Application Note

ARPES Spectroscopy and ToF Photoelectron Momentum Microscopy Driven by Table-top, High Repetition Rate UV-source at 80 MHz

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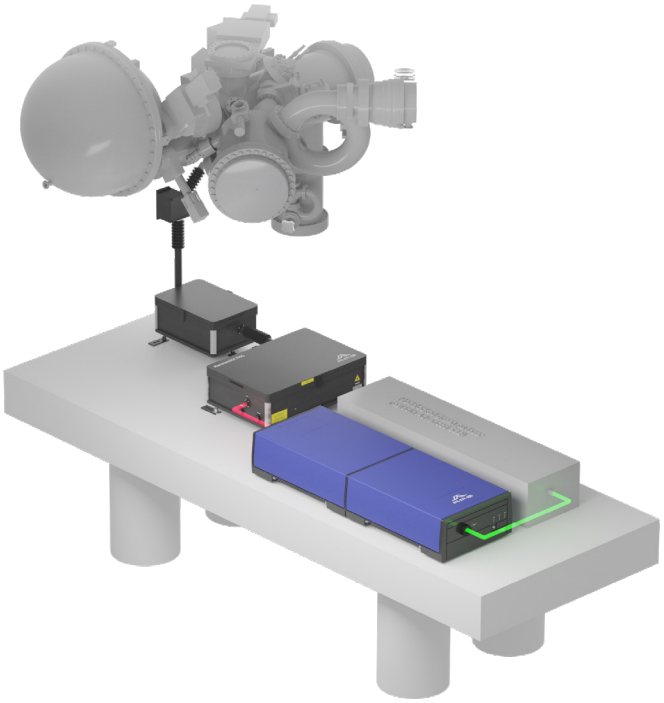
Abstract

We present a combination of a high photon-flux, ultraviolet light source for ARPES/ARUPS spectroscopy from APE GmbH with a novel time-of-flight photoelectron momentum microscope. The latter is comprised by a hemispherical spectral analyzer used as energy-bandpass pre-selector and by a subsequent time-of-flight spectrometer.

The ultraviolet, table-top light source provides extremely narrowband spectra (<0.2 meV) tunable from 2.58 to 6.5 eV at high average powers in the mW range. In combination with the photoelectron momentum microscope, the complete system yields a two orders of magnitude higher data recording efficiency in comparison to a standard approach using only a single hemispherical spectral analyzer. The data recording at the full 80 MHz source repetition rate can be facilitated at a measured energy resolution of ~ 4 meV and with an analyzed region-of-interest in the sub-µm range.



ARPES Introduction

Introduction

Angle-resolved photoelectron spectroscopy (ARPES) is a widely used measurement technique to characterize the electronic band structure and Fermi surface of crystalline solids. Hereby a narrowband light source (typical photon energies from few eV to hundreds of eV) excites photoelectrons from the material surface and the emitted photoelectrons are detected and analysed according to their kinetic energy and emission angle distribution.

A general challenge, when using multi-MHz repetition rate light sources, such as laser-based sources (~ 80 MHz) or synchrotron radiation in conventional multi-bunch mode (100 - 500 MHz) is the incapability of directly employing time-of-flight (ToF) based photoelectron spectrometers due to the small time gaps between two consecutive laser pulses. Since these devices are based on mapping the kinetic energy distribution of the photoelectrons onto a variation of electron flight times, small time gaps between consecutive pulses strongly limit the usable energy resolution of the ToF spectrometers. A few elaborate techniques have been demonstrated to circumvent this problem, with the downside of significantly reducing the usable repetition rate (e.g. transverse electron bunch separation schemes [1, 2], using fast-mechanical choppers for the radiation pulses [3, 4] or electron-optical pulse picking via fast electric beam deflectors [5]).

Here we describe the combination of an optical parametric oscillator (OPO) and a harmonic generation system from APE GmbH together with a novel time-of-flight photoelectron momentum microscope (a hybrid instrument combining a hemispherical analyzer with ToF energy recording). The latter solution allows momentum-resolved measurements at full laser repetition rate while permitting a high energy resolution. We characterize the output pulses of the ultraviolet (UV) laser source, validate its usability for ARPES experiments and demonstrate the state-of-the-art measurement capabilities in combination with the ToF photoelectron momentum microscope.

ARPES Description of the UV Laser-ARPES Light Source

Light Source Setup

The presented UV-light source consists of a synchronously-pumped optical parametric oscillator (Levante Emerald ps) combined with a subsequent frequency mixing device (HarmoniXX FHG), both designed by APE GmbH. The system is pumped by an ultrafast modelocked green laser (Paladin, Coherent Inc.), providing 20 W average power with 15 ps long pulses at 80 MHz repetition rate (see source layout in figure 1).

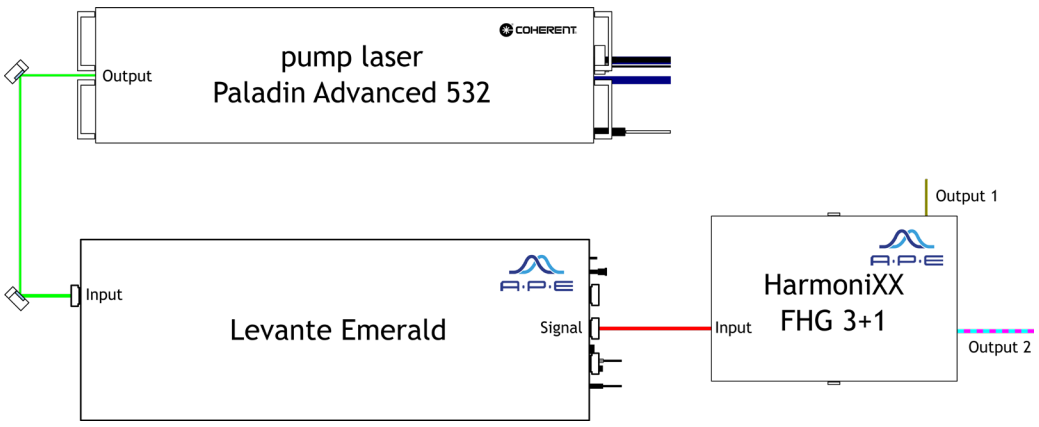


Figure 1: Schematic layout of the UV-light source consisting of a 532 nm pump laser, a sync-pumped OPO and a harmonics generator.

Light Source Working Principle

The pump radiation at 532 nm is converted by the Levante Emerald OPO to Signal and Idler pulses at wavelengths tunable from 690 to 990 nm and from 1150 to 2300 nm respectively. The OPO is wavelength and power stabilized via a computerized, internal feedback-loop seeded by the continuously acquired Signal spectrum.

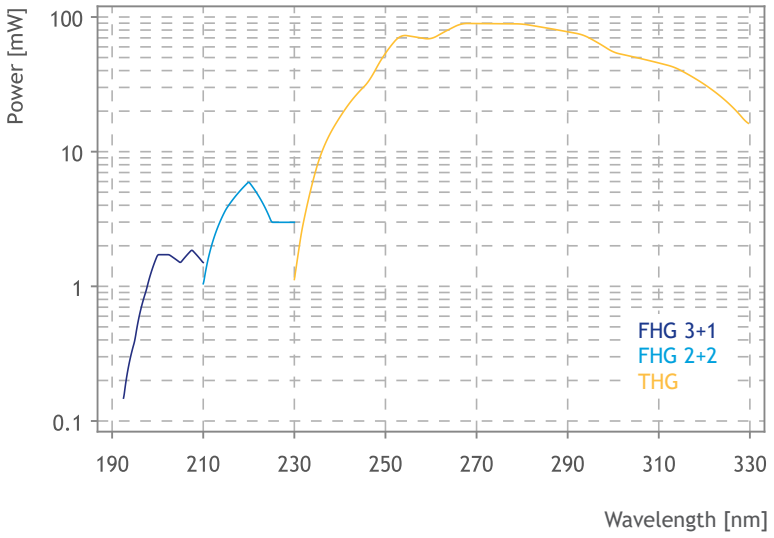
The subsequent frequency mixing device (HarmoniXX) allows the generation of the second harmonic (SHG), the third harmonic (THG) and the forth harmonic frequency (FHG) of the OPO Signal output. The FHG is further differentiated in FHG 3+1 (mixing of third harmonic plus fundamental) and FHG 2+2 (mixing of second harmonic plus second harmonic). Both FHG techniques are applied, due to their distinct phase matching conditions, since they provide slightly different UV output wavelength ranges.

ARPES UV-Light Source

Light Source Performance

The combination of all the facilitated frequency mixing techniques in one single device enables the continuous spectral coverage from 192 to 480 nm output wavelengths (photon energies from 6.5 eV to 2.6 eV). The output average powers as function of the center wavelength are shown in figure 2 for FHG and THG light generation.

Figure 2: Ultraviolet output powers as function of the center wavelength. The source is operated at a pulse repetition rate of 80 MHz.
FHG 3+1 .. sum frequency between third harmonic and fundamental,
FHG 2+2 .. frequency doubling of second harmonic,
THG .. third harmonic generation.



Light Source Advantages

With average powers in the 0.1 mW to 100 mW range over the ultraviolet tuning range, the source provides a much higher photon flux in comparison to synchrotron radiation, widely used in APRES spectroscopy. Another advantage compared to synchrotron radiation and noble-gas discharge UV lamps is a very narrow spectral bandwidth (<0.2 meV FWHM) and a good focusability to below 50 µm at 200 nm wavelength (6.4 eV photon energy).

Furthermore, the UV-light source presented here is fully motorized and allows remote wavelength tuning via a TCP-IP interface for output wavelengths from 210 to 480 nm (5.9 to 2.6 meV). Only wavelength tuning in the range from 192 to 210 nm (6.5 to 5.9 eV) involves additional manual adjustment of the FHG 3+1 nonlinear crystal.
Note that the system can also be pumped by other green, picosecond lasers or by high-power picosecond Ti:Sapphire lasers. Verified combinations are for example pump lasers such as Mira-HP-P (Coherent Inc.) or Tsunami HP ps (Spectra-Physics). Hereby a different average power or pulse duration of the pump laser will directly influence the generated UV pulse energies and spectral bandwidths available for Laser-ARPES experiments.

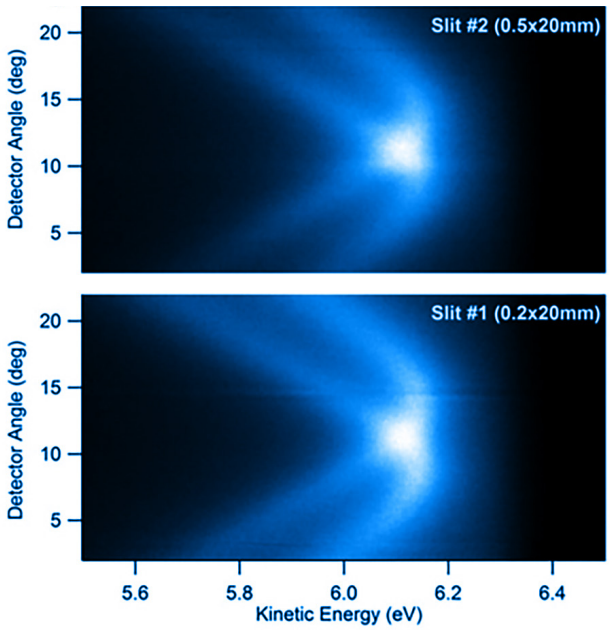
ARPES Measurements on Graphene/Ir(111) with the UV Laser

Description

In order to verify the capabilities of the ultraviolet, laser-based radiation source for ARPES experiments, a proof-of-principle ARPES measurement on Graphene/Ir(111) was performed. The employed detection system is a PHOIBOS 150 hemispherical analyzer (HSA) with 2D-CCD detector (SPECS GmbH). The center wavelength of the UV radiation source is set to around 196 nm (6.3 eV). The measured ARPES maps of Graphene/Ir(111) are shown in figure 3 using two different slit apertures of 0.2 and 0.5 mm resulting in different energy resolutions. The expected ARPES spectra could be obtained, visualizing the known Rashba-split.

These ARPES measurements validate the usability of the ultraviolet, laser-based light source for ARPES experiments. The low photon energy of the source tunable from 2.6 to 6.5 eV makes the system perfectly suited to probe electronic states close to the Fermi level. The below shown measurements were performed by Thorsten Kampen from SPECS Nano Surface Analysis GmbH. A full application report is available upon request from APE GmbH.

Figure 3: ARPES maps of Graphene/Ir(111) sample measured with a 196 nm (6.3 eV) excitation laser. The measurement shows the Rashbasplit of Graphene/Ir(111) surface state. Two different entrance slits (labeled in the figure) of the electron analyzer were used, resulting in distinct energy resolutions.



ARPES ToF Photoelectron Momentum Microscopy with UV-Source at 80 MHz

Setup Schematic

A general challenge, when using multi (10-100) MHz repetition rate light sources, is the incapability of directly using time-of-flight (ToF) based photoelectron spectrometers. In order to circumvent this problem, various techniques were developed, which use a reduction of the effective repetition rate reaching the detection system. This comes typically with the drawback of a reduced data recording efficiency (see references in the introduction section of this report). In contrast, the here described novel time-of-flight photoelectron momentum microscope is able to measure photoelectron momentum and kinetic energy distributions with a high energy resolution at the full UV-source pulse rate of 80 MHz. The device is based on the combination of an energy dispersive element (in this case a HSA) and a low-energy time-of-flight drift spectrometer (see layout in figure 4).

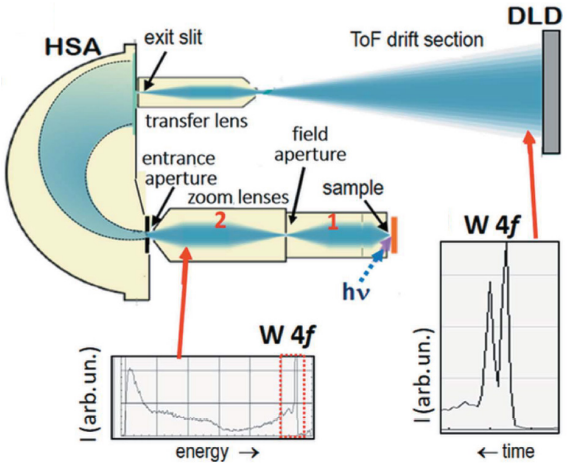


Figure 4: Schematic layout of high-repetition-rate-compatible ToF photoelectron momentum microscope. Device consists of a hemispherical analyzer (HSA) and a ToF-spectrometer. The HSA is used as bandpass-filter on the full energy spectrum (f). The pre-selected energy band is dispersed in the consecutive ToF-spectrometer, leading to a high-resolution spectrum. The various lens groups can project either a momentum pattern or a real-space image on the delay-line detector (DLD). (figure from [5])

Working Principle

First, the hemispherical analyzer cuts a well-defined bandpass from the full energy spectrum. Hereby, the transmitted energy band can be precisely selected via pass energy and slit width. Afterwards the ToF analysis provides a high resolution spectrum of the kinetic energy distribution. A pure ToF analysis, without the pre-filter, would not be possible at high repetition rates at around 80 MHz, due to the short time gap between two consecutive source pulses.

Note that measurements only using a hemispherical analyzer without ToF analysis are possible - but with much lower recording efficiency. In order to achieve similar energy resolutions as with the HSA-ToF combined system, the HSA-slit size and the pass energy E_{pass} would have to be significantly reduced. Furthermore, the measurement would require the classical scan through the transmitted kinetic energies and a combination of the kinetic energy dependent recorded data, leading to a significantly longer total acquisition time (see further details in the following section).

ARPES Bandpass Pre-filter in Combination with ToF Spectrometer

HSA Bandpass Pre-filter in Combination with ToF

As proof-of-principle measurement, the above described HSA+ToF hybrid photoelectron momentum microscope was used in combination with the presented UV-light source to investigate the ARPES spectra of Rhenium in the crystal orientation Re(0001). The wavelength of the UV-source is set to 194 nm (6.4 eV photon energy).

For the experiment the pass energy E_{pass} of the hemispherical analyzer was set to 200 eV and a 1 mm slit was used to pre-select an energy band with a transmitted bandwidth of 440 meV. The drift energy of the ToF spectrometer is set to E_{drift} = 9 eV resulting in an energy resolution of 9 meV of the recorded spectra. Figure 5 shows the obtained sections through the 3D data array $I(k_x,k_y,E_{kin})$ in both transversal momentum directions (k_x and k_y). The full recording time of this HSA + ToF hybrid measurement was 10 minutes.

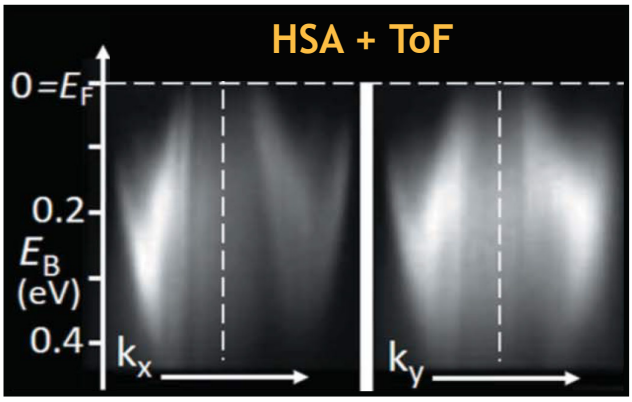


Figure 5: ARPES momentum measurement of Re(1000) surface using the bandpass pre-selection approach (HSA+ToF hybrid mode). The HSA is set to a transmitted energy band of 440 meV and the subsequent ToF analysis features an energy resolution of 9 meV. Shown are the binding energy E_B-versus momentum $k_{x,y}$ cuts of the measured data set. (data from [5])

ARPES Bandpass Pre-filter in Combination with ToF Spectrometer

Classical HSA Measurement

In comparison, figure 6 shows the results obtained with the classical approach of only using one HSA device. This measurement is also performed with the same HSA + ToF system, but with an effectively switched-off ToF-section by setting the ToF drift energy to a high value (E_{drift} = 200 eV). The HSA slit size and pass energy had to be reduced to 0.2 mm and 25 eV respectively in order to achieve similar energy resolutions as in the previous case. In this classic scanning approach, the kinetic energy was scanned in 5 meV steps over a 1.5 eV range. At each scan step a measurement was recorded, resulting in a total measurement time of 1.7 h (300 exposures of 20 s, total 1.7 h). The k_x - k_y momentum pattern of the Re(0001) Tamm state close to the Fermi energy E_F and the binding energy E_B-versus-k_x section from the composed data are shown in figure 6.

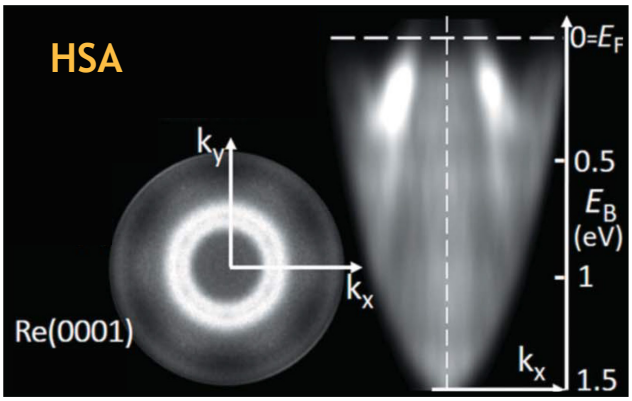


Figure 6: ARPES momentum measurement of Re(1000) surface using only a hemispherical analyzer in the classical scanning mode (scan of transmitted E_{Kin} with data collection at each scan step). For better comparability, the same device as in figure 5 was used, but with disabled ToF section. The shown data are a convolution of 300 measurements at different transmitted E_{Kin} (300 steps of 5 meV over a total range of 1.5 eV). Shown are the k_x - k_y momentum pattern close to the Fermi energy E_F and the E_B-versus-k_X cut section from the composed data. (data from [5])

Enhancement of Recording Efficiency due to Novel HSA + ToF Approach

In order to compare the recording efficiencies between the two measurement approaches (only-HSA vs. HSA+ToF hybrid mode), the acquisition time of the same data set with identical statistics has to be compared (equal number of counts in the [k_x, k_y, E_{kin}]-array with identical energy resolution). A detailed theoretical analysis of the influence of HSA exit slit size, pass energy E_{pass} as well as the influence of the HSA entrance slit on the acquisition efficiency for both approaches is given in section 2.5 of reference [5]. In this paper it is also experimentally shown that the HSA+ToF hybrid mode yields a roughly 150 times higher data recording efficiency compared to the only-HSA operation with parameters similar to the measurements shown in figure 5 and 6.

Hereby the great advantage of the HSA+ToF hybrid mode compared to the only-HSA operation is that the combination of a high pass energy of the HSA and the subsequent ToF analysis enables the parallel acquisition of many energy slices at the same time.

ARPES Sub-µm ARPES via Field Aperture and High-flux UV Radiation Source

Sub-µm ARPES

To enable the investigation of inhomogeneous or microstructured samples, it is important to have a high spatial-resolution (preferable in µm range) of the measurement device. Hereby simply focussing the excitation photons on a µm-spot size is technically very challenging (e.g. requiring zone plates, Sigray capillaries or others).
Here we demonstrate that the high photon-flux of the presented UV radiation together with the ToF momentum microscope enables the measurement of sub-µm ARPES spectra without the requirement of a small photon spot.

Therefore a field aperture is placed in the first intermediate real-space image plane of the ToF momentum microscope ("field aperture" label in figure 4) selecting the desired position and size of the analyzed region of interest (ROI) on the sample surface. Note that a small contrast aperture in the photoemission electron microscopy mode of the instrument typically confines the k-range acceptance necessary for momentum imaging - independent of the size of the photon spot. The scheme is illustrated in figure 7 by the example of the antiferromagnet Mn_2Au. In real-space imaging mode (PEEM; left and centre images) the antiferromagnetic domains are visible. As contrast we exploit linear dichroism from two measurements with perpendicular E-vector. Then the field aperture is driven either to a red or a blue domain and the microscope is switched to k-imaging mode (right side, figure 7). When the electrons pass the small aperture, their k-pattern is encoded in terms of the electron angular distribution (red arrows).

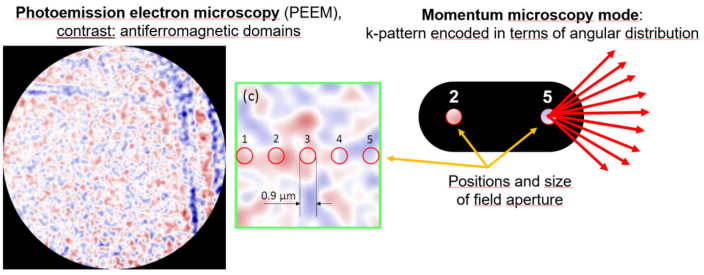


Figure 7: Small-area momentum microscopy (here with ROI 0.9 µm) of the antiferromagnet Mn_2Au. The domains (red/blue) are visible in the PEEM image (left, detail in centre) and can be selected by the position of the field aperture (right). The arrows indicate the angular pattern of electrons passing the aperture. PEEM images from [6].

Using a 10 µm aperture in the image plane, with a magnification factor of M=14 of the imaging lens system, a ROI size of below 0.8 µm on the surface was experimentally demonstrated [see 6]. Hereby the maximum measureable kinetic energy was 2.5 eV, corresponding to a photoemission momentum horizon of $k_{||}^{max} = 0.8$ Å^{-1}. The big advantage of this approach (aperture in image plane) is that the beam spot size on the sample does not influence the spatial resolution of the measurement. In fact the beam spot size was significantly larger than the analyzed ROI, requiring the high-photon flux of the UV-radiation source in order to capture sufficient signal from the sample ROI. For further information on these measurements, see section 2.6 of reference [5].

ARPES Conclusion

Summary

We presented a combination of an ultraviolet, high-flux light source (comprised of an OPO and frequency converter from APE GmbH) with a novel ToF photoelectron momentum microscope. The table-top source features high-power, narrowband output spectra (0.2 mV) tunable from 192 to 480 nm center wavelengths (6.5 to 2.6 eV photon energy). In combination with the ToF momentum microscope, ARPES experiments at the full 80 MHz source repetition rate supporting energy resolutions of ~ 4 meV and analyzed sample regions in the sub-µm range were demonstrated.

Further information

The ToF photoelectron momentum microscope and the performed measurements presented in the previous sections are published in a recent publication [5]. First scientific results can be found in [6].

References

[1] C. Sun et al., Phys. Rev. Lett. 109, 264801 (2012).
[2] K. Holldack et al., Nat. Commun. 5, 4010 (2014).
[3] M. Cammarata et al., Rev. Sci. Instrum. 80, 015101 (2009).
[4] D.F. Förster, et al., Opt. Lett. 40, 2265 (2015).
[5] G. Schönhense et al., J. Synchrotron Rad. 28, 1891–1908 (2021).
[6] O. Fedchenko et al., e-print on arXiv 2110.12186 (2021)

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