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STRUKTURA 2017 – INTRODUCTION

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During the last conference Struktura 2016 in Tábor (September 2016) and CSCA assembly, a proposal appeared to repeat colloquium with the title Experimental Methods in X-Ray and Neutron Structure Analysis. Such conferences were organized in past – in Bechyně, 1981 with proceedings [1], in Ostrava, 1994, with even more extended proceedings [2] and in Ostravice 2002 with extended abstracts [3] and some texts delivered that are currently published only on the web [4]. Probably, the most complete texts are in [2] which are already more than 20 years old, though. Some of the texts were updated in [3-4]. Nowadays, it is more and more difficult to convince anybody to write longer consistent text that may be very useful but it is is actually not taken into account on any evaluation of "scientific output". Looking at the programme of these meetings one can see the main topics – sources of X-rays and neutrons, optics and detectors, single crystal diffraction, powder diffraction, neutron diffraction, small-angles scattering, measurement of residual stresses and textures, topography, measurement at high pressures, low and high temperatures.

In the last decade, there has been fast development of X-ray instrumentation i.e. – sources, optics, detectors. Finally, one contribution at this conference is devoted to X-ray optics and several others to the news and experience in the instrumentation. So, here just a few remarks on X-ray sources and detectors are given.

Table 1. Wavelengths (in Å) of characteristic radiation for most common anodes, energy of K 1

Anoda	K 1	K	K	E(keV)
Ag	0.5594	0.5638	0.4971	22.11
Mo	0.7093	0.7136	0.6323	17.44
Cu	1.5406	1.5444	1.3922	8.04
Co	1.7890	1.7928	1.6208	6.93
Fe	1.9360	1.9400	1.7566	6.40
Cr	2.2897	2.2936	2.0849	5.41
W	0.2090	0.2138	0.1844	58.87

X-ray sources

Texts on conventional laboratory sources and also synchrotrons can be found in the above mentioned literature [2, p. 1-10], [3, p.79-81].

Conventional sealed-off X–ray tubes have been produced for many decades and basically look the same all the time – just different size of focus is offered and what is the most important – different anodes. Traditionally the tubes were made of glass but ceramic tubes appeared later. The power is typically 2-3 kW. Drawback of conventional tubes is necessity of water cooling and the spectrum with several components (mainly K $_{1,2}$ and K) some of which should be suppressed or removed by optics or high-energy resolution detectors. The main task is selection of the tube suitable for particular problem. The most common anodes are in Table 1. The reasons for selection may be related to – symmetry and size of unit cell, reduction of fluorescent background and penetration depth. The increase of power was allowed in the construction of *rotating anodes* that are still produced usually with power of 9 or 18 kW.

A new approach appeared when the tubes were combined with focusing X-ray optics. This allowed to construct simple *air-cooled tubes* with typical power of 30 W (introduced by Incoatec) that can be used in all experiments where small spot is sufficient or sometimes also for low-angle diffraction. Quite recently, completely new idea was realized overcoming the problem with cooling – *liquid anode*. The Metal Jet system is using Ga-alloy in a liquid form. The power achieved is more than100 kW/mm^2. Since this is the alloy, in addition to Ga K_a (1.34 Å, 9.25 keV) also In K_a spectral lines can be used in principle.

Different physical principle is used in *synchrotrons* where relativistic electrons with acceleration are used for generation of X-rays. The principle is old and synchrotron radiation was observed for the first time in the General Electric Laboratory in 1946 as more or less parasitic effect. First dedicated synchrotrons have been constructed since about 1965. Nowadays, nearly 50 synchrotrons are available over the world [5]. The so-called second generation of synchrotrons used mostly just bending magnets for circular motion of electrons, in the third-generation an importance was given to focusing and insertion devices – undulators and wigglers were frequently used. Synchrotron radiation has many well-known unique properties – high intensity and in particular, very high brilliance that is many orders higher than for laboratory sources, wide continuous spectral range, well-defined beam, high polarization in the plane of orbit, pulse structure, natural beam collimation, small divergence. These properties enable many different and unique experiments otherwise impossible in laboratories.

The plans for construction of CESLAB (Central European Synchrotron Laboratory) [6] in Brno were unfortunately stopped. However, one of previous Struktura conferences was devoted to synchrotron and this project [7]. Two years before, the first and last, till this moment, Struktura abroad was organized – a trip with two buses to synchrotron in PSI Villigen and ESRF in Grenoble [8].

Free electron laser (XFELs) is the last stage of these sources. If the electrons are bunched at radiation wavelength it allows coherent emission and consequently further increase of brilliance by several orders. 13 XFELs are currently reported in construction or partial work [5]. There is one contribution at the meeting also about XFEL.

X-ray detectors

A nice paper on classical X-ray detectors was presented many years ago at our meeting [2, p. 25-36]. Since that time, there was no systematic contribution on detectors but many talks, especially by companies, were devoted to new detectors since nearly every year there are some news in this field and new products are introduced.

General requirements on X-ray detectors are, of course, sensitivity to the corresponding energies, often high dynamic range, sufficient precision, for 1D or 2D detectors also space resolution, sometimes low mass etc. The detectors can be divided according to the principle (what is actually measured) or dimension. Classical detectors are *proportional* (using gas ionization) and *scintillation* ones. These are the mostly used point detectors. Proportional detectors are quite simple – glass tube with a wire anode and working gas (e.g. Xe + CO_2 or CH_4). The regime of work depends on voltage and for the application in X-ray range, the photons should initiate ionization avalanche restricted to the region around the wire electrode and not in the whole tube. Scintillation detectors use mostly the crystal NaJ + 1% Tl but ceramic alternative ($YAlO_3$ etc.) was introduced Crytur Turnov which have several advantages – unlike NaJ they are very stable in air, they can be very thin and have short flashes. Scintillation detectors have very high quantum efficiency but relatively high thermal noise and poor energy resolution. This is better for proportional detectors (with lower efficiency) where some electric pulse discrimination can be used to restrict detected photons in an energy window. By far the best energy resolution can be achieved by *semiconductor* detectors (Si(Li), Ge(Li)) that must be kept at low temperatures, which is their main drawback. Bruker used Sol-X detector cooled by Peltier cell down to 100 °C with energy resolution better than 300 eV.

Proportional detectors were also used for production of 1D wire detectors (Braun, Inel) where not only signal created by X-ray photons was detected but also the location from the delay the signal reached both ends of wire was evaluated. A grid version of this allowed construction of 2D detectors (Bruker High-Star, microgap detector, Våntec).

Of course, the first 2D detector is also the oldest one – simple *film*. This is also the cheapest X-ray detector. However, the wet process of developing is not very pleasant and nowadays we have a similar alternative – *image plates* that also work on similar principle when a latent image is created in the sensitive film and then developed in a scanner by scanning with focused laser beam – laser stimulated fluorescence. The image is then completely erased by white light and the plate can be used again. Such detectors have very high dynamic range, reasonable sensitivity but higher background. They are very flexible and can be quite large. For some experiments the disadvantage may be longer read-out time.

Since 1987, for long time CCD (Charged Couple Device) 2D detectors have been used intensively where the X-ray photon generates hundreds of electron-hole pairs. The detectors have lower dynamic range but excellent space resolution. Usually indirect detection is used via phosphor screen from which the signal is transferred by fibre optics into the CCD element This method has higher dynamic range and worse space resolution. Much effort has been put into the optimization of design of these detectors by several companies. Nowadays, the CMOS detectors seem to be preferred.

In last years, 1D strip detectors have become popular in powder diffraction (Panalytical X'Celerator – real time multiple strip detector, Bruker Lynxeye, Rigaku 1DTEX etc.). Variant LYNXEYE XE provides quite high energy resolution which allows electric discrimination of K lines and significant suppression of fluorescence (e.g. Fe, Co in samples measured with CuK).

Modern detectors are based on the so-called single photon counting technology, i.e. direct change of X-ray photon to electric charge with optimized solid-state sensors and CMOS readout in hybrid pixel technology. They were introduced in the XRD by Dectris (Pilatus, Mythen). They have excellent signal-to-noise ration, high dynamic range, zero dark signal, zero readout noise and short readout time. Such detectors (or similar) are now offered by other companies like Rigaku (Hypix), Panalytical (PIXCel). As mentioned above, nearly every year some news in the field appeared. In last years, CdTe detectors for hard X-rays were introduced, for example. So we may expect news also at this conference.

In addition to the mentioned references, one can find information on sources, optics and detectors in nearly each monography on X-rays, e.g. [8-10], in particular on 2D detectors [11].

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IMAGING AT BRIGHT X-RAY SOURCES

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X-ray imaging is a broad term which includes a number of techniques, ranging from raster scanning to full-field using attenuation, scattering or X-ray emission as contrast mechanism. Lately the real breakthroughs happened in terms of improvements in the temporal resolution of all these diverse techniques. With the new generation of X-ray sources (diffraction limited storage rings such as MAX IV and X-ray free electron lasers) it is likely that this trend will continue shedding light to nature's ultra-fast phenomena at the micrometer scale and beyond [1].

In the last 15 years of tomographic microscopy at synchrotrons we experienced in average an order of magnitude improvement in the acquisition speed every three years [2]. While in the early times the scan time of 60 minutes at 1-3 m voxel size was the state-of-the-art, today we can achieve a temporal resolution of 20 ms. Which are the main scientific drivers for this development? What technical and conceptual breakthroughs contributed to such a spectacular improvements? Are we at the technical or physical limits of the spatio-temporal resolution? What will the future of diffraction limited light sources bring? These will be some of the questions I will reflect on in my lecture and indicate some new directions in multimodal and multiscale imaging using advanced X-ray optics [3]

The standard imaging instruments at synchrotron beamlines perform very well down to about one micrometer spatial resolution. Breaking the one micrometer resolution barrier is conceived typically by transforming the nearly-parallel synchrotron beam to divergent beam to achieve magnification in the X-ray regime. At XFEL sources it is still not obvious which imaging modality is best suited to achieve spatial resolution in the range of some tens of nanometers. At this scale the organelles in cells can be viewed. Are such studies feasible at all? To shed some light onto this I will review the performence of different nanoimaging modalities in terms of sensitivity, dose efficiency, spatio- temporal resolution, requirements on the sample preparation and availability at synchrotron sources [4].

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SINGLE CRYSTAL DIFFRACTION METHODS WITH THE FILM REGISTRATION (2D)

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Film methods served for several decades in X-ray laboratories for determination of lattice parameters, space groups, and for checking of quality of crystals. During the era of point detector diffractometers, the candidate crystals were very often studied by film methods prior data collection. More recently the X-ray film was sometimes replaced by image plates. Moreover the film images can now be digitized with aid of a scanner.

Nowadays these methods, perhaps with few exceptions represent a chapter in a history of crystallography, rather than everyday reality in laboratories. Nevertheless, from time to time, let's remember this piece of history.

The methods are as follows [1, 2]:

Laue method [3, 4]: Stationary crystal and film, unfiltered polychromatic radiation. Diffraction pattern registered on the flat film. Reciprocal lattice image collapsed and distorted. Possible front reflection or back reflection arrangement. In the later arrangement relatively large crystals can be studied. Still used for the orientation of bulk crystals.

Oscillation and rotation method: Rotating or oscillating crystal is mounted on the crystallographic head in the axis of cylindrical stationary film cassette, monochromatic (usually filtered) radiation. Reciprocal lattice image distorted and collapsed. Reflections are arranged on layers, corresponding to reciprocal lattice planes.

Weissenberg method [1, 2]: Arrangement the same, one layer is selected by the cylindrical layer line screen. Moreover, the cassette is placed at the carriage moving there and back synchronously with the oscillation of crystal. The result is an un-collapsed but distorted image of one selected reciprocal lattice plane.

Precession method [5]: Both crystal and film are performing the precession motion in two synchronously moving gimbal rings. Circular layer line screen selects one reciprocal lattice plane. Because of the equality of crystal and film motions, an undistorted image of reciprocal lattice plane is recorded.

Cone-axis method [5]: Auxiliary to the precession method, provides circles corresponding to reciprocal lattice layers.

De Jong-Bouman method [1,2]: Crystal as well as the flat film is synchronously rotating. Alternative method to obtain undistorted and un-collapsed reciprocal lattice image.

Gandolfi method [6]: Provides powder patterns from single crystals due to an additional the rotation of the specimen.

The undistorted images of reciprocal lattice planes as well as powder diffraction data can be easily and more quickly obtained by processing of frames recorded by modern diffractometers with area detectors.

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