Nuclear Resonance Scattering:
Again a new era at the horizon.

Rudolf Rüffer

ESRF-The European Synchrotron, Grenoble, France

rueffer@esrf.fr

With synchrotron radiation, Mössbauer spectroscopy got not only new impetus but also opened up new fields and comprises nowadays as well quasi- and inelastic spectroscopies, Synchrotron Radiation based perturbed Angular Correlation (SRPAC) and other facets utilizing the nuclear resonances. We have seen exciting developments comparable to those in the early days of the Mössbauer age. This all was not only possible with the bright ideas in science popping up everywhere but as well with the bright ideas and heavy work by the users and beamline staff developing new techniques and instruments. Now with the ESRF-EBS we have again reached a landmark where exciting science and techniques are at the horizon. However, we will not stay alone, the other big synchrotron radiation facilities will soon follow and new ones are expected to come, both with the potential to surpass the existing one. Again, we need the very fruitful collaboration of users and beamline staff to get the best out of the development meaning to achieve new exciting results we may even not think about today.

During the workshop, we will hear about a wealth of new ideas on techniques and instrumentation. Looking around in the general synchrotron radiation world phrases such as "nano", "imaging", "coherence", and "holistic approach" are *en vogue.* What will be applicable to NRS? Where shall we go? We will highlight some aspects and developments, which might be crucial for the future. May be there are as well some dreams.

News of EBS machine, including timing modes

Jean-Luc Revol

ESRF-The European Synchrotron, Grenoble, France

revoljl@esrf.fr

The European Synchrotron Radiation Facility - Extremely Brilliant Source (EBS) is a facility upgrade bringing its scientific users a first-of-a-kind, low-emittance, high-energy synchrotron light source and new, cutting-edge beamlines.

On December 2018, after 30 years of operation, the beam stopped for a 12 month-month shutdown to dismantle the storage ring and install of a new and revolutionary X-ray source. On December 2019, the first beam was stored and accumulated in the storage ring, allowing starting vacuum conditioning and tuning. Beam was delivered to the beamlines on March 2020 for commissioning. On 25 August 2020, user program restarted with beam parameters very close to nominal values. From July 2020, delivery was mostly done in 7/8+1filling pattern, to allow the nuclear resonance beamline ID18 to work

With the new storage ring concept, allowing for an increase in brilliance and coherence by a factor of, the time structure filling modes are preserved. Nevertheless, due to difficulties with ceramic vacuum chambers, the delivery in 16 bunch time and associated filling patterns is today limited in beam current. New beam modes were developed in order to allow beam delivery for ID18 whereas not penalizing the other beamlines. This technical issue should be solved by the end of 2021. Reliability and stability of the beam is already very good.

EBS features, as seen from beamline

Dimitrios Bessas

ESRF-The European Synchrotron, Grenoble, France

bessas@esrf.fr

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On 10 December, 2018, the ESRF stopped the beam after almost 30 years of continuous operation of the original storage ring. An almost 20-month shutdown began with the dismantling of the storage ring and the installation of a new and revolutionary X-ray source. The nuclear resonance beamline ID18 took profit of the extended ESRF shutdown and refurbished all existing mechanics, electronics, optics, and software as well as it took actions to test new setups for the future new nuclear resonance beamline at ESRF.

On 30 January 2020, after reaching stable operation conditions of the EBS storage ring with 100 mA injection current, 65% injection efficiency and steady vacuum conditioning, the nuclear resonance beamline ID18 at ESRF opened its front-end with 5 mA of stored electron beam current and the commissioning phase of the beamline started.

During the commissioning phase the beamline recovered some *old* features, for example the optimum performance of the existing high-heat-load monochromator, and a high energy resolution obtained using the existing high-resolution optics, 0.5 meV at 14.412 keV, for the nuclear resonance energy of 57-Fe. New features were achieved with the existing focusing optics relevant to a small beamsize, [Hx V] 2 µm x 4 µm (for NIS and NFS) and 2 µm x 8 µm (for SMS), and may now routinely be provided in the user operation of the beamline.

In strong collaboration with the Accelerator and Source Division of ESRF the nuclear resonance beamline ID18 voluntarily contributed in selecting the optimum machine emittance and the storage ring cleaning in order to obtain a usable bunch purity for nuclear resonance scattering applications. Soon after the first tests a routine bunch purity between 10^{-10} and 10^{-11} in top-up mode (*i.e*, refill every 1 h) with the least beam perturbation during refill was achieved.

After the beamline commissioning phase, the nuclear resonance beamline ID18 was open for user operation. Despite the extended shutdown a very high demand for beamtime applications was received. A smooth full semester of remote user experiments (because of the global sanitary crisis) took place in the Fall of 2020.

With the revolutionary new storage ring concept, allowing for an increase in brilliance and coherence by a factor of 100 compared to present-day light sources, ESRF–EBS represents a new generation of synchrotron and an extraordinary new tool for scientists to study the heart of matter using nuclear resonance scattering.

Upgrade of the Nuclear Resonance beamline at ESRF

Aleksandr Chumakov

ESRF-The European Synchrotron, Grenoble, France

chumakov@esrf.fr

In 2024 the Nuclear Resonance beamline at the ESRF will be upgraded and it will change location from the ID18 to the ID14 straight section. The aims of the upgrade are to pursue *(i)* spectroscopies with extreme spatial resolution and *(ii)* studies of atomic dynamics with extreme energy resolution, and eventually *(iii)* allow one to use all advantages of the Extremely Brilliant Source of the ESRF.

For studies with extreme spatial (~150 nm) and extreme energy (~50 μeV) resolution, the beamline will be equipped with two new instruments, Nanoscope and Spectrograph, respectively. In both cases, the improvement in resolution will be achieved without essential losses of flux; i.e., keeping nearly the same count rate. Details of both instruments are discussed in two separate contributions to this workshop.

In order to assure stable operation with extreme spatial and energy resolution, the second optics hutch (containing optics of high-resolution monochromators, synchrotron Mössbauer source, and Spectrograph) and a new fourth experimental hutch (hosting Nanoscope) will be equipped with thermal isolation, and with high-quality heating, ventilation, and air conditioning (HVAC) system providing temperature stability of about 0.1°C.

The radiation source is expected to be upgraded to cryogenically-cooled permanent magnet undulators (CPMU), providing an increase of intensity by a factor of 1.4 for 14.4 keV (^{57}Fe), by a factor of 3-7 for 20-40 keV (^{151}Eu, ^{149}Sm, ^{119}Sn, ^{161}Dy, ^{125}Te, ^{121}Sb, ^{129}Xe), and by a factor of 15-20 for 60-90 keV (^{61}Ni, ^{99}Ru).

The high-heat-load monochromator (HHLM) will be upgraded by switching from vertical to horizontal scattering plane. This will allow for preserving best quality of the beam after HHLM in vertical plane for downstream high-resolution optics (working in the vertical plane of scattering). Simulations [1] show, that under these conditions the angular deteriorations of the beam after the HHLM due to heat-load effects are smaller than the angular size of the beam source (as seen from the HHLM position), i.e., that this instrument is free of the heat-load effects in the vertical plane. Similar to the improvement of spatial and energy resolutions, this will be achieved without noticeable (~3% percent only) decrease of intensity.

The Nanoscope and Spectrograph instruments are expected to be delivered to users by end of 2021, still at ID18, and moved to ID14 in 2023. ID18 is planned to be operational till second half of 2023, and user service at ID14 is expected to start from beginning of 2024.

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Spectrograph facility: towards 50-100 µeV energy resolution

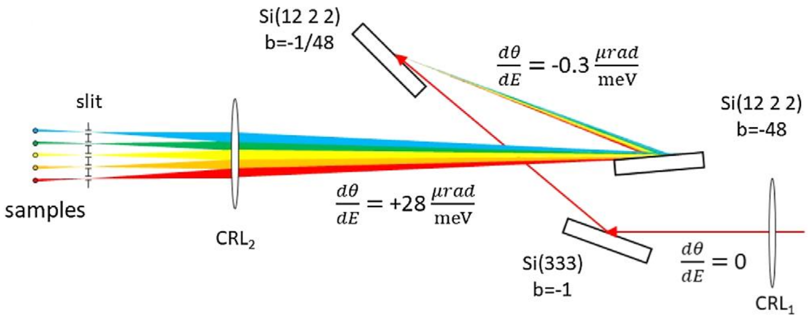
Dimitrios Bessas

ESRF-The European Synchrotron, Grenoble, France

bessas@esrf.fr

A high degree of monochromatization combined with a high incident to the sample flux are the basic requirements in scattering techniques for resolving collective excitations, such as phonons or magnons, in condensed matter. In the traditional approach a high degree of monochromatization is achieved by filtering the vast majority of wavelengths but one and as a result it works at the expense of high incident to the sample flux.

Recently, at the nuclear resonance beamline ID18 at ESRF we have developed a spectrographic approach [1] that is based on the Newton's prism principle, *i.e.*, the X-ray beam is dispersed in space in several monochromatic sub-beams, and we obtained an enhanced energy resolution of approx. 0.1 meV without sacrificing intensity, *i.e.*, all sub-beams (wavelengths) may be recorded simultaneously utilizing a position-sensitive detector. A proof of principle schematic of the spectrograph facility is shown in Fig. 1.



There are two key factors in order to achieve the optimum performance of such a spectrograph facility: (1) the quality of the crystals used for dispersing the X-ray beam that may deteriorate the obtained energy resolution, (2) the existence of a position sensitive detector with timing possibilities. In this talk, the principles of the spectrograph facility at the nuclear resonance beamline ID18 at ESRF will be discussed and hints on the quality of the crystals and a position sensitive detector with moderate timing capabilities will be given.

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Future Nanoscope facility: towards sub-micron spatial resolution

Aleksandr Chumakov

ESRF-The European Synchrotron, Grenoble, France

chumakov@esrf.fr

The Extremely Brilliant Source (EBS) of the new ESRF machine and the maintenance of beamline optics allowed the Nuclear Resonance beamline ID18 to improve the beam size from ~15 × 15 μm^2 [*] available in 2018 to ~2 × 8 μm^2 in 2020. The new Nanoscope facility will continue this development, providing sub-micron spatial resolution requested by high-pressure experiments in the TPa range, studies of inclusions in diamonds, investigations of cloudy zone of meteorites, studies of nano-objects, *etc*.

The Nanoscope includes the nano-platform and the short-focal Kirkpatrick-Baez mirror (KBM). Commissioning of the nano-platform with the accuracy of linear positioning of ~10 nm has been completed. The KBM is expected to come in autumn 2021. The Nanoscope will be delivered to users by end of 2021.

Thanks to the new EBS machine and the operational nano-platform, we determined typical slope errors of high-resolution optics in measurements of the size of the beam focussed by high-quality parabolic compound refractive lenses (CRLs). Previously, these data were not available because the spot size was dominated by geometrical demagnification and/or slope errors of existing KBMs.

For 14.4 keV x rays (^{57}Fe), the beam size of 580 × 700 nm^2 has been obtained with CRLs demagnification of ~1/75.8. Extrapolation of the results to a future short-focal KBM with horizontal and vertical demagnifications of 1/309 and 1/567, respectively, (including contributions of the KBM slope errors) gives the following estimations of the beam size: 170 × 110 nm^2 for ideal optics, 220 × 420 nm^2 for 0.5 meV high-resolution monochromator, and 620 × 660 nm^2 for synchrotron Mössbauer source.

The Nanoscope will provide sub-micron resolution nearly without losses of intensity, as the horizontally- and vertically-focusing mirrors will intercept ~180% and ~150% of FWHMs of the corresponding beam size. The sample space will be about 70 mm.

In order to assure stable operation with extreme spatial resolution, the Nanoscope will be located in a hutch with thermal isolation and temperature stability of about 0.1°C.

In future, the Nanoscope facility is expected to be upgraded by a KBM for 21.541 - 25.614 keV (^{151}Eu, ^{149}Sm, ^{119}Sn, ^{161}Dy) and another KBM for 67.4 keV radiation (^{61}Ni) with the expected beam sizes of 0.7 × 1.2 μm^2 and 0.87 × 0.55 μm^2, respectively. This will require additional budget and resources.

* All data are given as full widths at half maximum (FWHMs), for horizontal × vertical sizes.

Nuclear Resonance Measurements in the Laser Heated Diamond Anvil Cell

Georgios Aprilis
ESRF–The European Synchrotron, CS 40220, CEDEX 9, 38043 Grenoble, France

Laser heating inside the diamond anvil has already been used in high-pressure science for more than five decades, to study properties of materials *in situ*, simultaneously at high temperature and high pressure. The laser heated diamond anvil cell (LHDAC) technique has found numerous applications in mineral physics and high-pressure chemistry, physics, Earth and material sciences.
At synchrotron light source facilities, many beamlines working with samples at high pressure have coupled LHDAC with different techniques, such as X-ray diffraction, Non-resonant Inelastic X-ray Scattering (NIXS) and X-ray Absorption Near Edge Structure spectroscopy (XANES) showing the versatile nature and popularity of the technique. More specifically for nuclear resonant scattering beamlines, the LHDAC technique has found use Nuclear Inelastic Scattering (NIS)[1,2], Nuclear Forward Scattering (NFS)[3] and Synchrotron Mössbauer Source (SMS)[4,5] experiments. Over the years, the laser heating system at the Nuclear Resonance Beamline (ID18) of the ESRF has been employed for experiments at high pressures at temperatures producing important results for geoscience[6,7].
In addition to continuous-wave heating, pulsed laser heating has the advantage of achieving significantly higher temperatures due to the concentration of high laser power in a short impulse. The repetitive heating and cooling of the sample makes time an extra variable in addition to pressure and temperature, which is not possible with continuous-wave laser heating, allowing the possibility of time-resolved measurements.
Recently, a scheme was developed at ID18, allowing for fully time-resolved measurements of either SMS or NIS of the sample while pulse laser heating at high pressures. The proof-of-principle measurements reveal the modulated displacement of the sample inside the pressure chamber while pulsed heating, creating the possibility for a new field of research.

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New ideas in Synchrotron-based Perturbed Angular Correlation

Dimitrios Bessas

ESRF-The European Synchrotron, Grenoble, France

bessas@esrf.fr

The relatively unconventional nuclear resonance scattering technique of Synchrotron Radiation Perturbed Angular Correlation Spectroscopy (SRPAC) has great potential and was indeed used to study the properties of Mössbauer active nuclei, *e.g.*, iron-57 [1], tin-119 [2], nickel-61 [3]. SRPAC may be also used with less-known Mössbauer active nuclei [4] both in the solid or liquid phase. This technique probes isolated nuclei and is independent of recoil-free fraction.

The synchrotron radiation is used to incoherently excite the nuclei, *i.e.*, the excited transitions originate from a single ground state and terminate in a metastable state that decays towards the ground state by different paths determined by the hyperfine interactions between the nuclei and their electronic environment.

Although SRPAC and Nuclear Forward Scattering (NFS) are carried out using a very similar instrumentation, they are two essentially different experimental techniques. In short, NFS is an elastic coherent method that probes collective effects over a nuclear ensemble and thus is extremely sensitive to sample thickness, whereas SRPAC is an incoherent method and thus is in principle insensitive to sample thickness. Notably, NFS takes place by definition only in the forward direction, whereas SRPAC scattering occurs in the full solid angle.

An interesting feature in SRPAC is the so-called magic angle. The synchrotron radiation is a highly linearly polarized beam. In these conditions there is an angle, *i.e.*, 35.3 deg relative to the horizontal plane, at which the potential hyperfine interactions do not contribute to the measured spectra. Thus, the spectra measured at the magic angle may be used to quantify non-trivial multiple scattering phenomena or potential radiation trapping effects.

In this talk SRPAC will be briefly introduced and the concept of magic angle will be discussed in the context of revealing the hidden hyperfine interactions in epsilon-Fe.

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New Ideas in Time-domain Interferometry

F. Caporaletti[1], S. Capaccioli[2], S. Valenti[3], A. I. Chumakov[4], M. Mikolasek[4] & G. Monaco[5]

[1]Van der Waals-Zeeman Institute, Institute of Physics/Van 't Hoff Institute for Molecular Sciences, University of Amsterdam, Amsterdam, The Netherlands

[2]Dipartimento di Fisica, University of Pisa, Pisa, Italy

[3] Department of Physics, Universitat Politècnica de Catalunya, Barcelona, Spain.

[4] ESRF-The European Synchrotron, Grenoble, France

[5] Dipartimento di Fisica ed Astronomia, Università di Padova, Padova, Italy

Nuclear γ-resonance time-domain interferometry (TDI) provides access to atomic and molecular motions at the Ångstrom length-scale and in the ns-us time-window [1,2,3]. It is therefore the perfect tool to take a microscopic look at relaxation processes in supercooled liquids in that time window, and in the recent years has significantly contributed to the understanding of the glass-transition and of the associated atomic motions [4,5,6,7].

In this talk the state-of-the-art of TDI will be discussed [2,3,8] along with the new challenges and future perspectives, with a focus on the study of the glass-transition.

In particular the new opportunities open by the combination of multi-line TDI experiment [2,3,8] and dielectric spectroscopy measurements [6,7] will be explored. Such approach, being able to access both re-orientational and translational motions and to also determine the fraction of molecules involved in a relaxation process [7], has the potentiality to open a new window on the unique space-temporal properties of supercooled liquids and glasses.

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New ideas in Synchrotron Mössbauer Source: iron borate crystals, collimating lenses

Aleksandr Chumakov

ESRF-The European Synchrotron, Grenoble, France

chumakov@esrf.fr

The Synchrotron Mössbauer Source (SMS) at the Nuclear Resonance beamline ID18 at the ESRF operates with the (111) reflection of iron borate $^{57}FeBO_3$ crystal and the double-crystal Si(422)-Si(531) angular deflector (Fig.1). The advantage of this scheme is the practically horizontal beam from the SMS, adjusted in-line with the direct beam after the high-heat-load monochromator.

The compound refractive lens (CRL) is used in order to "squeeze" an incident radiation with the vertical divergence of ~12 μrad into the angular acceptance of the deflector (~3 μrad). In past, we used CRLs made of the O-30-H beryllium grade. With this CRL, the spectra were contaminated by contributions of Fe impurities in Be by about 2%. Therefore, in studies of non-enriched and thin samples, many users preferred to remove the CRL from the beam, even at the expense of about 40% loss of intensity.

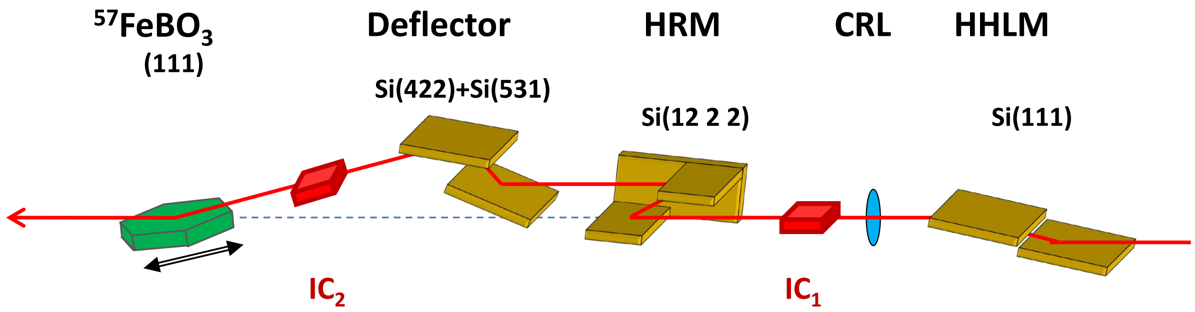

Figure 1. Synchrotron Mössbauer Source at ESRF. HHLM – high-heat-load monochromator with two Si(111) crystals; CRL – compound refractive lenses; HRM – high-resolution monochromator with two Si(12 2 2) reflections of a single channel-cut crystal, Deflector – double-crystal Si(422)-Si(531) angular deflector.

In 2020, ID18 tested new collimating parabolic CRLs (RXOPTICS) made from IF-5 and I70-H beryllium with ultra-low iron content. The contamination of spectra by contribution from iron impurities in Be decreased from 2% to 0.6%, where it is already dominated by a contribution from Be window (0.5%). Furthermore, high quality of CRLs allowed us to increase the flux by ~20% and, in addition, to gain further ~40% by slight detuning of the undulators from "best brilliance" to "best flux" magnetic gaps. Altogether, the obtained gain of intensity is ~1.7 for enriched or thick samples, and ~3 for non-enriched thin sample.

We also will discuss options to obtain enriched iron borate $^{57}FeBO_3$ crystal with better quality.