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DIFFRACTION MEASUREMENTS AT SOURCES OF SYNCHROTRON RADIATION

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Structure determination of biological macromolecules and their interactions with other molecules important for life play a principal role for understanding their function, i.e. for development of new generation of drugs, for environment, for agriculture, for food industry, for protection of human life, etc. Nowadays, the diffraction experiments performed at sources of synchrotron radiation dominate in successful structure determination of macromolecular systems. The last year, already more than 80 % of ~ 6000 papers publish works based on the use of synchrotron radiation and a percentage of the remaining experiments made using home diffraction equipment is continuously decreasing each year.

There are more than 70 synchrotron sources in 23 countries. Many synchrotrons have more beam lines for macromolecular crystallography, e.g. APS in Argonne USA have 19 beamline specialized for protein crystallography, ESRF in Grenoble 14 beamlines, Spring-8 in Japan 11 beamlines, etc. More information can be found in [1].

In spite of the fact that Czech crystallographers use extensive official and unofficial contacts with synchrotrons placed exclusively in west part of European Union, they cope with a critical shortage of measuring time. The average one beamtime a year per one laboratory is hopelessly much low than required for really competitive scientific work in structure biology.

The synchrotron CESLAB (Central European Synchrotron Laboratory) planned if frame of the Structural Funds of EU involves in addition to many other unique equipments devoted to medicine, spectroscopy, etc., also a modern macromolecular beamline devoted to protein crystallography solving practically all requirements of protein crystallographers for the next 20 30 years.

The CESLAB is not designed for protein crystallographers only. The synchrotron radiation is required almost in all fields of natural sciences. The construction of CESLAB allows almost any extension or modification of the equipment in future. In spite of the fact that the present design of synchrotron 12 beamlines devoted to different scientific areas, the construction allows an addition up to several tenth other experiments possibly claimed and designed in future without stopping the operation of the standing beamlines.

All equipment planned for modern synchrotrons of CESLAB type has been developed and produced many years by the best world laboratories. Therefore, it is very reliable and represents a top the world science.

Top of the experimental technique as far as the sources of X-ray radiation represent also newly developed "home sources of radiation". One of them **"LYNCEAN compact light source"** simulates the standard synchrotron source but instead of the insertion device (e.g. undulator with ~1 cm gap) it uses very intensive IR laser pulse (wavelength ~

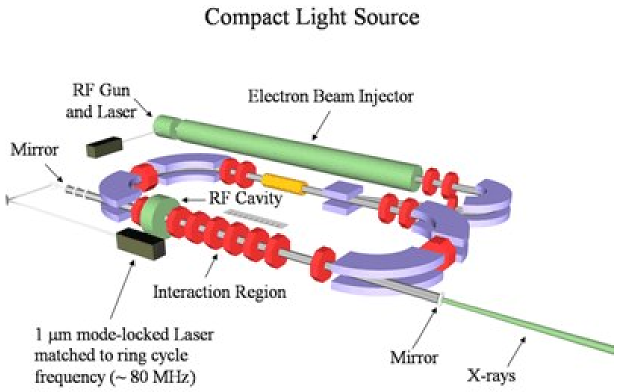

Figure 1. Schematic view explaining principles and a function of the LYNCEAN compact light source. Reprinted from http://www.lynceantech.com/sci_tech_cls.html.

1 m) synchronized and sent against the intensive electron beam in the storage ring (Fig. 1) http://www.lynceantech.com/sci_tech_cls.html.

In comparison with synchrotron, it allows substantial lowering of energy of the electron beam necessary to get the same wavelength of radiation and thus to reduce at least 10 times the size of the whole equipment (to ~ 20 m). Disadvantage is that a small size of the storage ring does not allow to place more beamlines on the ring. However, the only beamline available at LYNCEAN can be split to two or three devices inside of the experimental hutch.

The principle of LYNCEAN was verified in the factory in California on the functional prototype emitting the radiation. Because the first equipment specialized for protein crystallography is still in stage of verification only, a real brilliance of radiation (dependent strongly on the IR laser parameters) and other characteristics (polarization, divergence, etc.) are still a subject of guesses only. Estimates of performance are at level of a bending magnet of the second generation synchrotrons. The equipments of this type belong undoubtedly to the most perspective radiation sources for future, because it will allow higher diversity and a possibility to purchase their own specialized sources of radiation to the individual research institutions.

Another type of "intensive home source of radiation" is **MIRRORCLE RAY 20SX** designed in Japan. It is based on the same principle as a standard X-ray tube, however the electron beam falling on target is accelerated to very high energy (1 – 20 MeV). An impact of accelerated electron beam on target produces "bremsstrahlung" (i.e. white radiation) up to very high energies (Fig. 2). The very high energy radiation can pass through thick blocks of concrete or steel. It makes this product especially suitable for industrial testing of quality of buildings or heavy industrial products (the equipment with this determination that can be trans-

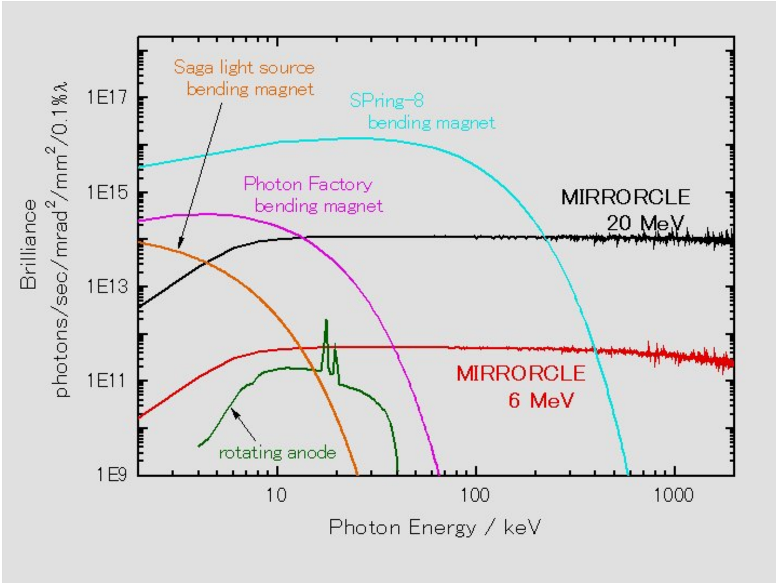

Figure 2. Performance of MIRRORCLE 6 MeV and 20 MeV. The calculated brilliance [photons/sec/mrad2/mm2/0.1%] in the whole spectrum. The 20 MeV model of MIRRORCLE is compared to the 6 MeV model for carbon target, to the standard rotating anode, and to the bending magnet of the older generation synchrotrons. The radiation used in protein crystallography should be highly monochromatic and is about 12 keV. Complete removal of radiation over 100 keV represents a serious problem. Reprinted from http://www.photon-production.co.jp/e/ PPL-monochroX-ray.html#XAFS.

Table 1. Comparison of radiation from MIRROCLE, X-ray tube and bending magnet. Reprinted from http://www.photon-production.co.jp/e/PPL-monochroX-ray.html#XAFS

	MIRRORCLE 20 MeV	X-ray tube	Synchrotron bending Magnet
Density @13keV (photons/sec/mrad^2/0.1%)	10^{10} (Present Value: 10^8)	10^8 @8keV	10^{11}
Brilliance@13keV (photons/sec/mrad^2/mm^2/0.1%)	10^{14} (Present Value: 10^{12})	(The case of Micro-focus; 10^{12} @8keV)	10^{15}
Density @13keV (photons/sec/mrad^2/0.1%)	10^{10} (Present Value: 10^8)	10^8 @8keV	10^{11}
Brilliance@13keV (photons/sec/mrad^2/mm^2/0.1%)	10^{14} (Present Value: 10^{12})	(The case of Micro-focus; 10^{12} @8keV)	10^{15}

ported on a large lorry is already in production). The use of MIRRORCLE in scientific laboratory (with the required energy of radiation in range 7-13 keV) is very problematic because of immense difficulties with shielding of unwanted very hard and intensive radiation. The efficiency for experiments with monochromatic radiation might be increased by a special design with a "parametric radiation" generated in the monocrystalic target properly oriented against the accelerated electron beam. However, there is no functional prototype and the theoretical data are not enough promising with respect to the price and operational costs (Tab.1).

Standard synchrotron source (as the CESLAB) is presently the only reliable and safe source of radiation with fully guaranteed quality. It provides an immense flexibility and universality and can satisfy up to several hundreds of users. Because it belongs to the large scale facilities, the design should always contain at least 6-10 beamlines for research in different fields of science because than the costs of purchase and maintenance per one beamline drop to the level expected for local intensive home sources. Additional beamlines for newly rising scientific or technological fields can be added to a large synchrotron ring of CESLAB any time without extensive costs and without interrupting the operation.

The home source LYNCEAN will be a good choice for individual institutes satisfied with a single beamline and for special research where a presence of public is not appreciated or allowed. The home source MIRROCLE may be good choice for special industrial applications. As far as the acquisition and operating costs per one beam line, synchrotron is usually more economical solution, however it looses this advantage rapidly when the number of beamlines drops significantly under ~ 10.

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1. J. Hašek, Opportunities for protein crystallography at the Central European Synchrotron Laboratory, *Materials Structure,* **15** (2008) 19-24.

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APPLICATIONS OF X-RAY PHOTON CORRELATION SPECTROSCOPY IN SOFT MATTER

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X-Ray Photon Correlation Spectroscopy (XPCS) is a new technique that takes full advantage of the high intensity of partially coherent X-ray beams at third generation synchrotron sources. XPCS is based on the same principles as Dynamic Light Scattering (DLS) employing visible laser light. Thanks to the short wavelength of X-rays, XPCS can probe dynamics on small length scales and turbid samples may be investigated.

The interaction of a coherent beam with a disordered system produces a “speckle” scattering pattern. The speckle pattern reflects the exact, instantaneous distribution of the scatterers in the system. If the scatterers change their position with time the corresponding speckle pattern also changes and hence the intensity fluctuations of the speckles can provide a measure of underlying dynamics. The XPCS technique is used to study mesoscale dynamics of soft matter systems like polymers, gels, colloidal glasses, liquid crystals and supercooled liquids. The latter is of high scientific interest and the object of the study presented in the following.

The glass transition of supercooled liquids is an interesting phenomenon which continues to attract a lot of interest. Supercooling of a glass former leads to a dramatically slowing down of the molecular motion and finally a vitrification below the glass transition temperature Tg. The complex molecular processes that take place during such transition are still poorly understood. It is believed that thermally activated motion between different minima of the energy landscape gets important in the supercooled state. The transition to landscape dominated dynamics was found to happen at temperatures higher than the glass transition temperature Tg. In this work we studied the dynamical properties of two supercooled liquids propanediol and polypropylene glycol (PPG) with molecular weight 4000 (PPG-4000) by means of XPCS.

The dynamics of probe nanoparticles suspended in propanediol was studied above the glass transition temperatures i.e. in the supercooled state. At high temperatures (> 230 K) the particles undergo Brownian motion and the measured correlation functions are simple exponential decays. Upon cooling further and approaching the glass transition temperature (Tg = 170 K) the dynamics of the particles change. This can be readily seen as the correlation functions suddenly decay faster that expected from a simple exponential behavior. To analyze the data we applied the continuous time random walk model where the motion of a particle during the time interval t is described by N discrete steps. The low-temperatures data indicate that the particles exhibit hyper-diffusive, cooperative behavior. Our measurements show that the transition to this behavior occurs about 40K above Tg but the detailed connection with the glass transition remains an open question.

Another study was devoted to the investigation of surface capillary wave fluctuations on supercooled PPG-4000. All liquid surfaces undergo constant fluctuations due to the presence of thermally activated capillary waves. The dynamics of the waves is determined mainly by the surface tension and the viscosity which is a bulk characteristic of the liquid. We investigated the capillary wave dynamics of supercooled PPG-4000 by grazing-incidence XPCS. Our results show a deviation of the dispersion relations from classical theory. We present a novel viscoelastic model that can successfully describe the experimental data. It combines the well known Maxwell-Debye and Voigt-Kelvin viscoelastic models pointing out to the presence of a low-frequency elastic plateau. This solid-like behavior of the supercooled low molecular polymer liquid is intriguing and its possible origin is discussed.

1. G. Grübel, F. Zontone, *J. Alloys Comp*. **362**, 3, (2004).
2. C. Caronna, Y. Chushkin, A. Madsen and A. Cupane, *Phys. Rev. Lett.* **100**, 055702 (2008).
3. Y. Chushkin, C. Caronna, A. Madsen, *Europhys. Lett*. (submitted).