



NUCLEAR RESONANT SCATTERING BEAMLINE AT CESLAB

O. Schneeweiss¹, V. Procházka², M. Mašláň²

¹*Institute of Physics of Materials AS CR, Žitkova 22, 61662 Brno*

²*Faculty of Science, Palacký University, Svobody 26, 77146 Olomouc*

Keywords

nuclear resonant scattering, hyperfine spectroscopy, Mössbauer spectroscopy

Abstract

The proposal of Nuclear Resonance Scattering (NRS) beamline at Central European Synchrotron Laboratory (CESLAB) is presented. The NRS is an experimental method needed a source of high intensive monochromatic photon beam and covering large number of experimental setups and modification which allow one to study wide spectrum of physical phenomena like hyperfine interaction, diffusion, surface and bulk thermodynamical properties, phonon density of states in solid state, vibration modes of molecules, molecule conformations etc. Efficient photon source is an undulator at synchrotron storage ring. The detailed proposal of beamline layout with several technical aspect and parameters is reported. The beamline was designed with respect to usage of NRS by scientific/research community (in the central European region) in many brunches, which are mentioned.

1. Short technical summary

Energy range: 5-50keV (the most often 14.4keV)

Methods applied: forward scattering (NFS), nuclear Bragg scattering (NBS), nuclear inelastic scattering (NIS), quasi-elastic and inelastic scattering with nuclear resonance analysis, synchrotron Moessbauer spectroscopy

Fields of research: hyperfine field (magnetic oxides, iron oxides, ...), phonon density of state, vibration modes of big molecule, molecule conformation, high speed processes, in-situ reaction study, reaction dynamic studies, diffusion in solid state, electron configuration study (high spin and low spin states of magnetic ions).

2. Introduction

The proposed beamline will offer application in the area of NRS and synchrotron Moessbauer spectroscopy. Theoretical principles of NRS were worked out just after the discovery of Moessbauer effect in the end of 50ties of 20th century [1]. Nuclear resonance scattering of synchrotron radiation includes a wide spectrum of different experimental techniques, such as nuclear forward scattering (NFS), nuclear Bragg scattering (NBS), nuclear inelastic scattering (NIS), quasi-elastic and inelastic scattering with nuclear resonance analysis. The synchrotron radiation was found to be very efficient source of photons for nuclear resonance scattering methods because of the high intensity and brilliancy and the pulse character of the radiation. The usage of synchrotron radiation as a source for Moessbauer spectroscopy (instead of the traditional radiation source) decrease rapidly the time of measurement in such scale that

the synchrotron radiation Moessbauer spectroscopy is suitable method for high speed process investigation like in-situ chemical reaction studies and diffusion studies. Possibility of the incident beam focusing allow one to study very small samples (around 100 x 100 mm). Hyperfine interactions are studied by NFS in forward or Bragg geometry. Broad range energy of incident beam with connection of sub meV resolution monochromator gives opportunity for direct measurement of phonon density of states. The NRS is an unique tool in many research brunches and the development of new applications and experimental setups still not finished.

3. Scientific case of the proposed beamline

Main NRS application field is for investigation of samples under extreme conditions like high pressure, fast temperature changes to low/ high temperatures, in high external magnetic field, fast processes, etc. The research is focused on two scientific area – hyperfine spectroscopy and structural dynamics.

3.1. Hyperfine spectroscopy

The field of hyperfine spectroscopy include static and dynamic magnetic and electric properties such as electron density, fields and fields gradient of magnetic or electric origin. Surface, interface and multilayer can be investigate using nuclear resonance reflectometry. Electric and magnetic structure of single crystal samples can be determined by using NBS.

Consider the recoilless process the nucleus can be excited from the ground state to the excited state by absorption of the phonon of characteristic energy. The nuclei deexcited accompanied by emission of photon with delay given by excited state lifetime. Lets consider an ensemble of nuclei incident by short photon pulse of given energy distribution nuclei are excited and then deexcite coherently, so the photons interfere and we can observe a beating in the time decay of photon intensity. In case the nuclei levels are split by hyperfine interactions the beating in the time decay carries an information about hyperfine interactions. The biggest advantage of time resolvable Moessbauer spectroscopy is in eliminating the prompt beam and the electron scattered beam from the nuclear scattered beam in time. The prompt beam and the electron scattered beam occur with delay up to several ps contrary to the nuclear scattered beam where the time decay usually is longer than hundreds of ns (according material) [2]. This method use with advance the pulse character of the synchrotron radiation, where the radiation occur in picoseconds pulses separated by hundred of ns [3]. The whole spectrum squeeze in the range of 2 ueV and the monochromated beam bandwidth usually is in order of meV thus the whole spectrum is excited at the same time



and the monochromator is tuned to the energy of nuclear transition.

Possible fields of applications are in materials science, physics, chemistry, geosciences and life sciences. Nuclear resonance scattering spectroscopies under extreme conditions are key methods to provide sound velocities and elasticity on iron, iron alloys and iron oxides [4, 5] or to study valence and high-spin to low-spin transition [6, 7].

3.2. Structural dynamics

The second area – structural dynamics of “non-resonant” samples are accessible for density of phonon states via inelastic scattering with nuclear resonance energy analysis and for diffusion using time interferometry. More details on physics and experimental techniques of nuclear resonant scattering can be found on the web page of the beamline ID 18 ID22N at ESRF [8] or in the review articles [9, 10].

The Nuclear Inelastic Scattering (nuclear resonant vibrational spectroscopy) does not exploit the recoilless absorption, contrary the phonon participation in the absorption and emission process is studied. When the photon energy of incident beam is not exact equal to the nucleus transition energy the phonon can participate in the process and the photon can be absorbed. The absorption and emission of photon is proportional to the phonon density of states thus it can be probed by changing of the photon incident beam energy. The whole spectrum overlay in the range up to 200 meV. The experiment is proceeded as an energy scan and the energy bandwidth of incident beam is crucial for measurement resolution [11].

Very interesting result on application the isotope-specific Fe^{57} nuclear resonant vibrational spectroscopy was published recently [12] where the authors report on elastic and thermodynamic properties of ultrathin Fe films on W(110). With decreasing thickness they observe a significant increase of the mean atomic displacement that goes along with an enhancement that goes along with an enhancement vibration modes at low energies as compared to bulk. The analysis reveals that these deviations result from vibrations of the single atomic layers at the two boundaries of the film, while the atoms inside the films vibrate almost bulklike.

3.3. Moessbauer isotopes

The NRS is method suitable for investigation by using large number of different isotopes. with nuclear transitions energies from 5keV to 50keV. Because of very strong requirement on the monochromation of the beam, it is technically extremely hard to achieve good properties of the beam in such a broad range therefore it is assumed building an experimental setup for Fe^{57} isotope measurement in the first period. The Fe^{57} (nuclear transition Fe^{57} energy is 14.4keV) is the most important Mössbauer isotope because of the wide presence of iron in nature (hemoglobin, clays, ferrites ...). Indeed, all equipment is designed to be easily extended for other isotopes.

4. Beamline layout

The proposed beamline has to be build up of several parts placed in separated hutches. The incident beam for experiment is prepared in several steps to have highly specific parameters required for chosen experiment in three hutches and front end. The first two hutches are designated for monochromators (head load and high resolution monochromator), the third hutch is designated for measurement devices (sample holder, goniometer, detectors, etc.).

The first part of the beamline is a front end where radiation source device is placed. Conception of the proposed NRS beamline is based on undulator as the X-ray radiation source [3]. In case of NRS experiments, where the intensity of the beam has crucial influence on the experimental proceeding, results and usage potential, the undulator could satisfied the requirements on the radiation intensity. The front end contain also Compound Refractive Lenses (CRT) which are used to focused the beam of undulator origin before it enter the monochromating process.

4.1. Beam monochromating

Monochromating of the beam is an crucial part of the experiment. Phenomena studied by NRS has energy band of spectra from 1 eV (NFS) to 200 meV (NIS). Good energy resolution is important factor. Narrow band of the beam is needed to minimize mainly the prompt radiation which can saturate or burn down the detector and for NIS it is crucial parameter for the resolution of the measurement.

The monochromating is planed to be hold in two steps. The first is a rough monochromating by so called heat load monochromator, which selects from the whole energy range (usually 5-25keV) narrow band of several eV around given energy. The heat load monochromator is planed as Kohzu Diamond (111) for energy range 5-25keV, resolution better than 2×10^{-4} , flux $> 1 \times 10^{12}$ photons/sec at 14.4 keV and unfocused beam size approx. 2×0.2 mm [13].

The monochromator is very vibration sensitive and even very small vibration could cause the lost of beam at sample position so any vibration have to be eliminated by antivibration devices. The heat load monochromator must load huge energy of unwanted radiation. Usually it is power of kW. That monochromator is situated in separate hutch together with an X-ray optics element such as a focusing crystals and collimators which spatially limited the beam before it enter the second step of monochromating process.

Different experiments of NRS such as NFS and NIS requires different beam properties. Very high intensity of the beam for NFS is required by contrast the band width should be wider contrary to the beam line for NIS where the bandwidth has to be minimized to the prejudice of the intensity. One monochromator cannot satisfy such a different requirements therefore the beamline has to be equipped by set of monochromators suitable for all planed experimental setup. In the first period of building we planed two monochromators (for NIS and NFS). The high resolution monochromator is based on the multiple sequence reflection in both cases. The difference is in the angle of the scat-

tering (high / low order). The monochromator has to be able to minimize the bandwidth to 1 meV at 14.4keV in case of NFS and to sub meV bandwidth in case of NIS. Huge attention has to be focused on the thermal stability, the temperature stability in the hutch with monochromator have to be better than 0.01 K at room temperature otherwise the beam energy can be tuned several meV from the needed value [13]. Therefore, the high resolution monochromator hutch has to be designed together with air-conditioner and precise temperature control system.

4.2. Experimental hutch

Since the highly monochromated beam leave the hutches of monochromator it enter the experimental hutch equipped by x-ray optics (lens, slits, colimators, etc.). The experimental hutch is planned to offer wide choose of experimental setups (NFS, NBS,...), therefore it will be equipped by several goniometers allowing one to rotate the sample and detector in many direction and detect in large space angle. To make the beamline excellent experimental place for users of different branches the experimental hutch is planned to be equipped by devices for experiments holding in different conditions. For temperatures from 2K to 1000K the cryostat and oven is designed. The oven allow in-situ study of chemical reactions and thermal processes. Superconducting magnet offers measurements in external magnetic field and the vacuum and ultrahigh vacuum system is planned also.

For nuclear resonance scattering experiments detectors with ns to sub ns time resolution, high dynamic range and fast recovery time are mandatory. The detector must survive this intense prompt flash and it has to be able to count few nanoseconds later a single photon event of the delayed nuclear radiation. These conditions nowadays completed by avalanche photo diode (APD) detectors [14].

The APD detector is connected with timing electronic which is responsible for the correct data collection and storing. In such measurement like NFS (in time domain) the synchronization of whole installation with the incident beam is crucial. Precise synchronization signal can be derived from the incident beam by detection of unused beam outgoing a monochromator (prompt beam), the second possibility is a "bunch clock" signal which is derived from the storage ring and lead to the experimental hutch. Both possibilities are sufficient.

4.3. Technical and operating background

All experiments are operated from the operating room where also all controlling devices are situated. The experiments are controlled and evaluated. For the evaluation exist several packages of software like MOTIF [15], EFFINO [16], CONUSS and PHOENIX [17], also development of new software tools is planned for the results evaluation. For

the beamline necessary is also a preparation room where the sample is prepared for experiment.

5. User community

The Czech user community utilizing synchrotron radiation for NRS are:

1. Institute of Physics of Materials AS CR, Brno
2. Centre for Nanomaterial Research, Palacký University, Olomouc
3. Department of Low-Temperature Physics, Charles University

Central European community: Mössbauer communities in Slovakia, Hungary, Poland, Ukraine, Romania, Bulgaria.

6. Conclusion

The NRS beamline is design as a fully equipped experimental research station of synchrotron Moessbauer spectroscopy and for NRS experiment in the first period for Fe⁵⁷ experiments with possible easy extension for other Moessbauer isotopes.

7. References

1. Gerda, E., et al., *Hyperfine Interactions* 123/124 (1999), 3.
2. Roehlsberger, R., *Nuclear Condensed Matter Physics with Synchrotron Radiation*, Springer (2004).
3. Smirnov, G.V., *Hyperfine interactions* 123/124 (1999), 13-30.
4. Mao, H.K., et al., *Science* 292 (2001), 914.
5. Lin, J.F., *Science* 308 (2005), 1892.
6. Jackson, J.M. et al, *Am. Mineral.* 90. (2005), 199.
7. Lin, J.F., *Phys. Rev. B* 73 (2006), 113107.
8. <http://www.esrf.fr/UsersAndScience/Experiments/HRRS/NRG/>
9. Leupold, O., *Hyperfine Interactions* 144/145 (2002), 21.
10. Sturhahn, W., *J. Phys. Cond. Matter* 16 (2004), S497.
11. Chumakov, A.I., et al., *Hyperfine interactions* 123/124 (1999), 781-808.
12. Stankov, S., et al., *Phys. Rev. Lett.* 99 (2007), 185501.
13. Toellner, T.S., *Hyperfine interactions* 125 (2000), 3-28.
14. Baron, Alfred Q.R., *Hyperfine interactions* 125 (2000), 29-42.
15. Shvyd'ko, Yuri V., *Hyperfine interactions* 125 (2000), 197-204.
16. Spiering, H., et al., *Hyperfine interactions* 125 (2000), 149-172.
17. Sturhahn, W., *Hyperfine interactions* 125 (2000), 149-172.