Condensed Matter Research with High-Energy Synchrotron Radiation

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The utilization of high-energy X-ray scattering at synchrotron radiation facilities has developed from a niche method to a fast-growing mainstream application over the last 30 years. After pioneering work using γ-radiation from strong radioactive sources to study electronic wavefunctions by means of Compton scattering [1] and for diffraction experiments to investigate with unprecedented high resolution large single crystals intended for use as neutron monochromators at the Institute Laue-Langevin (ILL) (France) [2], the first high-energy synchrotron radiation experiment at 150 keV was performed at CHESS[1] by the R&D Group of NSLS[2] [3]. With the high field wiggler beamline BW5 at DORIS III, also DESY[3] (Germany) played an important role to develop and promote the unique opportunities offered by photon energies larger than 40 keV [4]. As a result, early on dedicated beamlines for high-energy Compton scattering and diffraction experiments were realized at the hard X-ray 3rd generation facilities: the ESRF[4], the APS[5], SPring-8[6], and PETRA III[7]. Because of the high demand for high-energy X-ray beamlines for structural analysis and imaging of materials, facilities like NSLS and Diamond[8] also developed strong programs using wiggler beamlines.

The cross sections for the interaction of X-rays with matter depend on the number of electrons in the material and decays with photon energy as detailed in Fig. 1. In addition, the short wavelengths of high energy X-rays lead to very small Bragg angles, therefore the Ewald sphere of diffraction is almost flat. The scattering becomes concentrated in the forward direction enabling to work in even complex sample environments up to very high momentum transfers. The enormous, accessible reciprocal space facilitates the measurement of total scattering such as for pair distribution function (PDF) analysis to study even materials with only short-range ordering. The development of large area detectors recording data of large fractions of reciprocal space in sub second time slots was groundbreaking for a wide range of application of high-energy X-rays for *in situ* and *operando* research in materials science, in chemistry, on nanomaterials, on engineering materials and functional devices, in geology etc. In general, the limit for the highest accessible momentum transfer is given by the minimum signal/noise ratio required for the interpretation of the scattering data. The latest detector development using CdTe as sensor material gave access to single photon counting detectors with high quantum efficiency even for high energy X-rays with low background noise, high frame rates, and gating options, which empowered fast in situ and operando experiments and precise structure determination by detecting strong and weak scattering at the same time. High brilliance synchrotron radiation facilities provide intense incident beams at even 100 keV and enable focusing

1 Cornell High Energy Synchrotron Source, Cornell University (USA)
2 National Synchrotron Light Source at Brookhaven National Laboratory (USA)
3 Deutsches Elektronen-Synchrotron, Member of Helmholtz Association (Germany)
4 Advanced Photon Source at Argonne National Laboratory (USA)
5 European Synchrotron Radiation Facility (France)
6 Super Photon Ring-8 GeV of Institute of Physical and Chemical Research RIKEN (Japan)
7 Originally "Positron-Electron-Tandem-Ring-Anlage" at DESY (Germany)
8 Diamond Light Source (United Kingdom)

to µm range spot sizes. The focusing allows the study of surfaces, interfaces, as well as thin layers, and small volume elements buried inside large samples. Imaging techniques like tomography and grain mapping are widely used especially in applied research, Compton imaging of light materials in a heavier matrix is of great potential.

This SRN Special Issue on High-Energy Synchrotron Radiation Research includes contributions from seven facilities with dedicated programs in the spectral range above 40 keV. A collection of examples of science shows the broad field of applications and demonstrates how high-energy X-ray scattering experiments enlarge the toolbox for fundamental and applied research, and how the techniques using high-energy photons complement X-ray scattering in the hard X-ray regime as well as neutron scattering.

Worldwide the next level of progress in X-ray science will be stimulated by the availability of storage ring facilities with much lower emittance getting closer and closer to the diffraction limit even for hard X-rays. This became possible by using a novel multibend achromat lattice pioneered at MAX IV in Lund, Sweden. All large high-energy X-ray facilities perform or plan upgrades, ESRF already concluded the upgrade to ESRF-EBS and is back to user operation. APS will follow in 2023, and also SPring8 and PETRA are going to upgrade the lattice. The development of highly precise insertion devices as in-vacuum or superconducting undulators will improve significantly the flux. Fig.2 shows the enormous brilliance gains as realized by ESRF-EBS. The source size of the electron beam shrinks horizontally to an almost round beam, and the brilliance at 100 keV photon energy will become comparable to the brilliance at 10 keV today. The combination of these improvements with the high penetration power and the large accessible reciprocal space of high-energy X-rays will enable novel research prospects. The study of dynamics using X-ray photon correlation spectroscopy can be expanded to complex sample environments and buried materials as common in functional devices. Another opportunity will be coherent diffractive imaging applied to an extended reciprocal space, or to study buried grains in engineered materials. Also, the high resolution of computed tomography (CT) methods can be applied to extended objects with phase information on the micrometer-scale for diffraction experiments (XRD-CT). To take full advantage of the upgraded sources, the further development of detectors for high-energy X-rays will need to continue. Moreover, the development of novel algorithms and faster computing speed is essential for the reconstruction of images on the flight, especially with more material in the observed volume. Machine Learning approaches will enable the interpretation of *in situ* and *operando* data taken in complex environments involving plentiful information of various materials, which is essential for the design and improvement of functional materials

The Special Issue in hand demonstrates the great potential and the bright future of high-energy Synchrotron radiation research. The articles outline the history of the programs using high-energy photons, showcase the great scientific impact with several examples to pique the curiosity for this energy range, and conclude with a look to future developments at the facility.

References

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Figure captions

Fig. 1: Photon cross section for the interaction with matter as a function of photon energy for the example of Cu [5]. Photo electric cross section and elastic Thomson scattering decrease, while inelastic Compton scattering increases with increasing energy. Overall absorption decreases and the picture demonstrates the advantage of using high energy X-rays for studying properties of bulk materials, as e.g. in engineering materials science and functional devices.

Fig. 2: Spectral brilliance plots calculated for different insertion devices at MAX IV and ESRF-EBS, the first synchrotron radiation facilities with the new multibend achromat lattice (7-bend), which are by now in user mode. MAX IV spectra were calculated for 3 GeV electron energy and the nominal current of 500 mA, the ESRF spectra for 6 GeV electron energy and nominal current of 200 mA. A comparison is made between the operation with the new lattice of the ESRF and the previous double-bend achromat lattice in use until 2018 (dotted lines) showing a gain in brilliance of about a factor of 30 for typical insertion devices for high-energy x-rays. (calculated by Joel Chavanne, ESRF)

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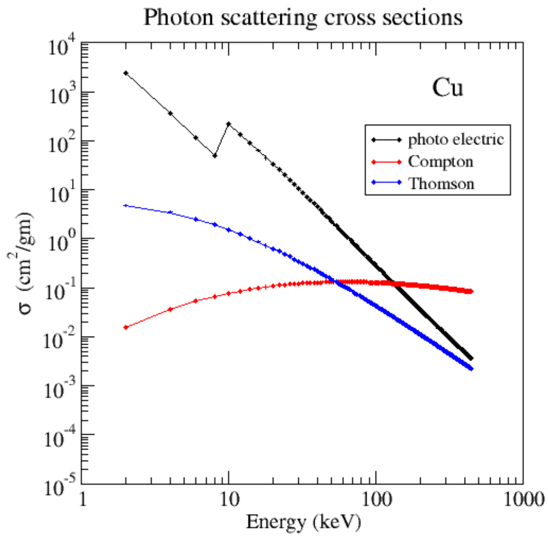
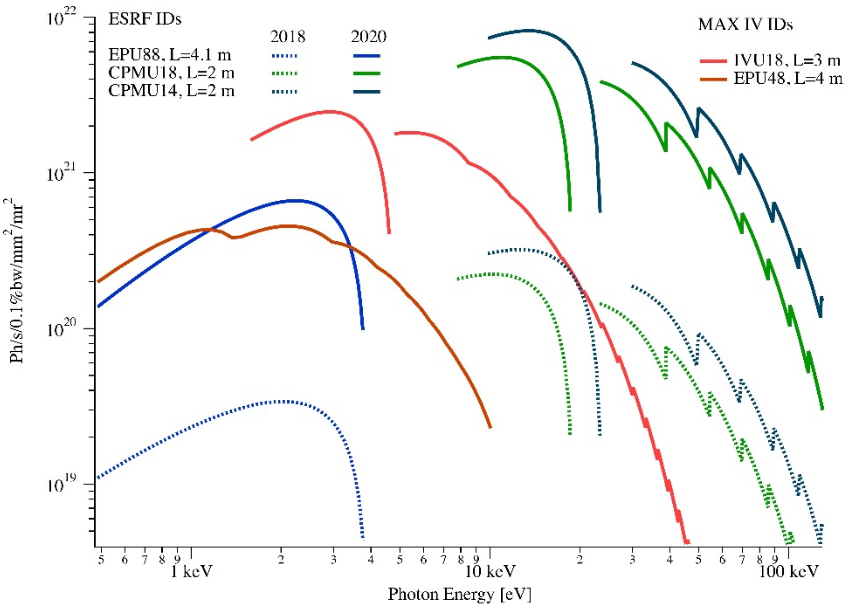
Fig. 1



Photon scattering cross sections
Cu
photo electric
Compton
Thomson
σ (cm²/gm)
Energy (keV)
10^4
10^3
10^2
10^1
10^0
10^-1
10^-2
10^-3
10^-4
10^-5
1
10
100
1000



ESRF IDs
2018
2020
EPU88, L=4.1 m
CPMU18, L=2 m
CPMU14, L=2 m
MAX IV IDs
IVU18, L=3 m
EPU48, L=4 m
Ph/s/0.1%bw/mm²/mr²
Photon Energy [eV]
1 keV
10 keV
100 keV

Fig. 2