersion $\mathrm{d}s/\mathrm{d}\lambda$ ($\mathrm{d}s$ is the arc length of conical diffraction at the exit slit corresponding to $\mathrm{d}\beta$), that is,

$$\Delta\lambda = \frac{\mathrm{d}\lambda}{\mathrm{d}s}\times w_{\mathrm{slit}} = \frac{\mathrm{d}\lambda}{\mathrm{d}\beta}\frac{\mathrm{d}\beta}{\mathrm{d}s}w_{\mathrm{slit}} = \frac{2\cos\beta}{\sigma m f}w_{\mathrm{slit}}. \quad (5)$$

In the experiment, β is 4.9° for the wavelength of H23 with the grating of $\sigma = 300$ gr/mm, the focal length $f = 500$ mm, and the slit width $w_{\mathrm{slit}} = 50$ μm, and the calculation from Eq. (5) gives a resolution $\Delta\lambda = 0.7$ nm or a resolving power $\lambda/\Delta\lambda = 49$. Therefore, our measured value is reasonable as the theoretical value which is little better than the measured value. This can be confirmed by a further simulation by the ray tracing for the monochromator using SHADOW code.[28] The results are shown in Fig. 5 at 50 μm exit slit. The photon energies of rays are 35.7 eV and 36.4 eV, respectively. The ray tracing simulation gives a resolving power $\lambda/\Delta\lambda = 51$.

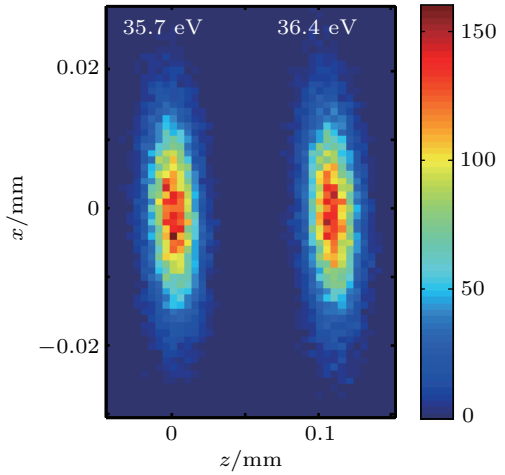

Fig. 5. (color online) Ray tracings at the exit slit plane using SHADOW code. The photon energies of rays are 35.7 eV, 36.4 eV, respectively. The z direction is the dispersion direction and the x is the exit slit direction.

A small value of the measured resolution for the monochromator, compared to the theoretical and ray tracing values, results from several experimental factors, such as the aberration of the toroidal mirror, the size of HHG generating spot, etc.

4. Conclusions

In conclusion, we accomplish a laboratory facility for producing femtosecond XUV coherent monochromatic radiation with a broad tunable range of 20–75 eV, based on spectrally selected HHG driven by ultrafast intense laser fields in atomic gases. By designing a Czerny–Turner type monochromator with the conical diffraction grating mount, a time preserving for the selected harmonic radiation is achieved, which minimizes the time broadening from grating diffraction and keeps a relatively high diffraction efficiency. This radiation source provides an ultrashort monochromatic XUV radiation with a tunability in a broad energy region and can be useful for further studies in ultrafast electronic and structural dynamics in a large variety of atoms, molecules, and condensed matters.

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