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Efficient low-density grating setup for monochromatization of XUV ultrafast light sources

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Abstract: Ultrafast light sources have become an indispensable tool to access and understand transient phenomenon in material science. However, a simple and easy-to-implement method for harmonic selection, with high transmission efficiency and pulse duration conservation, is still a challenge. Here we showcase and compare two approaches for selecting the desired harmonic from a high harmonic generation source while achieving the above goals. The first approach is the combination of extreme ultraviolet spherical mirrors with transmission filters and the second approach uses a normal-incidence spherical grating. Both solutions target time- and angle-resolved photoemission spectroscopy with photon energies in the 10-20 eV range but are relevant for other experimental techniques as well. The two approaches for harmonic selection are characterized in terms of focusing quality, photon flux, and temporal broadening. It is demonstrated that a focusing grating is able to provide much higher transmission as compared to the mirror+filter approach (3.3 times higher for 10.8 eV and 12.9 times higher for 18.1 eV), with only a slight temporal broadening (6.8% increase) and a somewhat larger spot size (∼30% increase). Overall, our study establishes an experimental perspective on the trade-off between a single grating normal incidence monochromator design and the use of filters. As such, it provides a basis for selecting the most appropriate approach in various fields where an easy-to-implement harmonic selection from high harmonic generation is needed.

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1. Introduction

Extreme ultraviolet (XUV) radiation with a narrow bandwidth is one of the key ingredients for many high-resolution spectroscopic techniques used in research today. Beyond this, XUV light also has considerable application value for the industry, for example in lithographic patterning adopted in semiconductor manufacturing processes. With the advent of femtosecond high-power lasers and the progress of high harmonic generation (HHG) techniques, table-top XUV light sources with pulse duration in the femtosecond range have become a reality. These sources offer a more compact and affordable alternative compared to the synchrotron radiation facilities, or free-electron lasers, while simultaneously supporting spectroscopic, imaging or scattering techniques with high temporal resolution. Presently, HHG has become a viable approach to generate bright and ultrafast XUV-light sources, but monochromatization of the generated radiation still remains a challenge – in particular from the point of view of maintaining a high photon transmission through the monochromator, while simultaneously minimizing time broadening of the light pulses, as well as keeping the optical design robust and compact.
HHG is a well-developed technique used to convert long-wavelength laser light into radiation with shorter wavelengths, typically in the XUV or soft X-ray regime [1–3]. The HHG process can be understood conceptually by the three step model [4,5]: firstly the electrons experience the ionization tunneling, followed by a propagation in the electric field from the driving light, and finally recombination with the ionic nuclei, yielding high energy and coherent photons during the process. The HHG spectrum consists of a series of discrete harmonics spaced by twice the driving frequency, $2\omega$, up to a maximum photon energy determined by the single atom cut-off relation,

$$h\nu_{\text{cut-off}} = I_p + 3.17U_p,$$

where $h$ is Planck’s constant, $I_p$ is the ionization potential and $U_p$ is the ponderomotive energy of the electron. This relation implies that the energy cut-off can be increased by using a longer driving wavelength, as it scales with the relation

$$U_p \propto I_L \lambda_L^2,$$

where $I_L$ is the intensity of the driving laser and $\lambda_L$ is the wavelength. On the other hand, the HHG efficiency scales proportional to $\lambda^{-\delta}$, where $\delta$ is in between 5 and 6 [6]. This inherent trade-off means that, in practice, one needs to choose suitable driving conditions based on the light source requirement. In our case, to maximize the photon flux at photon energies up to 25 eV, a cascaded scheme [7], using the third-harmonic of the driving laser, is chosen for HHG in a gas target.

The monochromatization requires a selective response to different frequency components, and gratings are extensively adopted for this purpose. A typical monochromator layout is the X-ray Czerny-Turner (XCT) design [8], which is a single-grating based grazing incidence monochromator (GIM), that provides the most necessary elements needed for the collimation, diffraction and re-focusing. The line density is usually chosen based on the trade-off between efficiency and spectral resolution. With high line-density, very high resolving power and energy resolution have been achieved [9,10]. Another common GIM layout is a spherical grating based configuration, which serves as a compact layout [9,11]. As the counterpart of GIM, the normal incidence monochromator (NIM) has also been employed in synchrotrons to access the low photon energy range and provide high energy resolution [10]. Compared to GIM, NIM is quite advantageous in terms of compact layout, ease of alignment, and good XUV imaging properties.

Stepping into the ultrafast regime in the XUV range, monochromators based on diffractive optics exhibit intrinsic drawbacks, as the induced pulse-front tilt broadens the pulse length. Nevertheless, there have been designs targeting HHG [12–18] and free-electron-laser (FEL) light sources [19–21]. One approach is to use a single-grating, but lowering the line density to reach the design requirements [17,18,22], concerning the pulse length trade-off for a certain spectral linewidth. The double-grating design is an alternative approach that permits complete pulse stretching compensation and ideally provides no extra temporal broadening [12,14,16,21]. Pulses as short as 8 fs have been reported [13] using this design. However, such a configuration reduces the transmission and makes the design and alignment somewhat complicated. The pulse stretching induced by a single grating can, however, be reduced by using the off-plane-mount. In this case the diffraction occurs in the direction perpendicular to the direction of propagation thus minimizing the number of illuminated grooves and, thus decreasing the temporal broadening [15,22,23].

In this work, we compare the performance of two types of NIM configurations, as illustrated in Fig. 1. The mirror+filter approach and the spherical grating monochromator are compared in terms of their focusing properties, photon flux and the introduction of temporal broadening of the HHG pulses. The ultrafast light source is part of an experimental setup for time- and angle-resolved photoemission spectroscopy (tr-ARPES) [24]. For the comparison, we designed a spherical grating NIM specially for 10.8 eV and 18.1 eV photon energies. In this particular case, it is found that the mirror+filter design results in a slightly better focus, 100 µm spot size, while the focusing grating focus is about 30% larger (∼33 µm). The larger spot size could be due to fabrication imperfections of the grating profiles or overcoating. On the other hand, in terms of transmission efficiency, the focusing grating is remarkably effective for both photon energies,
resulting in 3.3 times higher efficiency for 10.8 eV and 12.9 times higher efficiency for 18.1 eV as compared to the mirror+filter solution. Consequently, this enabled us to achieve a photon flux on the level of $10^{11}$ photons/s. As for the temporal response, the mirror+filter combination, which is used as a reference, is assumed to provide negligible temporal broadening. Pump-probe measurements on graphene show an overall temporal resolution of 205.3±7.6 fs for mirror+filter and 219.5±1.9 fs for the spherical grating design. By excluding the temporal contribution from the pump, this corresponds to a temporal broadening of 72.4±27.4 fs. In Table 1, the various performance parameters are listed, to compare the applicability of the grating and filter designs for a specific experimental design.

**Fig. 1.** Schematic drawing of the two types of monochromatization for ultrafast applications. a) The focusing mirror plus the thin film filter. b) The focusing grating. ROC stands for the radius of curvature, $m$ is the diffraction order, $\sigma$ is the line density and $\beta$ is the beam incidence angle. $p$ and $q$ indicate the entrance and exit arms, which are 0.6 m and 2.9 m in our case, respectively. HH stands for high harmonic.

**Table 1. Criteria for two configurations of ultrafast XUV pulse monochromatization.**

<table>
<thead>
<tr>
<th></th>
<th>Mirror+filter</th>
<th>Focusing grating</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Pulse-front tilt</strong></td>
<td>Negligible</td>
<td>Below 100 fs for $h\nu&gt;10$ eV$^a$</td>
</tr>
<tr>
<td><strong>Spectral broadening</strong></td>
<td>Negligible</td>
<td>Negligible</td>
</tr>
<tr>
<td><strong>Efficiency</strong></td>
<td>Moderate. Limited by reflectivity of the mirror and transmission through the thin filter</td>
<td>High. Limited by reflectivity of grating, which is set by the groove depth, groove-to-land ratio and coating material</td>
</tr>
<tr>
<td><strong>Focusing</strong></td>
<td>Determined by source size and spherical aberration from focusing mirror</td>
<td>Broadened compared to spherical mirror. Mainly limited by the fabrication errors</td>
</tr>
<tr>
<td><strong>Costs</strong></td>
<td>High. Requires multiple band pass mirrors as well as thin film filters</td>
<td>Moderate. Single optical element low-line-density grating produced by standard nanofabrication processes</td>
</tr>
</tbody>
</table>

$^a$This is for the experimental design presented in this article. The temporal broadening can be further decreased by lowering the number of illuminated grooves through lower line density, or smaller beam spot size.
2. Monochromatization of an ultrafast light source

For monochromatization of ultrafast light sources, the mirror+filter configuration is preferred in terms of time resolution as it can eliminate the temporal broadening issue, providing a significant advantage compared to monochromators based on gratings. In our setup, the mirror+filter configuration is comprised of a concave normal incidence mirror, and a thin film filter that is used to further suppress unwanted harmonics. The mirror coating method is critical for the harmonic selection, but it is difficult to completely eliminate unwanted harmonics with the coating alone. For this reason, the thin film filter with an appropriate transmission window is necessary. The drawback of this approach is that the performance and working wavelength range are primarily determined by the available thin film filters, and they do not always fit the experimental demands. Since thin film filters need to be as thin as hundred nanometers to maintain reasonable transmission, they are delicate and can easily be damaged mechanically or by the beam. Oxidation and contamination are other issues often experienced in the use of thin film filters.

Here we propose a novel and efficient grating-based NIM that has been designed, fabricated and experimentally compared with the mirror+filter solution. It is in principle a spherical focusing grating, which serves as a single optical element-based NIM, with a low line density of 40 l/mm. The main design criteria is to have as low line density as possible while still being able to separate the harmonics geometrically at the sample position. In the present context, the low resolving power caused by the low line density is not an issue, as the spectral resolution is intrinsically determined by the HHG conditions. The purpose of using the lowest possible line density is to mitigate the pulse-front tilt to a negligible level. Note that the value of 40 l/mm adopted here, is dependent on the particular geometry of the setup and the trade-off between pulse stretching and focal properties. In the present comparison, the two compared solutions have similar geometry and focusing properties so that the performance can be compared in a relatively straightforward manner. Depending on the availability of filters and coatings the two solutions are also more or less difficult to implement for various wavelengths, in the present case we target photon energies (∼10-20 eV). For the spherical grating efficiency, the simulation with a groove depth of about 17 nm, together with the 80 nm SiC coating, shows a 15% overall efficiency centered at 18.1 eV photon energy, and overall, above 10% efficiency in the range from 10 to 20 eV, as presented in Fig. 2(c).

3. Mirror+filter configuration

For the direct comparison of the filter and grating-based monochromator solutions, we focus on two photon energies, 10.8 eV and 18.1 eV, generated by the HHG setup. For the mirror+filter
configuration, and for a photon energy of 10.8 eV, a concave MgF$_2$-coated Al mirror with a radius of curvature (ROC) of 1000 mm is used to focus the diverging high harmonic radiation. The filter in this case is a thin (500 µm) LiF (Eksma Optics) window that blocks photon energies higher than 11 eV. For 18.1 eV photons, the previous mirror and filter were replaced with a concave silicon mirror substrate (ROC=1000 mm) coated with SiC and a 150 nm thick Sn filter (Lebow), respectively. The Sn filter provides a band-pass window for photon energies between 17 eV and 24 eV. For more information on the band-pass windows for different thin film filters, see Supplement 1.

4. Focusing grating configuration

With only a few changes, the experimental setup of the mirror+filter monochromator described above can be turned into a grating-based NIM. The key is to replace the spherical focusing mirror with a spherical grating with the same ROC and instead of thin film filters use an output slit to select the desired diffraction order from the grating and to suppress the background noise caused by unwanted orders. In the following section, we elaborate in detail on the focusing grating part, in particular regarding the design principles, and the fabrication recipes.

4.1. Design principles

To design a NIM using a focusing grating, and to match the specific requirements regarding high efficiency and temporal resolution, several constraints need to be considered. Compared to the mirror+filter solution, one major challenge for designing the focusing grating is to limit temporal broadening of laser pulses. The pulse-front tilt experienced on the grating is proportional to the number of grooves that are illuminated, resulting in a correlation between the line density, $\sigma$, and the illuminated area, $A$, on the grating. The spot size $A$ is given by the HHG divergence and the focusing geometry of the setup and, therefore, in this work, it is regarded as a fixed parameter. The illuminated area on the grating can be modulated by an aperture at the cost of reduced photon flux. Keeping the line density $\sigma$ low can minimize time broadening of the pulses, but a sufficiently large $\sigma$ is required to spatially separate the different diffraction orders of the harmonics at the position of the focus. The groove depth, $d$, and the ratio of the groove and peak widths, $r$, determine the efficiency and the band-pass center of the grating. The choice of a coating, and its thickness, are also factors affecting the final efficiency.

Figure 1(b) depicts the concept of the spherical grating-based monochromator that reflects and refocuses the diverging HHG radiation generated in the gas target, while simultaneously separating the harmonics into different diffraction spots. The desired harmonic can then be selected with a slit or an aperture. For a grating with $N$ grooves illuminated by the incident beam, the resolving power, $R$, is expressed as:

$$ R = \frac{\lambda}{\Delta \lambda} = |m|N = \frac{\Delta t \cdot c}{\lambda} \quad (1) $$

where $m$ is the diffraction order, $\Delta t$ is the total pulse-front tilt, $c$ is the speed of light and $\lambda$ is the radiation wavelength. The expression implies a proportional relation between the resolving power and the induced time broadening for a grating-based monochromator. In our case the spectral linewidth is determined by the HHG conditions, so as a design principle, one should pursue a low resolving power for the minimum temporal broadening, which directly links to the line density $\sigma$. Another constraint for $\sigma$ is the wavelength spacing between the different harmonics, as presented in Fig. 2(a), since there needs to be sufficient spatial separation for an aperture to select the harmonics.

Following the aforementioned design principles, a focusing grating is explicitly designed for a photon energy of 18.1 eV. The chosen line density $\sigma$ is 40 l/mm. With the distance from the grating to the sample in the photoemission chamber being 3 m at the sample position, the first
order diffraction spots for 18.1 eV and 10.8 eV are positioned 8 mm and 14 mm away from the zero-order diffraction maximum, respectively. This separation is sufficient so that an aperture can be used to select the desired harmonics and block the remaining undesired diffraction spots. The 17 nm groove depth $d$ and the 1:1 ratio value of $r$, provide optimal efficiency for the 18.1 eV photon energy. The grating pattern is etched into a silicon mirror substrate with a ROC of 1000 mm, and finally, a SiC coating layer with a thickness of minimum 30 nm was deposited. This minimum coating layer thickness yields the best performance according to calculations (see Supplement 1 Section 1 and Figure S1). Figure 2 presents calculations of key performance parameters for the focusing grating for the photon energy range between 5 eV and 25 eV, using the parameters presented above. Figure 2(a) presents the calculated space-separation of the diffracted harmonics. The distances between the zeroth order and the first diffraction order for specific photon energies, are plotted. The entrance and exit arms are set to 0.6 m and 2.9 m, respectively. The inset shows the results of separation for the HHG conditions and setup geometry in our case. Figure 2(b) shows the pulse-front tilt calculated based on Eq. (1), as a function of photon energy. The footprint size of the HHG beam on the grating, is determined by a knife-edge measurement, where the drain current from a mirror down-stream from the grating is measured while a Ta-foil is used to gradually block the beam in front of the grating. The full width at half maximum (FWHM) of the footprint on the grating is found to be approximately 3.5 mm, which corresponds to a source divergence of 5.8 mrad. The total simulated efficiency of the grating is illustrated in Fig. 2(c). A ray-tracing simulation [25] is performed to numerically evaluate the grating performance. For these simulations, a point source is defined as the HHG source, with a beam waist of 10 µm and a divergence in accordance with the experimentally determined divergence of the HHG beam. The experimentally determined grating parameters such as curvature, line density, groove depth, land-to-groove ratio, as well as coating are also used as input. The simulation is then looped for varied photon energies to obtain the result shown in Fig. 2(c).

4.2. Fabrication recipe

The fabrication of the spherical grating was conducted in a clean-room using standard semiconductor processing steps such as lithography, chemical etching, and sputter deposition of the coating material. The different steps of the manufacturing process are schematically shown in Fig. 3(a). The plano-concave 1 inch silicon mirror substrate (Eksma Optics), is first spin-coated with a photoresist (Microposit, S1813) on the concave surface. A photomask made of chromium is used in the exposure step with a high dose exposure (30 mW/cm$^2$ at 365 nm) from a helium lamp (Karl Suss MJB3). The exposure time was 120 seconds. The substrate was then transferred into a reactive-ion-etching (RIE) system (Oxford instruments) where argon plasma milling for $\sim$6 min was used to etch the grating pattern and reach the desired groove depth ($\sim$17 nm). The etching was followed by a soft ashing procedure with oxygen in order to remove the residual photoresist. As the final step, the substrate was coated with SiC in a thin-film deposition system (AJA Orion-8) to enhance the reflectivity in the XUV range. The fabricated piece, and the overall quality of the etched pattern, was then examined with an optical microscope and a scanning electron microscope (SEM; FEI Nova 200). The results are shown in Fig. 3(c) and (d), respectively. The depth and the ratio of the grooves were measured with a stylus profilometer (KLA Tencor) and are shown in Fig. 3(e).

One major issue limiting the fabrication quality for such a grating is the nature of the curved surface, as this causes non-uniformity in terms of the groove depth $d$ and the ratio $r$. With a ROC of 1000 mm for the concave surface of the mirror substrate, there is a height difference of $\sim$ 80 µm between the rim and the center of the mirror. This makes it challenging to achieve a homogeneous thickness of the photoresist layer, as well to have an even exposure of the photoresist since the curvature leads to the UV light being defocused towards the center compared to the rim of the
mirror. Simulations (see Supplement 1 Section 2 and Figures S2 and S3) show that the value of the groove depth dominantly affects the band-pass center, but the efficiency is almost unchanged for a ±2.5 nm tolerance of the depth. This requirement can be fulfilled in the fabrication used here. As Fig. 3(e) shows, the groove depth is about 18.4±2.5 nm which is within the range of optimal performance.

5. Experimental setup

The performance of the two types of monochromatization is characterized using the HHG-based light source and tr-ARPES setup described in Ref. [24]. This system yields ultrafast, few-hundred-femtosecond long XUV pulses. Figure 4 shows the schematic diagram of the setup, illustrating the specific optical layout. Briefly, the HHG light source consists of two Ytterbium-Doped Fiber Amplifiers (YDFA) lasers, one (Tangor, Amplitude Systems) for driving the HHG of the probe source, and another (Tangerine, Amplitude Systems) for driving an optical parametric amplifier (OPA) which provides the pump beam. Both lasers deliver pulses with a wavelength centered at 1030 nm, and are physically synchronized to share one common oscillator. For the pump line, the infrared (IR) beam has a duration of about 280 fs and is compressed from the OPA to ∼100 fs at an output wavelength of 1.2 µm, which is used for the temporal response measurement shown in the present work. For the probe line, the fundamental light source has maximum pulse energy of ∼300 µJ at a 250 kHz repetition rate. The pulse duration is ∼461 fs. Prior to the HHG, the IR output is firstly tripled in frequency through a two stage up-conversion using non-linear optical crystals, to 343 nm, with an efficiency of about 30%. The UV light is focused by an off-axis parabolic mirror into a gas jet for HHG process, which generates output in the XUV range from 10.8 eV to 32.5 eV. The HHG is conducted in the tight focusing geometry with argon gas as the medium. In this work, we focus on the 3rd (∼10.8 eV) and 5th (∼18.1 eV) harmonic. Following the HHG, a mirror wheel carrying the refocusing optics is placed, with the incidence angle of ∼1°.
6. Performance

Three fundamental properties are characterized to evaluate the performance of the two types of monochromatization investigated in this work. These are: optical quality (focusing), photon flux, and temporal broadening. The experimental results for the mirror+filter based and the grating-based optical setups are presented, and compared, below.

6.1. Optical quality and focusing

The focusing properties of both the simple spherical mirror and the grating are characterized by observing the light spots on a fluorescence screen, and a YAG (Y₃Al₅O₁₂) crystal. The fluorescence screen, which is mounted as a powder-coated vacuum viewport on the vacuum setup, is used to view the diffraction patterns for the corresponding photon energies. Figure 5(a) displays, from top to bottom, the results obtained from the SiC/Si mirror in combination with an Al filter, the grating, and the grating in combination with the Al filter, respectively. For these measurements, the zeroth order diffraction spot from the grating is aligned to the same position on the fluorescence window as the spot from the mirror. It is clearly seen that the grating generates resolvable diffraction orders with spatial separation. For the bottom panel of Fig. 5(a), the spots on the far left and far right are blocked by the Al filter. This verifies that the remaining spots are indeed the fifth harmonic ($h\nu = 18.1$ eV) spots from the HHG, as the Al filter provides a band-pass filter for photon energies above $\sim 15$ eV, thus blocking the third harmonic light ($\sim 10.8$ eV). The spatial separation between the first order spots for 10.8 eV and 18.1 eV, obtained from the image, is about 7 mm. The experimental result is consistent with the simulation presented in

Fig. 4. The experimental setup used for characterizing the monochromator. a) The schematic drawing of the beam path. b) HHG spectral flux on sample as a function of photon energy. c) Model of the HHG and refocusing chamber showing detailed mounting and placement of the optical elements for the example configuration. Mono-components are marked by golden color.
the inset of Fig. 2(a). The 7 mm distance is sufficiently large so that the two harmonics can be selectively filtered out using a small aperture, in case one would like to prevent the unwanted higher orders from reaching the sample. The latter is not always necessary since the distance puts the unwanted orders outside the view of the electron analyzer of the photoemission setup. In the middle panel of Fig. 5(a), the very bright zero order spot, located in the center of the image, is blocked by a metallic foil to avoid damaging the fluorescence window and to allow the weaker first order spots to be imaged with the camera.

Fig. 5. The analysis of the diffracted beam spots from the fluorescence screen. a) The observed spot pattern based on different combinations: mirror+filter (top), grating (middle) and grating+filter (bottom). The ability to separate the harmonics is clearly demonstrated. b) and c) depict the spot size measured with a YAG crystal for the mirror+filter and the focusing grating setups, respectively. Horizontal and vertical spot sizes are marked in the figure.

A YAG crystal is used for accurate determination of the light spot size produced by the two NIM setups. The YAG crystal is placed at the sample position in the photoemission vacuum chamber, and its exact position can be controlled by a motorized manipulator (SPECS) with a calibrated millimeter scale. Figure 5(b) and (c) show the results of the fifth harmonic ($h\nu = 18.1$ eV), as imaged by the spherical SiC/Si mirror and the grating, respectively. The spherical mirror provides a spot size of about $104\times95 \, \mu m^2$ (H×V), while the grating gives $123\times144 \, \mu m^2$. From ray-tracing simulations, presuming a beam waist of the HHG radiation of 10 \, \mu m and a divergence of 5.8 mrad, we obtain a focus of $47.7\times48.1 \, \mu m^2$ (H×V), ~2.1 and ~2.7 times smaller than the measured values, respectively. The deviation between the simulated and measured spot sizes for the mirror+filter configuration, is possibly due to underestimation of the HHG plasma size and to a residual misalignment of the spherical mirror. The 10 \, \mu m HHG source size, determined by the focus of the off-axis parabolic mirror ($f = 101.6$ mm), is in practice difficult to realize due to several reasons, including misalignment, UV beam quality, HHG phase-matching, as well as the roughness of a mirror coating. These uncertainties are present for both types of NIMs, but the additional broadening of the spot produced by the spherical grating mainly comes from the fabrication imperfections, as discussed previously.

6.2. Efficiency comparison

Figure 6 shows the photon flux measurement for the two types of monochromators at 10.8 eV and 18.1 eV photon energies, reflecting the efficiency. The photon flux is determined by measuring the drain current from a piece of annealed tantalum foil placed in the beam path. The yield efficiency is estimated based on the Ref. [26]. The laser that drives the HHG operates at a repetition rate of 250 kHz, and the pulse energy is tunable up to approximately 80 \, \mu J. The flux measurement using the focusing grating is based on the first order diffraction spots from the corresponding harmonics. Overall, the focusing grating provides a considerable enhancement of the monochromator efficiency compared to the mirror+filter solution. In the case of our design
that is specifically optimized for the 18.1 eV photon energy, there is an order of magnitude improvement in the photon flux compared the mirror+filter based solution.

![Flux measurement of the mirror+filter and focusing grating NIM for a) 10.8 eV and b)18.1 eV photon energy.](image)

Fig. 6. Flux measurement of the mirror+filter and focusing grating NIM for a) 10.8 eV and b)18.1 eV photon energy. The results are plotted as a function of the pulse energy driving the HHG. The specific configuration of the monochromator, as well as the efficiency enhancement are specified in the figure.

6.3. Temporal broadening

Figure 7 compares pump-probe measurements of the electronic excitation in graphene measured with the tr-ARPES setup described in Ref. [24], and using the two different types of monochromatization. Since the fifth harmonic with 18.1 eV photon energy has the shortest pulse duration of the two wavelengths considered here [24], the temporal properties were investigated using this photon energy. A p-type graphene sample was selected as a test specimen since it has sufficiently fast intrinsic electron dynamics to precisely reflect the system-limited temporal resolution [27,28]. The sample used is a quasi-freestanding monolayer graphene on a 6H-SiC (0001) substrate [29]. The measurement was conducted at room temperature, and the data acquisition time for each delay point is 5 min.

![Time-and-angle-resolved photoemission spectroscopy measurements on a monolayer p-doped graphene, performed with 18.1 eV (fifth harmonic).](image)

Fig. 7. Time- and angle-resolved photoemission spectroscopy measurements on a monolayer p-doped graphene, performed with 18.1 eV (fifth harmonic). a) The momentum-integrated spectrum as a function of time delay, taken by using focusing grating NIM. The pump beam wavelength is 1.2 µm. Purple box indicates the energy integration window used to obtain data shown in b) and c). b) The excitation intensity as a function of delay (black dots) and fit to the data (red line). The gray line shows the fitted Gaussian peak representing the temporal resolution. c) Same as in b), but for the mirror+filter NIM. FWHM stands for full width at half maximum. All data taken at room temperature.

Figure 7(a) shows the excitation spectra with integrated momentum as a function of delay time, taken with the focusing grating NIM. The pump wavelength is 1.2 µm, and the fluence is approximately 35 µJ/cm². In Fig. 7(b) the energy-integrated intensity within the purple box drawn in Fig. 7(a) is plotted as dot markers. The fit (red line) to the data points is a convolution of a 2-τ-parameter exponential decay curve and a Gaussian function, using the two τ’s, the decay
curve amplitude, and the Gaussian standard deviation $\sigma$ as fitting parameters. The FWHM of the fitted Gaussian distribution (gray line) represents the overall temporal resolution of the system, while the $\tau$-parameters describe the rate of decay after the excitation. Similar to Fig. 7(b), the data in Fig. 7(c) is acquired by using the mirror+filter solution. The same energy window for integration is selected as for the grating case.

For the focusing grating NIM, an overall temporal resolution of about $219.5 \pm 1.9$ fs is found, while the mirror+filter results in a $205.3 \pm 7.6$ fs temporal resolution. These results suggest that the focusing grating NIM introduces a temporal broadening of the probe pulse, as expected. By deconvoluting the broadening from the pump beam, the pulse duration of the probe beam is found to be $\sim 195$ fs and $\sim 179$ fs, for the focusing grating and mirror+filter, respectively. In comparison, the contribution from the grating monochromator is about $72.4 \pm 27.4$ fs, which agrees well with the calculation presented in Fig. 2(b). The additional broadening from the focusing grating gives an increase by 6.8% compared to the mirror+filter, this increase is negligible for most experiments under our experimental conditions. The two sets of data were collected with the same pulse energy used to drive the HHG ($\sim 60 \mu$J), and the same acquisition time. The statistics presented here, as indicated by the noise level and error bars, reflects the increased photon flux at 18.1 eV photon energy when using the focusing grating (error bars for grating data is smaller than the measurement points).

7. Discussion and conclusion

Figure 6 shows that using a low-density focusing grating for monochromatization of 10.8 eV and 18.1 eV photon energies from a HHG source provides higher efficiency compared to a solution based on a focusing mirror in combination with thin film filters. Using the grating instead of the mirror+filter results in a decrease in temporal resolution from 205 fs to 220 fs, which corresponds to a temporal broadening of less than 10%. Since the particular HHG setup used in this work is designed to achieve a high energy resolution, and thus have relatively long laser pulses, the pulse duration of the driving laser is 461 fs [24], and the pulse stretching caused by the grating has little influence on the overall temporal resolution of the system. For HHG sources with shorter pulses, however, the pulse-front tilt induced by the focusing grating can be minimized by reducing the line density. However, this has to be weighed against the reduction in resolving power of the grating, as well as the spatial separation of the diffraction orders at the sample position. In the present work, a line density of 40 lines/mm was selected since this gave the best trade-off between the temporal broadening and the separation of the harmonics. The pulse-front tilt can also be reduced by decreasing the beam size on the grating by an iris or an aperture. Doing so, however, comes at the cost of reduced photon flux. Since the grating based solution provides considerably higher transmission this is however a viable trade-off strategy that provides increased flexibility.

It is worth noting that very few thin film filters work well in the low photon energy range below 20 eV due to high absorption at these wavelengths. In practice, band-pass thin film filters normally require ultra-thin thickness to have decent transmission. Some materials, such as Sn or Si filters ($\sim 100$ nm), are very delicate and can easily be destroyed by handling or by absorption of residual light from the driving laser. Furthermore, frequent replacement is time-consuming and costly. As a filter-less solution, the focusing grating circumvents these challenges. Applying the focusing grating in the higher photon energy range above 20 eV could be achieved by optimizing the groove depth, in combination with a multi-layer coating specifically tailored for the photon energy of interest. The multi-layer coated mirror has recently been demonstrated [24] to provide enhanced reflectivity at photon energy higher than 20 eV, yet it is insufficient to suppress unwanted harmonics and thus demands thin film filters to work. Notably is that even when thin film filters are used, the mirror+filter solution can under certain circumstances let unwanted harmonics pass through provided a sufficiently high intensity in that harmonic. In terms of the fabrication quality of the grating, the most pressing issue is the curvature of the surface, as that can cause a
non-uniformity of the etched pattern. This can be addressed by further optimizing the specific procedures, in particular the lithography process, or by using an error-compensated photo-mask.

In summary, we have compared a monochromator design based on a normal-incidence spherical grating and compared it to a solution based on a spherical mirror and filter. The experimental comparison targets the characteristics important for applications in ultrafast electron spectroscopy. We have designed and fabricated a spherical grating as a compact, single-element monochromator, specifically optimized for a photon energy of 18.1 eV. The performance of the focusing grating monochromator is compared to a solution based on a spherical mirror in combination with a thin film filter – the latter is a commonly used monochromatization scheme for HHG-based ultrafast light sources. Overall, the mirror+filter solution can provide a smaller spot size at the focus with no temporal broadening. The focusing grating can, on the other hand, provide an order of magnitude higher efficiency in terms of photon flux with only a small amount of focus size increase and temporal broadening. The single-element design of the grating enables a very compact optical layout with minimal need for maintenance. The focusing grating, furthermore, requires only a low-cost, commercially available Si-mirror substrate, and can be produced using standard nano-fabrication processing. The final product is both mechanically robust, and can handle a high power loads. In short, the mirror+filter solution is more appropriate in cases where there is no tolerance in temporal broadening, while the focusing grating is more advantageous in efficiency. The presented grating solution also presents significant advantages in terms of low-cost, compact size and high efficiency, and could be relevant for applications outside of photoelectron spectroscopy as well.

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**Data availability.** The data that support the findings of this study are available from the corresponding author upon reasonable request.

**Supplemental document.** See Supplement 1 for supporting content.

**References**